

Atom probe tomography

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Abstract

Atom probe tomography (APT) provides three-dimensional compositional mapping with sub-nanometre resolution. The sensitivity of APT is in the range of part-per-million for all elements, including light elements such hydrogen, carbon, or lithium, enabling unique insights into the composition of performance-enhancing or lifetime-limiting microstructural features and making APT ideally suited to complement electron-based or X-Ray-based microscopies and spectroscopies. Here, we provide an introductory overview of APT ranging from its inception as an evolution of field-ion microscopy to the most recent developments in specimen preparation, including for nanomaterials. We touch on data reconstruction, analysis, and various applications including in the geosciences and the burgeoning biological sciences. We review the underpinnings of APT performance and discuss both strengths and limitations of APT, including how the community can improve on current shortcomings. Finally, we look forward to true atomic-scale tomography with the ability to measure the isotopic identity and spatial coordinates of every atom in an ever wider range of materials through new specimen preparation routes, novel laser pulsing and detector technologies, and full interoperability with complementary microscopy techniques.

[H1] Introduction

APT provides three-dimensional compositional mapping of materials with sub-nanometre spatial resolution¹, and, in principle, no lower or upper limits of elemental mass². The mass

42 resolution is generally sufficient to distinguish each isotope of each of the detected
43 elements. This information is used to infer how the distribution of certain elements affects
44 the properties of a material, to guide the design of new materials, or better predict when a
45 material will fail over time in service. It can also reveal the material's history or that of the
46 geological region where it was found. Typical bulk materials have complex, hierarchical
47 structures on multiple scales, as depicted in **Error! Reference source not found.**a. This
48 applies to human-made and natural solid materials – including engineering alloys,
49 semiconductor devices and minerals. In crystalline solids, atoms are organised on a lattice
50 that can be populated by **solute** [G] substituting for solvent atoms on the lattice or located
51 in between lattice atoms, i.e. interstitially. The crystal contains atomic and **microstructural**
52 **imperfections** [G] including lattice defects such as **vacancies** [G], **dislocations** [G], **stacking**
53 **faults** [G] and **twins** [G], and **grain boundaries** [G], as well as **secondary phases and phase**
54 **boundaries** [G], or voids, for example. Multiple stable and metastable phases may also be
55 present in the material. In addition, a material has an interface with its operating
56 environment, where a range of interactions can modify **composition** [G] and microstructure
57 resulting in, for example, cracks or oxidation. Other materials can be amorphous with
58 different defects. Some devices are fabricated with well-defined structures, such as thin
59 films with stacks of layers of an expected composition and numerous interfaces.

60 APT stems from field-ion microscopy (FIM)^{3,4}, and before it, from field-electron emission
61 microscopy (FEEM)⁵. The elegance of the FIM and FEEM techniques lies in their simplicity: a
62 high voltage applied to a needle-shaped specimen generates an intense electrostatic field –
63 the same effect underpinning the lightning-rod effect⁶. The needle is positioned in front of
64 an ion detector, which can be a phosphorescent screen or a particle detector. A negative
65 electrostatic field can cause the emission of electrons. Reverting the **polarity** [G] enables
66 two distinct phenomena, **field ionisation** [G], whereby atoms near the specimen's surface
67 can be ionised, and **field evaporation** [G], whereby atoms that constitute the surface of the
68 specimen can get ionised and desorbed. The specimen itself acts as a **projection optic** [G],
69 with no additional lenses. The charged particles are projected nearly radially during the
70 early stages of the flight, and their trajectory is defined only by electrostatics⁷. The tip-
71 shaped specimen has an end radius typically below 100 nm, which makes the projection
72 highly divergent and provides a magnification in the range of 10⁶. Interatomic distances,
73 typically 10⁻¹⁰ m, hence become 10⁻⁴ m, which is discernible by the human eye. FIM provided
74 the first direct images of surface atoms in the 1950s⁸. The atom probe combined this
75 imaging with a **time-of-flight mass spectrometer** [G]² to provide analytical capabilities. The
76 name 'atom probe' was seemingly coined as a counterpart to another materials analysis
77 technique, the electron-probe microanalyser, which uses X-Rays to provide micron-scale
78 compositional mapping. However, 'atom probe' is misleading; the ions that allow us to
79 probe the material are the primary beam produced by and from the specimen, whereas
80 other techniques typically have an external primary illuminating source, for instance light or
81 electrons in the most common microscopy techniques. Early atom probes allowed for depth
82 profiling⁹. Step-changes in the design^{10,11} led to 'three-dimensional' or 'tomographic' atom
83 probe, with the implementation of a position-sensitive detector^{12,13}. Thorough historical
84 perspectives are available on the development of the technique^{14,15}.

85 Not all microstructural features can be analysed by APT. Primarily a compositional mapping
86 tool, crystallographic lattice features such as vacancies and structural defects such as
87 dislocations, faults, and boundaries cannot be readily imaged. However, chemical

88 segregation leading to compositional heterogeneity at these structural defects often reveals
89 their presence^{16–20}. The strengths of APT complement other microscopy and microanalysis
90 techniques. Among a wide arsenal that allows for nanoscale imaging and analysis, **Error!**
91 **Reference source not found.** maps some that are commonly used with APT, along with the
92 typical imaged/analysed feature size and compositional sensitivity. In Figure 1b, between
93 scanning and scanning transmission electron microscopy (SEM and (S)TEM) with energy-
94 dispersive X-ray and electron-energy loss spectroscopy (EDS and EELS), and secondary-ion
95 mass spectrometry (SIMS), APT occupies a unique space in terms of combining a high
96 sensitivity in the range of 10s of parts-per-million and feature size in the range of below 1
97 nm. APT is also inherently three-dimensional which is not the case for the other techniques
98 in Figure 1b.

99 The higher throughput and wider applicability in APT instrument design, as well as
100 improvements in specimen preparation and transfer techniques, has led to an international
101 expansion of APT, with a hundred equipped groups in 2020, at shared facilities across the
102 world²¹ and at internationally-leading materials companies. In this Primer, we cover the
103 fundamentals of the technique, specimen preparation and data processing, show examples
104 of applications, and discuss reproducibility issues and the intrinsic limitations of the
105 technique. We finish by discussing prospective new technique developments and the
106 research frontier of the burgeoning cryo-APT for the fields of soft-matter and liquids.

107 [H1] Experimentation

108 This section describes the general aspects of atom probe experimentation. It covers the
109 setup contained in the ultra-high vacuum chamber, some of the practical aspects of the
110 fundamental scientific principles enabling the technique, and the preparation of specimens
111 suitable for atom probe analysis.

112 [H2] Atom probe design

113 There are a number of common traits in the design of modern atom probes (**Error!**
114 **Reference source not found.**a). Here, we describe some generalities as review articles and
115 textbooks are available for more details^{14,15,22,23}. The analysis takes place in an ultra-high
116 vacuum chamber, with pressures in the range of 10^{-8} Pa to 10^{-9} Pa. The specimen is
117 mounted on a stage that is cooled to cryogenic temperatures in the range of 20 K – 80 K,
118 typically by a combination of a cold finger at the end of a closed-circuit helium-based cryo-
119 cooler combined with a resistive heater that regulates the temperature. The stage holds the
120 specimen in front of a counter-electrode located from a few tens of microns to a few
121 millimetres away and is connected to a direct-current high-voltage power supply commonly
122 of up to 15 kV. In the most frequently found commercial design (the Local Electrode Atom
123 Probe or LEAP^{22,24}), the stage can move in three-dimensions to facilitate specimen
124 alignment in front of a micro-electrode with an aperture diameter of ~ 40 μm . Prototypes
125 using a similar assembly had previously been designed^{25,26}, but the high throughput of the
126 LEAP instrument, which allows for loading multiple specimens at once to analyse them
127 successively, significantly facilitated the spread of the technique. Note that commercial
128 equipment is identified here to adequately specify the experimental conditions and does
129 not imply any endorsement by the National Institute of Standards and Technology that it is
130 the best for the purpose.

131 The field evaporation and subsequent measurement of the time-of-flight of each ion is
132 triggered by pulsing either the high voltage (to vary the electric field), or by laser pulses (to
133 vary the temperature). In the former case, negative high-voltage pulses lasting a few
134 nanoseconds^{27,28} and with an amplitude of 10 %–25 % of the applied DC voltage are
135 transmitted onto the counter-electrode. In the latter case, a pulsed-laser beam is focused
136 towards the specimen apex. While early designs of atom probes used nanosecond pulsed-
137 laser^{29,30}, state-of-the-art instruments use pulses in the pico- to femtosecond times range<sup>31–
138 33</sup> with wavelengths typically in the near-ultraviolet (UV) (near 350 nm). Research
139 instruments can also be equipped with laser sources with a variable wavelength^{34,35}.

140 Beyond the counter-electrode on the path of the ions, ion-optical devices are sometimes
141 used^{36,37}, with the most popular so far being the **reflectron** [G]^{38–40}. A reflectron is the
142 equivalent to an electrostatic mirror that bends the ion trajectories, as shown in **Error!**
143 **Reference source not found.**b. It offers the possibility to extend the time-of-flight of the
144 ions and hence the relative precision of the measurement and the mass resolution⁴¹.

145 Finally, 10 cm–50 cm away from the specimen is the single-particle delay-line detector,
146 which combines an assembly of microchannel plates (MCPs) and an anode. The MCPs
147 convert the impact of a single ion into hundreds of thousands of electrons⁴². The anode
148 collects the electrons, and on state-of-the-art atom probes, contains three **delay-lines** [G]
149 ^{43,44}. The electronic signals detected at each end of each delay-line are processed to
150 calculate the impact position. In cases where multiple ions strike the detector nearly
151 simultaneously, the signals from the third line are used to help disambiguate the signals on
152 the other lines to maximise the accuracy of the positioning information recorded⁴³. The
153 processing of the signals, their recording, and the association of an impact position to a
154 time-of-flight for each detected ion are done digitally, either during the acquisition or during
155 post processing of the data.

156 [H2] Field evaporation

157 [H3] Metals

158 APT is underpinned by the fact that an intense electrostatic field can cause the desorption
159 and ionisation of atoms from the surface of a material in a field evaporation process^{45,46}.
160 This requires that a critical field, termed the evaporation field, is reached. Field evaporated
161 ions are accelerated by the electric field away from the specimen's surface. Field
162 evaporation is primarily a thermally assisted process⁴⁷, even though there have been
163 reports of field evaporation via an ion tunnelling process⁴⁸. The prevalent theory states that
164 the departing ion is singly-charged and, during the early stages of the flight, is further
165 ionised as one or more electrons tunnel back into the specimen⁴⁹ (**Error! Reference source
166 not found.**c). The probability of these post-ionisation events is directly related to the
167 successive ionisation energies of each atom and the strength of the electric field. The ratio
168 of the charge states of the various species present in the specimen can therefore be used as
169 a proxy to estimate the intensity of the electric field^{50–53}.

170 [H3] Non-conductors

171 The mechanisms discussed above have been established in the study of metals. Several
172 differences appear in semiconductors and insulators (see BOX 1 and Figure 2d). One
173 difference is a high fraction of multiple events where more than one ion is detected after a
174 single high-voltage or laser pulse, with these multiple events having two origins. First,

175 following the field evaporation of a surface atom, the charges at the surface rearrange to
176 maintain the screening of the electrostatic field, as recently imaged by FIM⁵⁴, which can lead
177 to the rapid field evaporation of neighbouring atoms. This seems to occur more for
178 materials that are less electroconductive, which may be due to differences in carrier density
179 or mobility. For some semiconductor materials, for instance thermoelectrics, the proportion
180 of multiple events was extraordinary high^{55,56} suggesting that materials properties play a
181 crucial role in how the bonds break during APT experiments. Second, the high fraction of
182 **molecular ions [G]** increases the likelihood of dissociation.

183 Advanced analyses of the APT data containing a high proportion of molecular ions and/or
184 multiple events can provide information about which species are most susceptible to
185 simultaneous evaporation and information about dissociation pathways^{57,58}. These issues
186 affect, for instance, carbides⁵⁹, leading to known issues in the quantification of carbon in
187 steels⁶⁰. These analytical problems arise as a result of the functional properties of the
188 surface such as band gap, low effective mass of electrons, or the high anharmonicity of
189 chemical bonds (equivalent to a high Grüneisen parameter) in part modified by the intense
190 electrostatic field.

191 While many details of the physics of the field evaporation of insulators and semiconductors
192 remain elusive, there are indications^{61,62} of similarities with processes in metals. Although
193 maybe not quantitatively transferable, the dependencies of the charge states of the emitted
194 ions as a function of the electric field^{30,63,64} and the relative stability of molecular ions⁶⁵
195 appear to follow the same trend.

196 There are numerous reports of the influence of the field evaporation conditions on the
197 resulting analytical performance of APT^{64,66-70}, and in BOX 2 we provide some guidelines to
198 help optimise data quality.

199

200

201 [H2] Specimen preparation

202 [H3] From bulk materials

203 The preparation of specimens suitable for APT can be seen as a (dark) art. Field ion
204 evaporation requires electrostatic fields on the order of ~ 10 V/nm in order to selectively
205 and sequentially remove surface atoms. To achieve the fields required, atom probe
206 specimens are formed in the shape of a sharp needle, with an apex radius in the range of \sim
207 100 nm. An analytical model ($F=V/kr$) relating the surface electrostatic field (F) to the end
208 radius (r) and applied voltage (V) through a field-reduction/field-factor (k) has been derived
209 from simple electrostatics⁶ and typical k values range from 2–8^{71,72}. A modest applied high
210 voltage, on the order of 10^3 V, can be used to achieve the necessary electrostatic field.
211 Historically, before the advent of focused ion beam (FIB) milling systems, metallic specimens
212 were prepared from a wire or a small match-stick-shaped blank and turned into a needle by
213 electrochemical polishing with various alkaline or acidic solutions^{73,74}, or, specifically, for
214 noble metals, molten salts⁷³. The blank is moved through a thin layer of the solution in order
215 to progressively reduce the cross-section. At the point of rupture, the end radius or tip apex
216 was often below 100 nm. Targeting specific microstructural features, however, was
217 challenging and could only be achieved via successive observations by TEM followed by
218 pulsed electropolishing, for example^{75,76}.

219 Instrumental to routine targeted preparation was the development of liquid-metal ion
220 sources⁷⁷ and the development of dual-beam SEM/FIB systems, which are now used to
221 prepare multiple site-specific atom probe specimens in a single session⁷⁸. The SEM is used to
222 identify a region of interest and the FIB is used to prepare the specimen itself. The current
223 established protocol is briefly outlined in **Error! Reference source not found.a** and discussed
224 as follows: first, a 15 μm – 20 μm long cantilever is cut with the FIB from the material
225 sample's surface; second, using the in-situ gas-injection system, the micromanipulator is
226 attached to the wedge, which is cut free and lifted out; third, the wedge is attached on a
227 support, most often a **microtip coupon [G]**⁷⁹ or a TEM half grid, and sliced; fourth, each slice
228 is then turned into a needle-shaped APT specimen through a series of sequentially smaller
229 annular milling routines in the FIB. The final end apex radius is approximately 100 nm
230 depending on the material of interest. While FIBs typically use a liquid metal gallium source,
231 plasma-based sources have been used with increased success^{20,80–82}. Alternative and
232 variations around this principle have been proposed and are thoroughly reviewed in^{83,84}.

233 For site-specific sample preparation, microstructural features of interest are identified by
234 using, for instance, back-scattered imaging, electron-backscattered diffraction, electron-
235 channelling contrast imaging, or EDS in the SEM⁸⁵. Additional information can be gathered
236 by inspecting the APT specimen by TEM⁸⁶ using an appropriately-sized TEM grid as a
237 support⁸⁷, as outlined in **Error! Reference source not found.b**. In this case, a feature is
238 identified by SEM and marked by electron-beam or ion-beam deposition inside the SEM/FIB.
239 The marked region is extracted, and a slice is attached to a partly electropolished half-grid⁸⁸.
240 The slices are sharpened and the feature of interest positioned within 200 nm of the apex.
241 The final specimens are subjected to low acceleration voltage (2 kV – 5 kV) milling to
242 remove the regions of the materials damaged by the energetic incoming ions.

243

246 APT analyses of nanostructures such as nanoparticles or nanowires require dedicated
247 specimen preparation approaches⁸⁹. Direct deposition of nanoparticles onto pre-sharpened
248 specimens in vacuum⁹⁰ or electrophoresis in solution^{91,92} have led to successful analysis. The
249 second approach is illustrated **Error! Reference source not found.a** where the specimen is
250 inserted into a drop of solution containing nanoparticles, and, by applying voltage pulses,
251 negatively charged nanoparticles are attracted to the positively-charged needle. The
252 number of deposited layers is controlled by adjusting the concentration, pulse amplitude or
253 duration. TEM shows one to two layers of nanoparticles deposited on the support needle in
254 **Error! Reference source not found.b**. This method is suitable for 1 nm – 60 nm
255 nanoparticles and is limited by the size of the support needle. **Error! Reference source not
256 found.c** exemplifies successful APT analyses of 8 nm – 10 nm silver atoms (in grey) forming
257 the core of nanoparticles and palladium (in yellow) forming a shell, with two different sets
258 of nanoparticles imaged with various core-shell thicknesses. A range of other
259 nanostructures have been successfully analysed following this preparation approach^{91,93–95}.
260 The main drawback is a low yield and poor data quality because the particles are only
261 loosely bonded to the support needle and complete nanoparticles can simply detach. A
262 similar method based on the use of a pre-sharpened metallic needle has enabled the
263 analysis of a variety of materials ranging from metallic multilayers^{96,97} to polymer films⁹⁸ or
264 self-assembled monolayers^{99,100}.

265 Flat-top Si microtips, individual needles, or TEM half-grids have been used as supports with,
266 for instance, a single nanoparticle or nanowire placed on the surface by using a
267 micromanipulator or a microgripper to grab individual^{101,102} or agglomerated
268 nanoparticles¹⁰³ (Figure 4d). Nanowires have been grown^{104,105} and particles deposited by
269 inert gas condensation directly on coupons or flat substrates with a geometry suitable for
270 APT (**Error! Reference source not found.e**)^{106,107}. Subsequent deposition of a protective
271 metal film such as chromium or platinum can help ensure adhesion during preparation and
272 analysis^{101,108} and prevent damage from the gallium-ion beam. The methods in Figure 4a-e
273 enable the analysis of complete nanoparticles. However, agglomerated particles can
274 simultaneously evaporate, causing problems with the APT data reconstruction and
275 interpretation, and they are not amenable to all particle sizes or composition. So far, the
276 community has not established clear guidelines regarding which approach to use to obtain
277 optimal results for a specific sample nature and geometry.

278 An alternative approach is to embed spatially separated nanoparticles in a metal or oxide
279 matrix and then prepare the specimen by traditional FIB lift-out. In **Error! Reference source
280 not found.f**, a drop of a diluted solution containing nanoparticles is deposited on a flat
281 substrate, subsequently coated with a metal film, and finally lifted-out by FIB. Here the
282 wedge is rotated by 90° to prepare the APT specimen with the particle/substrate interface
283 aligned along the specimen's main axis¹⁰⁹. This method has enabled analysis of silica-
284 supported (SBA-15) particles and carbon nanotubes^{110–114}. However, sputter or electron-
285 beam assisted deposition often leads to a non-conformal coating that may leave voids at the
286 interface between nanoparticles and substrate, which lowers the specimen survival rate and
287 can lead to severe trajectory aberrations in the reconstructed data¹¹⁵. Larson *et al.*¹¹⁵ used
288 atomic layer deposition (ALD) to produce a void-free and highly conformal film to sandwich
289 nanoparticles (**Error! Reference source not found.g**) followed by lift-out and sharpening

290 (Error! Reference source not found.h). Kim *et al.*¹¹⁶ used electrodeposition to embed the
291 nanoparticles in a metallic matrix (Error! Reference source not found.i): Error! Reference
292 source not found.j shows a TEM image in which palladium nanoparticles were successfully
293 embedded in a Ni matrix and no voids were observed, and Error! Reference source not
294 found.k shows the corresponding APT dataset. This approach is versatile and has enabled
295 the analysis of nanosheets¹¹⁷, nanowires and nanorods¹¹⁸ as well as nanoparticles. However,
296 it may not be as advantageous for surface sensitive nanoparticles as the techniques in
297 Figure 4f and g: the solution used for the electrodeposition can then react with the surface
298 and modify the nanoparticles.

299 [H2] Recent developments in instrument design

300 Most atom probes in operation worldwide are commercial instruments. CAMECA currently
301 leads the market with the LEAP and, to a lesser extent, the EIKOS series. A start-up company
302 (INSPICO) has recently started commercially manufacturing instruments. Some groups
303 develop their own instruments for a bespoke setup or to pioneer the implementation of
304 new capabilities such as detectors or extreme vacuum levels, for instance. Since its
305 introduction in 2015, the current leading commercial instrument is the LEAP 5000 series. It
306 combines the local electrode [G] concept with micron-size laser spot size, with 10 ps laser
307 pulses in the near-UV (355 nm) to minimize thermal tailing effects, as already demonstrated
308 in the earlier LEAP 4000 generation instrument. The current detection efficiency, defined as
309 the fraction of field evaporated ions that are detected and counted, reaches 80 %. The
310 voltage pulsing repetition rate reaches 500 kHz and in laser mode 1 MHz, promising faster
311 measurements. In reality, the evaporation behaviour of the material and the geometry of
312 the specimen dictate the optimal conditions and hence acquisition time.

313 APT specimens are small, with a high surface-to-volume ratio, and materials can be reactive
314 so their transport through air can modify their composition and change their survivability
315 during the analysis. Worldwide, there are increased efforts regarding sample preparation,
316 transport, processing and handling in protective atmospheres or under cryogenic
317 conditions. This was in part inspired by approaches developed in the biological sciences
318 where sample environment can be crucial. These new workflows are particularly critical for,
319 for example, liquid materials or air sensitive samples likely to react with oxygen, and involve
320 cryogenic-vacuum-transfer solutions. Custom-designed or commercial (Leica, Quorum,
321 Ferrovac) enclosed controlled environmental transport systems, termed ‘suitcases’, have
322 been adapted or developed to enable controlled specimen transfer into the atom probe<sup>119–
323 121</sup> from the FIB. These suitcases can reach high or ultra-high vacuum conditions and can
324 typically be cooled down to liquid nitrogen temperature. These cryogenic capabilities have
325 enabled the analysis of hydrogen/deuterium-charged specimens^{120,122,123}, frozen
326 liquids^{124,125}, and hydrated porous specimens¹²⁶. There are also reports of the importance of
327 using cryogenic cooling during the final stages of specimen preparation^{127–129} or for full lift-
328 outs^{126,130} to avoid introduction of spurious species and limit damage, or maintain sample
329 hydration. These approaches fit within the effort to facilitate correlative microscopy (BOX
330 3), which combines specific techniques to deliver a more complete set of information on a
331 material, and thereby enable scientists to derive well-founded conclusions.

332

[H1] Results

Following specimen preparation and setting up the experiment in order to perform an APT analysis, the collected data needs to be extracted and processed. The raw data consists of a sequence of 2D detector-space coordinates, and the corresponding time-of-flight of every ion is measured over the course of the experiment. This section goes step-by-step into the data 'reconstruction' process to transform the raw data into a real-space point cloud representation of the specimen, within which both the 3D position and the chemical identity of every detected ion has been determined. The processing of the data is similar on commercial and bespoke instruments.

[H2] Mass spectrometry

The first step in the data processing workflow is to assign a chemical identity to each ion. Time-of-flight can be converted to mass-to-charge-state ratio through a simple equation relating the kinetic energy of the ion $\frac{1}{2}mv^2$ to its potential energy neV , where m is the mass, v is the ion velocity, n is the unit charge of the ion, e is the elementary charge of an electron, and V the applied acceleration voltage. At a given acceleration voltage, an ion's kinetic energy is related to its mass (a heavy ion travels more slowly than a lighter ion) and its charge (a doubly charged ion acquires twice the energy of a singly-charge ion). A couple of calibration and correction steps for the difference in flight length as a function of detector coordinates are commonly applied¹³¹. A histogram, better known as a mass spectrum, is generated; an example for a steel sample is shown in a–b. Each peak or series of peaks is related to the detection of specific types of ions. Each peak in the mass spectrum, and therefore every detected ion contributing to this peak, is assigned a chemical identity. This process is known as 'ranging' [G]. The boundaries (lower and higher mass-to-charge values of each individual mass peak) are defined to associate a range of mass-to-charge to a single element or a combination of elements and the discrete counts in the peak are then calculated through integration¹³². Ranging allows for measuring the composition, and each ion that has been identified is assigned a certain volume to contribute to the three-dimensional reconstruction.

The resolution is such that individual isotope peaks are routinely resolvable. The ability to resolve different isotopes and to locate them individually within the microstructure also enables some unique insights into a variety of investigations, such as the age and history of ancient geological materials¹³³ or the role of irradiation-induced transmutation in the microstructural degradation of components in nuclear reactors^{134,135}.

Typically, ions are detected in the 1^+ or 2^+ charge states, but higher charge states can also be observed depending on the element and the intensity of the electric field⁴⁹. Molecular ions are also commonly detected. For example, in the mass spectrum resulting from the analysis of a steel shown in a, $^{12}\text{C}_2^{2+}$ and $^{56}\text{Fe}^{14}\text{N}^+$ are observed. In most cases, all isotopes are expected to be detected in their natural abundances. However, rare exceptions may exist for elements that have undergone biotic fractionation or elements subject to nuclear reactions. Detection of higher charge states and complex ions complicates the ranging process as it increases the likelihood of peak overlap, because the detection of two or more distinct types of ion with the same mass-to-charge-state ratio will contribute to the same

377 peak in the mass spectrum. For example, in a (inset), the peak at 14 Da could potentially be
378 assigned to $^{14}\text{N}^+$ or $^{28}\text{Si}^{2+}$. If this peak were to incorporate only silicon, then we would also
379 expect to see adjacent peaks at 14.5 Da and 15 Da corresponding to $^{29}\text{Si}^{2+}$ and $^{30}\text{Si}^{2+}$, and
380 expect that the ratio of the peak heights to one another would match the ratio of their
381 respective natural isotopic abundances. Such isotopic fingerprints provide confidence that
382 peak identities have been correctly assigned during the ranging process. Furthermore, the
383 existence of adjacent peaks due to the detection of isotopes provides a route for estimating
384 the relative contributions of different ionic species to the same overlapping peak¹³⁶.
385 Additional discussion on these aspects can be found in Box 5.

386 387 [H2] Three-dimensional reconstruction

388 APT, like FIM, is a point projection microscope, wherein the specimen acts the projection
389 optic, with no additional lenses. The typical configuration of the atom probe is depicted in
390 c: ions are emitted by the specimen with a radius R of less than 100 nm, and fly over a
391 distance L to the detector with a diameter of 80 mm–200 mm. The trajectory of the
392 evaporated ions towards the detector is determined by the electric field generated by the
393 specimen and is therefore influenced by the electrostatic environment, which includes both
394 the surrounding instrumentation and the shape of the specimen itself. Although initially
395 near-radial, the ions' trajectories are curved towards the centre of the detector and the
396 projected image is subject to what is known as **image compression [G]**. Importantly,
397 provided that they fly in a field-free tube, the trajectory of the ions is not dependent on the
398 ion's mass, its charge nor the specific value of the voltage⁷. The resulting magnification onto
399 the detector is of the order of 10^6 . Multiple projection models have been shown to reflect
400 well the imaged emitter's surface onto the detector, including a quasi-stereographic and
401 azimuthal equidistant projections^{137–139}. d shows the results from projection models used
402 to describe the ion projection in APT. Ultimately, the interest of using a projection law is
403 that it is not necessary to calculate the trajectory of each ion from the detector back to the
404 specimen, and one can then use one of these projection laws as an approximation to relate
405 a set of detector coordinates (X_D^i, Y_D^i) to a unique position at the specimen's surface. Even
406 though it is not the most accurate^{137,139}, the most commonly implemented projection is
407 **quasi-stereographic [G]**, and the image compression factor ξ is experiment-dependent with
408 a value generally between 1 and $2^{72,140,141}$.

409 Assuming a projection model, the generation of the three-dimensional atom-by-atom
410 images is a two-step process. First, based on their impact coordinates, each ion striking the
411 detector (X_D^i, Y_D^i) is 'de-magnified' by using a reverse-projection model to position them back
412 to their original real-space location within the specimen $(x^i, y^i, z^i)^{141,142}$. The key assumption of
413 the current implementations of the reverse-projection approach is that the geometry of the
414 atom probe specimen always takes the form of a perfect hemispherical cap on a truncated
415 cone and that the radius of this cap is known at every point in the experiment. As material is
416 removed from the specimen during the experiment, the tip becomes increasingly blunt. The
417 implication is that the magnification is not constant throughout the analysis and that the
418 accurate reverse-projection of each ion from detector space to real space requires an
419 instantaneous estimate of the end-form radius of the specimen at the moment of every
420 detection event. This is usually achieved either monitoring the evolution of the voltage
421 applied throughout the experiment given that $R^i = \frac{V^i}{Fk}$, or by using an assumed specimen

422 shape to derive an estimate of the contribution of each ion to the blunting of the tip^{141,143}.
423 Either of these approaches involves several parameters, such as the strength of the field F
424 and the geometric field factor k , or the shank angle of the specimen.

425 The second step is the calculation of the depth coordinate. The conventional algorithm
426 assumes that the first detected ion is evaporated from a hemispherical surface, $R^{i=1}$, and
427 that the depth of this emitting surface moves down by an increment $\Delta_z^{i=1}$ that is
428 proportional to the volume occupied by an individual atom within the material's
429 lattice^{141,142}. The need to assign a realistic volume is why the ions must each be identified
430 through ranging prior to reconstructing the point cloud. Indeed, a volume is assigned to
431 each detected and ranged atom, and that volume is considered proportionally larger to
432 account for the limited detection efficiency. The depth increment represents the thickness
433 of the volume of the preceding detected atom if spread across the entire surface of the
434 specimen that is within the field of view of the detector. In general, for the i^{th} detected ion,
435 this increment is additive, incorporating contributions from every previously detected ion in
436 the sequence. Finally, the z-coordinate of an ion is the cumulative sum of these increments
437 added to the z-position from the reverse-projection. This is summarised in **Error! Reference**
438 **source not found.e**.

439 Calibration, or incorporation of physical data about the specimen obtained through a
440 correlative or complementary technique, plays an important role in ensuring the accuracy of
441 subsequent spatial measurements made within the APT reconstruction (BOX 4). This is
442 especially important since there is not necessarily a unique set of reconstruction parameters
443 leading to a specific reconstruction. An example of a reconstruction is shown in **Error!**
444 **Reference source not found.aError! Reference source not found.**, where elemental
445 identity, in this case aluminium, zirconium, magnesium, and copper were first assigned
446 through ranging of the mass spectrum. The point cloud was then built and calibrated based
447 on the known spacing of the (002) planes in one of the grains.

448

449 [H2] Data visualisation and analysis

450 Visualisation of APT reconstructions can offer immediate and striking insights into the
451 microstructure of a complex material. For example, the points pertaining to atoms of
452 different species can be selectively displayed to reveal their respective spatial distribution,
453 as in **Error! Reference source not found.Error! Reference source not found.b** where
454 chromium partitioning to the γ phase and aluminium to γ' can be seen visually. Following
455 the reconstruction process, some of the most unique and powerful analyses that can be
456 undertaken have roots in the fundamental nature of the 3D atom-by-atom APT
457 reconstructions. For example, changes in chemical composition can be characterised along a
458 specific direction within a region of interest in order to calculate a one-dimensional
459 composition profile, for example to quantify segregation at a microstructural feature of
460 interest such as a grain boundary, as seen in **Error! Reference source not found.c**. More
461 advanced APT data analysis can be broadly divided into two classes based upon either
462 **voxelization [G]** of the data or the interrogation of inter-atomic distances.

463 [H3] Voxel-based methods

464 Voxel-based methods segment the dataset into an array of cubes of equal volume or
465 number of atoms. The content of each voxel is then inspected, enabling a wide variety of

466 visualisation and analysis. Possibly the most commonly-used voxel-based analysis is the
467 **isosurface [G]** that link together an array of adjacent voxels based on a threshold criterion.
468 Two examples are displayed in **Error! Reference source not found.Error! Reference source**
469 **not found.b** for aluminium and boron. The threshold can either be a molar fraction or a
470 number density of a chosen element (or elements)¹⁴⁴. A molar fraction threshold leads to an
471 **iso-concentration [G]** or composition surface encompassing regions within the point cloud
472 that contain over a certain molar ratio expressed in atomic %, while an element number
473 density threshold is an isodensity surface expressed in at.nm^{-3} . The type of isosurface and
474 the value of the threshold are selected by the user based on what they wish to visualise.
475 Features revealed by isoconcentration or isodensity surfaces may not be similar. In
476 particular, variations in point density are typically associated with local magnifications (see
477 below) and hence with microstructural features that do not necessarily show compositional
478 variations, for example grain boundaries^{145,146}. With careful adjustment of the threshold,
479 the user can define the interface between contiguous microstructural regions, for example a
480 second phase precipitate and the surrounding matrix, or two adjacent layers in a multi-layer
481 device. This provides a means to both isolate regions of the data for more targeted analysis,
482 as shown in the inset of **Error! Reference source not found.Error! Reference source not**
483 **found.d**, and characterize the chemistry at and in the vicinity of the interface. Statistical
484 measurements, such as voxel-concentration frequency distributions, can also identify and,
485 to some extent quantify, subtle chemical inhomogeneity within the microstructure¹⁴⁷⁻¹⁴⁹.

486 The compositional evolution of interfaces delineated by isosurfaces can be derived from a
487 profile calculated in the form of a proximity histogram, better known as a proxigram^{150,151},
488 which represents chemical concentration as a function of the distance normal to the
489 isosurface. The proxigram in **Error! Reference source not found.Error! Reference source**
490 **not found.d** provides similar information to that of a 1D composition profile but facilitates
491 the analysis of curved and/or enclosed interfaces. This approach is not without issues¹⁵², but
492 it readily enables an average measurement across a number of disconnected surfaces such
493 as a population of second phase precipitates. Concentration profiles can be further
494 extended to actually quantify the elemental excess number of atoms segregated to a
495 surface^{153,154}. Voxelization also provides a means to create a 2D map or 3D visualisation of
496 how concentration and density fluctuate throughout the reconstruction. To this end, fixed
497 voxel size can be advantageously replaced by a local meshing on which the composition or
498 excess can be efficiently mapped^{155,156}, revealing and quantifying patterns in the segregation
499 that can be associated to grain boundary curvature¹⁵⁷, faceting¹⁹, or to the presence of
500 linear defects at low-angle grain boundaries for instance¹⁵⁸.

501 [H3] Interatomic-distance-based methods

502 Other approaches involve the direct measurement of interatomic distances and enable the
503 interrogation of the local neighbourhood surrounding each atom. The simplest of such
504 analyses is a nearest neighbour analysis^{159,160}. By examining the distribution of distances
505 separating a solute atom from its 1st, 2nd, 3rd, or nth nearest solute neighbour, in comparison
506 to the same measurement undertaken on a complementary randomised system, the
507 presence of subtle fluctuations such as the very onset of nanoscale clustering effects¹⁶¹
508 (**Error! Reference source not found.Error! Reference source not found.e**) and interactions
509 at longer distances can be used to deduce the matrix composition^{162,163}. Radial or pair
510 distribution functions¹⁶⁴⁻¹⁶⁶ revealing specific interactions between solutes can also be

511 extracted from integrating the average three-dimensional neighbourhood of each selected
512 solute atom within the reconstruction.

513 A unique feature of APT is to characterise solute clusters identified based on inter-atomic
514 distance. The most widely used approach is known as the 'maximum separation'
515 algorithm^{167,168}. This assumes that the distance separating two nearest neighbour solutes
516 within a cluster is less than between two nearest neighbour solute atoms within the matrix.
517 Hence, a maximum distance, d_{\max} , can be assigned such that any pair of solute atoms within
518 this separation are considered to be clustered together. Further, if one of these pairs is also
519 clustered to another solute, then all three can be defined as being in the same cluster. In
520 this way, a larger distinct network of linked solutes can be built that ultimately defines a
521 population of clusters. This initially excludes solvent atoms from the cluster definition.
522 However, in a second stage these can be incorporated into the analysis in processes known
523 as 'enveloping' and 'erosion'^{168,169}. Once clusters have been identified, a variety of
524 measurements can be applied to characterize each one individually in terms of size, shape,
525 composition, structure, etc^{170,171}. As such, statistical analyses can be generated to describe a
526 population of clusters and identify correlations between physical characteristics. Typically, a
527 comparison to a randomly-labelled dataset is needed to differentiate non-random clustering
528 from the clusters that might be expected even if the solute was randomly distributed. Other
529 algorithms have been developed^{161,172}, underpinned by different cluster definitions based
530 on concentration¹⁷³, higher-order nearest neighbour distances^{159,160,174}, Gaussian mixture
531 models¹⁷⁵, and more^{1,176,177}. These methods were authoritatively reviewed previously^{161,172}.

532 Ultimately, results from applying either of these approaches must be considered very
533 carefully, in particular when claiming quantitiveness and reproductibility¹⁷⁸. Objectively
534 defining the extent of a nanoscale cluster of atoms within a reconstruction remains a
535 challenge, whichever approach is used. In particular, even though the APT reconstruction
536 can be highly accurate, it is nevertheless both imperfect and incomplete. The inherent
537 premise upon which the approach is based is not amenable to certain situations; for
538 example, delineating the clusters from the matrix becomes difficult if the solute
539 concentrations are relatively close to that of the matrix. The difference in the evaporation
540 field between the solutes and the matrix, and the dependence of this critical field on the
541 local neighbourhood, also introduce aberrations in the trajectories that break
542 neighbourhood relationships. For example, two nearest neighbours at the specimen surface
543 are unlikely to be so in the reconstructed data if they are both high-field solutes¹⁷⁹. Finally,
544 the main source of error is likely the parameter selection, which has the potential to
545 considerably affect the resulting analysis¹⁸⁰. Protocols exist, but none are universally
546 applicable nor adopted as a community standard. Selecting parameters by defining a
547 heuristic^{69,170,181,182}, including using statistical methods¹⁷⁵, has been proposed but the
548 outcome of applying cluster-finding algorithms remains highly dependent on user input and
549 expertise level^{178,180}. Alternative approaches using radial-distribution functions to extract
550 the typical characteristics of a population of solute clusters or precipitates¹⁸³ have also been
551 proposed, with the option to select parameters.

552 [H3] Structural analysis

553 For some alloys, the APT reconstruction can retain elements of the crystallography from the
554 original sample¹⁸⁴, as illustrated in **Error! Reference source not found.a**. This partial
555 information appears as sets of atomic planes in certain regions of the data, most often
556 associated to the projection of low-Miller indices lattice planes parallel to the local tangent

557 to the reconstructed emitting surface. Means to extract quantitative information from this
558 crystallographic information have been developed based on direct Fourier Transforms¹⁸⁵,
559 3D-Hough Transforms^{186,187} and techniques based on radial-distribution functions¹⁶⁵
560 including the spatial distribution maps (SDMs)¹⁸⁸⁻¹⁹⁰.

561 The Fourier transform is akin to performing diffraction from the three-dimensional
562 distribution of atoms in the reconstructed volume. It makes use of a voxelised reciprocal
563 space and calculates the contribution of all points in the real space to this particular location
564 in the reciprocal space. The presence of sets of planes results in loci of high intensity in the
565 reciprocal space at a distance to the central spot inversely proportional to the distance in
566 the real space. This approach enables multiple sets of planes to be characterised at once,
567 and the angle between sets of planes can be directly estimated as they are maintained
568 through application of the Fourier transform. The 3D Hough transform provides a similar set
569 of information but is more computationally intensive, as the computation is done
570 completely in real space and looks for planar features positioned at all possible polar and
571 azimuthal angles.

572 A SDM is built by using a sphere, typically 2 nm in diameter, located on a first atom within
573 the input point cloud and all atoms within this sphere are copied into an output. The sphere
574 is then moved onto the next atom in the input data and these new neighbouring atoms are
575 copied into the output. This procedure is reproduced for all atoms in the input data, and the
576 output hence depicts the average three-dimensional atomic neighbourhood around each
577 atom within the input dataset. If the dimensionality of the data was reduced to 1, then this
578 would result in a radial distribution function. SDMs are displayed either as a histogram of
579 atomic offsets in z along a specific direction (z-SDM) or as a two-dimensional map showing
580 the average density of atoms within the plane of the reference atoms (xy-SDM). The peak-
581 to-peak distance in the z-SDM indicates inter-planar spacing and measures the inter-atomic
582 separation along the direction perpendicular to the planes. Being computationally
583 expensive, SDMs are usually only used in regions of the data where planes are resolvable¹⁸⁸⁻
584 ¹⁹⁰. If angles between a set of a planes and a reference plane can be deduced from another
585 SDM implementation^{190,191}, it is typically not performed because of the computational cost.

586 The term atom probe crystallography was coined to refer to the use of this type of
587 structural information to provide additional information on the analysed material^{158,184}. The
588 main applications of these approaches, in particular the Fourier transform and SDMs, has
589 been to facilitate the calibration of tomographic reconstruction¹⁹² and assess the spatial
590 resolution of APT^{193,194}. The high computational cost and mostly manual operations to
591 extract information and interpret the data may explain why it is not more widely used
592 despite great potential¹⁹¹.

593 [H1] Applications

594 3D compositional data provided by atom probe is highly complementary to, for instance,
595 the high spatial resolution 2D images obtained from TEM. Atomic maps provide valuable
596 information about the common microstructural features in engineering materials, most of
597 which involve variations in the local composition and can often be examined within the size
598 scale of a typical atom probe dataset volume. These microstructural features include
599 precipitates or clusters, solid solutions and/or ordered structures, grain/phase boundaries,
600 dislocations, and point defects/vacancies. Numerous reviews detail the applications of APT

601 to characterise for example aluminium-alloys^{161,195}, nickel-based superalloys^{196–198},
602 intermetallics¹⁹⁹, steel^{200,201}, high-entropy alloys²⁰², nuclear materials^{203–205}, and, beyond
603 metallurgical systems, thermoelectrics²⁰⁶, semiconducting devices for microelectronics^{207–}
604 ²⁰⁹ or light-emission²¹⁰, and geological materials^{211,212}. Here we give some details of what
605 information APT provides in these various contexts.

606 [H2] Solid solutions

607 With access to a large enough 3D dataset, the statistical approaches discussed above
608 provide information on the arrangement of solute atoms in a solid solution. These insights
609 enable the study of interactions between atoms from a single species or from one species to
610 one or more species. In solid solutions that are not at an equilibrium state, it is common to
611 observe certain species being preferentially located at shorter distance to one another
612 compared to a random distribution, as illustrated in Figure 6**Error! Reference source not**
613 **found**.e. There are many examples of the use of APT to study the early stage of
614 decomposition of a solid solution in metals^{166,195,213–216} or dopant distributions in
615 semiconductors^{160,217,218}, sometimes in conjunction with SIMS²¹⁹. Short-range order can be
616 detected in principle, but depending on the solute concentration, the results may be
617 obfuscated by the effect of the combination of detector efficiency and the spatial resolution
618 within the APT data^{1,220}.

619 [H2] Precipitation

620 Precipitation of a secondary phase from a metastable solid solution is a common strategy to
621 modify properties of materials. APT is commonly used to provide the composition of
622 precipitates, as shown in **Error! Reference source not found**.a and b**Error! Reference source**
623 **not found**.. APT can also be used to study compositional gradients in the vicinity of grain
624 boundaries or interphase interfaces, as in **Error! Reference source not found**.**Error!**
625 **Reference source not found**.c for instance, which can help to understand their growth or
626 migration behaviour. Differences in the field evaporation properties between the matrix
627 and precipitate can affect the measured size and composition²²¹ and should be considered
628 when interpreting data. Gradients can originate from coarse binning of data or from data
629 voxelisation^{144,152} and again, great care should be taken during data interpretation. The
630 study of precipitation has been at the core of APT research for decades and many of the
631 aforementioned review articles are focused on this subject.

632 [H2] Grain boundaries and interfaces

633 Grain boundaries and interfaces are important factors in relating a material's properties to
634 its microstructure. Their precise influence often depends on their composition, which in
635 turn, depends on their crystallographic nature¹⁸. It is common for certain elements to
636 segregate to grain boundaries and interfaces, typically driven by the minimisation of the
637 system's free energy. This segregation can be beneficial or detrimental, depending on the
638 desired set of physical properties. For instance, grain boundaries affect electronic transport
639 and therefore underpin the performance of multi-crystalline photovoltaic materials^{222–224},
640 and several studies have focused on using APT in correlation with electron-beam induced
641 current^{225,226}. This is also the case across interfaces in thin films for instance, finding
642 applications in microelectronics and memory^{227–232}, light emitting^{233–235} devices,
643 photovoltaics^{236–238} or magnetic reading/recording^{96,239,240}, and thermoelectrics^{206,241–243},
644 amongst others.

645 The inherently three-dimensional nature of APT data offers a unique opportunity for
646 measuring segregation at grain boundaries and interfaces. Targeted analysis provides
647 quantitative information about the extent of segregation either in the form of an atomic
648 fraction or an **interfacial excess** [G]²⁴⁴. This is exemplified in panels **Error! Reference source**
649 **not found.a–b** of BOX 3 on correlative microscopy in a solar cell material, where grain
650 boundary segregation of sodium passivates the existing detrimental charge defects. It is also
651 displayed in panels c–d of BOX 3, which shows an intermetallic phase where the formation
652 of the twin boundary locally changes the composition, thereby modifying the magnetic
653 domain structure and underpinning promising magnetic properties²⁴⁵. In **Error! Reference**
654 **source not found.Error! Reference source not found.c**, boron appears almost completely
655 segregated at the grain boundary, and it is typically added to Ni-based superalloys to
656 strengthen grain boundaries.

657 It is possible to map the extent of segregation across certain surfaces¹⁵⁵ in two-dimensions
658 and relate it to the grain boundary type²⁴⁶. Segregation mapping reveals that, while special
659 boundaries such as twins tend to have lower segregation levels than high angle boundaries,
660 the segregation behaviour at high angle boundaries is highly spatially variable even across a
661 single boundary¹⁵⁵. Grain boundaries have a distinct structure and composition and can
662 undergo phase-like transitions, called complexion transitions, that occur under conditions
663 distinct from phase transformations²⁴⁷. Taking advantage of these processes to tailor alloy
664 properties is the focus of much contemporary research²⁰¹. APT, combined with TEM, has
665 been used to identify complexions such as segregation-induced faceting transitions^{19,248,249}
666 and spinodal-like fluctuations²⁵⁰.

667 [H2] Dislocations, stacking faults and twins

668 Dislocations are linear defects that accommodate strain in crystalline materials. It is
669 common for dislocations to be surrounded by an excess of certain solute species known as a
670 Cottrell atmosphere. The solute can affect the mobility of dislocations and have an effect on
671 the mechanical behaviour. APT provided the first direct observations of Cottrell
672 atmospheres, first for carbon in steel²⁵¹ and later in many other systems such as boron in
673 FeAl²⁵², and arsenic in Si²⁵³. A dislocation with segregation is shown in panel **Error!**
674 **Reference source not found.f** of BOX 3. Dislocations can also form loops with segregation at
675 specific edges or facets^{254,255} for instance. The three-dimensional nature of the data allows
676 the mapping of segregation along the dislocation line^{256,257}, sometimes referred to as the
677 'line excess'. It has been found that this segregation varies along the line of a defect²⁵⁸,
678 presumably due to the different local crystallographic environment. Atom probe has also be
679 used to demonstrate the existence of linear complexions—chemically and structurally
680 distinct regions located inside a linear defect²⁵⁹, and to study other structural defects such
681 as stacking faults^{260–263}, anti-phase boundaries^{264,265} or (nano-)twins^{266–268}, and twin
682 boundaries^{269–271}.

683 [H2] Microstructural degradation processes

684 An important application of APT has been to understand the microstructural evolution
685 leading to the degradation of a material's properties that limit their service lifetime, such as
686 for materials used for nuclear power generation. Handling radioactive materials can be
687 challenging. To shield the user from the source of radiation, dedicated facilities such as
688 'hot'-FIBs and atom probes are available in the UK (Oxford), France (Rouen, CEA Saclay), the
689 USA (Idaho National Laboratory), and Japan (CRIEPI), amongst others. Studies on nuclear

690 materials include the formation of secondary phases in materials already in service or in
691 candidate materials^{203,272–274}, welds in engineering parts^{275,276}, and the oxidation and
692 corrosion of pipes and fuel cladding^{277–281}. Research in the area of fuel and waste is also
693 burgeoning^{282,283}. In these systems, a significant challenge is the detection of very light
694 elements, including helium²⁸⁴ and hydrogen.

695 Beyond just nuclear materials, spatially-resolved characterization of hydrogen has been a
696 long-standing frontier in microscopy and microanalysis. Hydrogen is known to embrittle and
697 cause catastrophic failure in many materials, but is also a strong focus for clean energy
698 storage and carbon-emission-free mobility. However, its distribution within material
699 structures is difficult to determine. While hydrogen is readily detected in APT, it is
700 challenging to establish whether it originates from the specimen itself or is residual
701 hydrogen from within the vacuum chamber. Isotopic labelling has been proposed as an
702 approach to circumvent this issue. Samples are charged with deuterium, which serves as a
703 marker for hydrogen. This approach has successfully detected hydrogen trapping^{285,286},
704 although quantification is not straightforward²⁸⁷ and will depend on the analysis
705 conditions^{288,289}. In order to minimise diffusion of hydrogen out of the specimen, it is also
706 necessary to keep the sample cold between charging and analysis. The study of hydrogen by
707 APT has been facilitated in the past few years by developments in specimen transfer under
708 cryogenic conditions. APT has shown that hydrogen can be trapped at grain boundaries,
709 phase boundaries, and dislocations^{120,122,123,290}, and there have been several studies
710 reporting the analysis of hydrides and their growth mechanisms^{291–293}.

711 [H2] Geology

712 While early applications of APT to geological materials were studies of metallic
713 meteorites²⁹⁴ and metamorphic magnetite crystals²⁹⁵, the technique rose to prominence
714 with its application in geochronology^{133,296}. Today, a large proportion of the published work
715 using APT on geological materials falls within geochronology, extra-terrestrial materials and
716 economic geology, but the technique is rapidly spreading into a broad range of minerals of
717 interest in chemical geology, petrology, mineralogy, and economic geology, as recently
718 reviewed in detail^{211,297}.

719 In geochronology, APT is used to investigate the nanoscale distribution of radiogenic
720 isotopes, such as uranium and lead, in accessory minerals (for example zircon, monazite,
721 titanite, baddeleyite)^{133,298,299}. The reliability of accessory minerals as geochronometers
722 relies on the assumption that trace elements diffuse negligible distances through the crystal
723 lattice. Geochronology studies reveal that deformational and metamorphic events can allow
724 the diffusion of radiogenic isotopes, leading to local changes in the isotopic ratios. The
725 diffused atoms form nanoscale clusters^{212,300,301}, and the characterisation of these clusters
726 has the potential to be used to resolve the timing of the cluster-forming geological events.
727 These studies have improved our understanding of the mechanisms for parent-daughter
728 isotopic mobility and yielded important information about the formation and evolution of
729 the Earth's crust³⁰².

730 APT has also been used to study extra-terrestrial and terrestrial proxy materials such as
731 accessory minerals from lunar soil samples and meteorites (for example ilmenite, zircon,
732 baddeleyite, nanodiamonds, and refractory metal nuggets) to resolve the timing of
733 planetary events, and better understand formation of planetary crusts^{294,303,304}. These
734 works have allowed determination of the timing of lunar transient thermal episodes³⁰⁵;

735 characterization of the composition and texture of space weathering products³⁰⁶;
736 constraining the processes driving the migration of early solar system materials in the
737 protoplanetary disk³⁰⁷; and determination of the astrophysical origins of meteorites³⁰⁸.

738 In the field of economic geology, APT has mostly been applied to ore minerals such as pyrite
739 and arsenopyrite to investigate the mechanisms of precious- and base-metal incorporation
740 and mobility^{309,310}. The characterization of metal occurrences within their host minerals
741 provides key information for understanding the fluid-rock interactions and crystal growth
742 kinetics³¹¹ that lead to the formation of viable economic deposits. Recent studies that have
743 used APT for the study of ore minerals have made significant advances towards
744 understanding the paragenesis of ore deposits including Carlin-type gold³¹², orogenic
745 gold³⁰⁹, porphyry Cu-Au³¹³, and platinum-group-alloy deposits³¹⁴.

746 Other interdisciplinary studies include investigations on the interaction between trace
747 elements, fluids and crystal defects and their effect on mineral physical properties³¹⁵,
748 characterization of the composition and structure of minerals to understand fundamental
749 processes associated to phase formation in, for example, feldspar^{304,316}, vapor-phase
750 mineral deposition, glass corrosion, and magma unmixing^{317,318}. Studies also include
751 assessment of the mechanisms for dissolution/precipitation on mineral surfaces^{319,320},
752 element diffusion during deformation, metamorphism and metasomatism used to shed light
753 on interface reactions, and mineral intergrowth and exsolution processes^{316,321–326}. These
754 insights can only be gained through APT, with its high elemental sensitivity combined with
755 three-dimensional imaging at sub-nanometre spatial resolution.

756 [H2] Beyond bulk materials

757 In addition to understanding the structure of the bulk of materials, understanding surfaces
758 is also of great interest especially, for example, for chemical conversion systems such as
759 catalysts and electrocatalysts that are involved in renewable energy generation. Most
760 devices use nanoparticles to increase their surface-to-volume ratio and maximise their
761 activity. To optimise the catalytic performance of these nanostructured materials, one must
762 have a detailed understanding of atomic-scale microstructure on surfaces, near-surfaces,
763 and where internal defects and interfaces intersect the surface. APT and related techniques
764 have long been used in surface science and catalysis based on the hypothesis that the near-
765 spherical cap at the end of the needle-shaped specimen is akin to an individual
766 nanoparticle^{327–330}. Upon adapting the specimen preparation strategies to protect
767 catalytically-active surfaces from ion- or electron-beam damage, APT can potentially provide
768 the distribution of elements within microporous and nanoporous materials such as
769 zeolites^{331,332}, metallic-organic frameworks³³³, and nanoporous metals^{334,335}. APT has also
770 been used to study the intermediate species formed at the surface of thin-film catalysts at
771 different stages of the oxygen evolution reaction^{336–338}.

772 [H2] Biological and organic materials

773 The study of organic matter with atom probe is considered an emerging application area.
774 The most significant contributions to date have been in the study of biominerals, which are
775 solid, mostly inorganic and readily amenable to atom probe analysis, as first demonstrated
776 by Gordon and Joester³³⁹. Subsequently, atom probe data has revealed the nanoscale
777 distribution of important elements such as magnesium and fluorine in dental enamel^{340,341},
778 and has added to the understanding of the nanoscale structure of the shells of marine
779 organisms^{342,343}, and various studies on apatite, bone^{344,345}, or biogenic carbonates^{346,347}.

780 Soft materials can be much more challenging to examine as they can be unstable in the high
781 vacuum environments encountered in atom probes and in FIB systems. There have been
782 several reports of the analysis of carbon-based molecules^{348–350}, polymers⁹⁸, and self-
783 assembled monolayers formed directly on metallic specimens^{99,100,351,352}. Although
784 interesting results were obtained pertaining to the field evaporation behaviour of organics,
785 much more is required to understand the intricacies of the analysis of organics by APT^{353–355}.

786 Soft biological materials are normally hydrated, but dried proteins have been deposited and
787 imaged by FIM and analysed by APT including DNA³⁵⁶, ferritin^{357–359}, and amyloid fibrils³⁶⁰.
788 Different approaches to maintain the proteins in their pristine, hydrated state include
789 freeze-drying³⁶¹, fixing in resin^{359,362}, and freezing^{125,130}. Freeze-drying removes the water
790 and greatly modifies the original structure. Fixing can be an alternative but can alter the
791 sample on a molecular level and disrupt the distribution of ionic species. In an approach
792 similar to fixation, Sundell et al.³⁶³ examined an antibody protein by using a sol-gel method
793 to embed individual proteins in an amorphous solid silica matrix, followed by a standard FIB
794 lift-out. The hydration shell around the molecule was completely replaced with silica and
795 the shape of the features in the 3D reconstructions showed good agreement with the
796 crystal structure in the protein databank. Cryogenic developments for APT specimen
797 preparation and transfer, recently reviewed by McCarroll et al.³⁶⁴, have the potential for
798 cryogenic preservation. However, more developments are needed to maintain the
799 hydration shell around proteins and determine more specifically what APT can bring to the
800 biological sciences.

801 [H1] Reproducibility and Data Deposition

802 [H2] Standards and community-led protocols

803 Although APT is fast-growing, it remains a rather immature field compared to other analysis
804 techniques. Like many relatively small communities that grew from isolated groups, defining
805 a common vocabulary across APT researchers is sometimes arduous, and using standard
806 protocols even harder. Nomenclature for the technique itself in published works over the
807 past two years only includes 3D-AP, 3-DAP, 3D-APT, TAP, LEAP, or AP microscopy, which
808 makes it confusing to outsiders and often confuses the technique with the instrument. This
809 does not happen with TEM, SEM, or SIMS. Historically, each group had its own software
810 toolbox. While sharing good practice was not prioritised, recently created geology-focused
811 groups³⁶⁵ have published the first article on how to appropriately report APT in scientific
812 publications.

813 The evolution of the commercial landscape and the fast spread of the LEAP has accelerated
814 the change towards more homogeneity with common tools and an, albeit imperfect,
815 terminology. For example, a commercial software is now used by most across the
816 community. In addition, there was a reckoning of the importance of creating active sub-
817 groups within the International Field Emission Society (IFES) to build a common structure
818 across research groups, thus the situation is evolving as we write. For instance, although
819 newly developed tools often are primarily used within individual groups, there are
820 continuous efforts to share tools that are very often open-source and document their usage
821 and application^{136,366–371} – a list is available [here](#). This is in part related to the efforts of the
822 APT Technical Committee³⁷². Another example, although in existence for over a decade, is a
823 standards committee to establish a common vocabulary and a first term compliant has been
824 submitted to ISO. For those involved, defining one term represents over 150 emails

825 exchanged over nearly two years, giving an indication of how titanic the task will be in the
826 coming years. These efforts will make **round robin experiments [G]** more common.
827 Currently there are only a few examples^{178,373} facilitating exchange of good practice and
828 comparison of results on common grounds and accelerating progress.

829 There are also critical questions that arise from performing experimental sciences that the
830 APT community has not yet fully addressed. APT is a destructive technique, so we have to
831 assume that the specimen was representative of the material, and when comparing across
832 multiple specimens from different locations in the same sample we must assume that they
833 are similar. There is a need to improve on the statistical analyses of the data and ensure
834 comparability. Experimental setups and data outputs are not yet standardized, even though
835 there are efforts in this direction coordinated by the Technical Committee of the IFES, which
836 proposed the use of an [HDF5-based file structure](#).

837 While there are currently no minimum reporting requirements when conducting and
838 reporting on APT measurements, some forays have been made in this direction by Blum et
839 al. for geological materials analysed by APT³⁶⁵, and this part of the community is pushing to
840 establish this as standard practice. It should be emphasised that generalising this practice
841 would be extremely beneficial to the community as a whole, especially considering the
842 variability in the processed data demonstrated by a recent interlaboratory experiment on a
843 reference zircon³⁷³. No public or standard location or repository currently exists for APT
844 data, even though there were early efforts by groups in, for example, Sydney³⁶⁹ and
845 Colorado³⁷⁴.

846 These are critical issues that are yet unaddressed and on which the community needs to
847 progress. For example, virtual workshops or dedicated symposia at targeted scientific
848 meetings could encourage the adoption of standard procedures and practices.

849 [H1] Limitations and optimizations

850 [H2] Spatial resolution and performance

851 APT is often presented as a microscopy technique, so naturally the question arises as to its
852 actual spatial resolution. For a conventional microscopy technique, the **Rayleigh criterion**
853 **[G]** is commonly used, and it corresponds to when the diffraction-limited image of two
854 point-sources can no longer be separated. In APT, an equivalent is not, strictly speaking,
855 accessible: the reconstructed position of the ion is subject to error but is not diffraction-
856 limited. Over the years, several criteria have been reported based on a statistical analysis of
857 the reconstructed atomic planes, either in real or reciprocal space^{188,193,194,375}. These studies
858 were performed on pure materials, with depth resolutions reported down to 20 pm³⁷⁶ for
859 aluminium or 60 pm¹⁹³ for tungsten, and lateral resolutions in the range of 200 pm, with
860 variations associated to the materials^{193,194,377} and set of atomic planes considered, as well
861 as the experimental conditions^{194,378}.

862 None of these investigations inform on the smallest feature size that can be confidently
863 analysed, which is more intimately related to the imaging process. The magnification in APT
864 is associated with the distribution of the electrostatic field at scales ranging from the
865 mesoscale to the near-atomic scale, and some aberrations are inherent to the physics of the
866 field evaporation process.

867 On the mesoscale, the magnification is related to the specimen itself⁷ and several projection
868 laws can describe the projection rather well^{137–139}. However, the parameters associated to
869 the projection differ from specimen to specimen³⁷⁹, evolve over the course of an
870 experiment³⁸⁰, and ultimately also depend on the analysis conditions³⁸¹. This is unlike most
871 other microscopy techniques. In addition, since the magnification is related to the local
872 curvature, and the field evaporation probability is dependent on the local atomic
873 neighbourhood, inhomogeneities in the specimen's composition in the near-surface region
874 lead to the development of local curvatures and changes in magnification. This is apparent
875 in experimental detector maps shown in **Error! Reference source not found.a**, in which
876 dense regions correspond to precipitates sitting at a grain boundary and within each grain,
877 as delineated by white circles and ellipses. **Error! Reference source not found.b** explains
878 schematically why the image of a precipitate on the detector can be larger or smaller
879 depending on its evaporation field relative to the surrounding matrix, which leads to
880 compression or divergence of the ion trajectories.

881 At the near-atomic scale, the distribution of the electrostatic field in the vicinity of the
882 surface is highly dependent on the neighbourhood of the departing partly-charged atom or
883 ion³⁸², which has been studied extensively by electrostatic simulations³⁸³ and density-
884 functional theory-based calculations^{384,385}. Gradients of electrostatic field can also modify
885 the path followed by the departing particle causing, for instance, short-range rolling motion
886 of atoms on their neighbours before desorbing, making the ion start its flight from a close-
887 by position to where the atom initially was inside the material.

888

889 These effects combine to blur the atomic positions following reconstruction and result in a
890 limited spatial resolution, which depends on the species considered and on the population
891 of particles that is being imaged. De Geuser et al. recently reviewed the literature
892 comparing small-angle scattering (SAS) techniques and APT on a range of precipitate-
893 strengthened alloys¹. SAS, using either X-rays or neutrons as a source, in principle has no
894 limits to the minimum size of particles that can be detected, and the size distribution
895 extracted from the fitting procedure is not affected by a spatial resolution. The reported
896 feature size by APT and SAS is plotted in **Error! Reference source not found.c**, along with, in
897 grey, the expected size for an effective spatial resolution of 0.25 nm – 1.25 nm. The actual
898 resolution is probably somewhere between these boundaries, but pinpointing to a single
899 value is problematic as it intrinsically depends on the analysed material system, and
900 particularly the size and composition of the microstructural features of interest. Ultimately,
901 the problem does not lie in the APT's capacity to detect these particles – ions from these are
902 emitted and detected – but the resolution limit implies that their size and composition can
903 no longer be directly measured.

904 Interfaces such as grain boundaries are important microstructural feature analysed by APT.
905 There have been numerous studies comparing the chemical or compositional width of an
906 interface with its structural width, which may be different. Structural discontinuity was
907 shown to introduce trajectory aberrations even without segregation^{145,146}. Correlative
908 studies with TEM performed directly on an atom probe specimen¹⁸, including at high
909 resolution¹⁹, have highlighted that the common assumption of a width of 0.5 nm is close to
910 the values obtained from experiments only for the structural width of a boundary. Recently,
911 APT measured segregation of a similar width for a transformation interface analysed along a

912 specific set of atomic planes¹⁵⁴. Yet typically, the chemical widths of grain boundaries and
913 interfaces vary even along a single boundary^{386–388}, and the limited spatial resolution of APT
914 can limit the precision of the measurement to 1 nm to 2 nm full-width-half-maximum¹⁵⁴.

915 [H2] Sensitivity and analytical performance

916 APT is a mass spectrometry technique, which naturally leads to the question of its sensitivity
917 and the precision of the measured composition. There have been efforts in recent years to
918 define statistically meaningful metrics to assess APT sensitivity²³ but they have not been
919 adopted across the entire community. Here we will only discuss some aspects affecting the
920 sensitivity without dwelling on a specific metric, to give a sense of the important parameters
921 that need to be monitored.

922 More conventional mass spectrometry techniques, such as inductively-coupled plasma mass
923 spectrometry or SIMS, typically analyse large volumes of materials of several cubic microns
924 to cubic millimetres, and hence contain billions and billions of ions. APT volumes are
925 minuscule in comparison, with ion counts in the tens to hundreds of million ions. This is a
926 first limitation when looking for trace elements (see additional discussion of limitations to
927 species detection in BOX 5). When these are agglomerated within the material, their local
928 concentration can be high enough to be detected.

929 [H2] Error estimations

930 There are numerous reports of imprecisions in the composition, and species-specific losses,
931 when comparing the overall composition from APT to that obtained from other bulk
932 techniques^{60,66}. The precision is often assumed to simply be that of the counting statistics –
933 the larger the measured number of ions N , the more precise the measurement with

934 $\sigma_i = \sqrt{\frac{C_i \times (1 - C_i)}{N}}$ where C_i is the atomic fraction of element i . When it comes to reporting
935 local composition for example in a profile, only the counting statistic is typically quoted for
936 precision. Danoix³⁸⁹ showed that the detection efficiency could be accounted for, but would
937 not change the error estimation if it is assumed to be the same across all species, which is
938 an approximation. There are also known dependencies of the measured composition on the
939 experimental conditions, which can often be traced by changes in the electrostatic
940 field^{66,67,390}. This can become crucial when considering error estimations, for example when
941 reporting on the concentration of hydrogen within materials^{289,391}.

942 As per the spatial precision, there have been reports and discussions on the possible
943 migration of atoms at the surface prior to field evaporating on a local^{46,47} scale or at the
944 mesoscale^{392,393}, as well as high-field solutes being retained on the surface while
945 neighbouring atoms from multiple lower layers field evaporate^{394,395}. However, data
946 representation continues to be based on a point cloud that implies an almost infinite
947 precision of the reconstructed position. Most composition profiles will only display errors on
948 the composition and not on the measured distance, which is problematic, particularly as
949 some of the errors can be induced by user selection of grid and delocalisation
950 parameters^{144,152}. Local changes in the field evaporation properties can also affect the
951 reported size of the imaged microstructural feature^{396,397}. While there are proposed
952 protocols to correct the distance or the data reconstruction³⁹⁸, they are scarcely used and
953 ultimately rely on another set of assumptions, and may therefore not represent a significant
954 advance after all. Ultimately, the community must accept that APT is extremely valuable

955 despite its intrinsic limitations and include these in the discussion of results, including
956 precision and accuracy.

957 [H1] Outlook

958 The outlook for the future of APT is bright, and it may be possible to finally achieve true
959 atomic-scale tomography³⁹⁹ with the ability to measure the isotopic identity and spatial
960 coordinates of every atom in a material of interest. This would require that every atom is
961 unambiguously counted and identified, with accurate and precise spatial reconstruction of
962 its original location in the specimen with true-atomic resolution. Ideally, such a method
963 would be applicable to any material that can be fashioned into a suitably-shaped specimen
964 whether solid or liquid, soft or hard, organic or inorganic. Finally, all this information should
965 be collected in large, experimentally-relevant volumes. APT would then be close to being a
966 standard reference technique for measurement of chemical composition since, in the most
967 fundamental sense, it is simply counting individual atoms. Uniquely, and unlike other
968 standardized methods used to measure chemical composition, it can provide this
969 information on highly local, arbitrarily shaped and oriented analysis volumes.

970 We are not there yet, but this lofty goal guides the technique's progress. Below, we discuss
971 recent advances in some of these areas including standards, experimental hardware, data
972 analysis, simulation, reconstruction, sample preparation and handling, and integration of
973 complementary and correlative analytical techniques.

974 [H2] Hardware

975 Some of the most exciting recent developments in hardware have come in the form of
976 either combining other instruments with the atom probe itself or integrating an atom probe
977 into working versions of other analytical tools. These can aid in specimen preparation,
978 provide additional data to inform reconstruction and analysis, give complementary
979 measurements, or provide correlative information for multi-modal microscopy studies. For
980 example, atom probe instruments have recently been successfully integrated with both
981 SEMs and FIBs⁴⁰⁰⁻⁴⁰². A start-up at the University of Stuttgart⁴⁰³ is developing a modular
982 instrument directly attached to a FIB enabling direct transfer from electron and ion imaging
983 to APT analysis. Dedicated chambers are also in development or operation to perform
984 chemical reactions at the specimen's surface and probe its response^{404,405}. In a similar vein,
985 an in-situ micro-photoluminescence bench that can be operated during atom probe analysis
986 has recently come online⁴⁰⁶ with the ability to interpret optical data through analysis of
987 three-dimensional chemical structure. Finally, there are ambitious projects underway to
988 integrate an atom probe directly into the pole gap of a TEM as a module^{407,408} or via a
989 dedicated holder following earlier designs⁴⁰⁹.

990 Similarly, there has been great progress with integration of sensitive-atmosphere specimen
991 preparation and handling capability with atom probe instruments^{364,410}. These include
992 cryogenic specimen preparation, handling, and transfer^{119,130,411,412} as well as in-situ
993 environmental treatment cells^{404,413} for performing studies related to hydrogen
994 embrittlement, phase transformations, and catalysis.

995 There have been recent developments that make use of ionizing radiation to enable new
996 pulsing paradigms. Ultrafast femtosecond-pulsed extreme ultraviolet radiation^{414,415} have
997 been used to trigger alternative field ion emission schemes such as photoexcitation or

photoionization⁴¹⁶, or to enable a more localized heating mechanism with the aim of eliminating some of the uncertainty associated with the current state-of-the-art near-ultraviolet laser pulsing. Finally, detectors that will not only detect every emitted ion but unambiguously identify its isotopic species have recently been experimentally demonstrated^{417,418}.

[H2] Revisiting FIM

FIM, the predecessor to APT, does not suffer from the spatial resolution limitations of APT since atoms are imaged prior to departing the sample surface. Indeed, aberrations occur in the early stages of the ionic flight. The magnification in FIM can still be affected by local variations in curvature, and the contrast in field-ion imaging is not yet fully understood. To date, there have been several efforts to revive FIM⁴¹⁹ as one more way to complement APT⁴⁰⁴. First, three-dimensional FIM was introduced^{420,421}, and multiple digital image processing routines developed to extract atomic positions and build atomically-resolved tomograms⁴²²⁻⁴²⁴. New imaging simulations have also been proposed⁴²⁴, aiming in part to explain the nature of the observed contrast by using density-functional theory to model the image gas ion current based on the local density of state of the surface atoms under intense electric field¹⁷⁹. To support these insights, FIM was performed within an APT and filtering routines deployed on the time-of-flight mass spectrometry data to distinguish the mass-to-charge ratio of the field evaporated surface atoms from the high background caused by the ionisation of the imaging gas¹⁷⁹. This first study on analytical-FIM demonstrated a higher spatial resolution compared to APT and the ability to image segregation at structural defects with atomic resolution. Today, analytical-FIM is in its infancy and will require hardware and software developments to make it an established technique⁴²⁵.

[H2] Moving the state-of-the-art

Much work remains to be done in the area of data analysis, simulation, and reconstruction. Most fundamentally, a better and more thorough understanding of the field evaporation process is required. Great progress has been made in recent years^{385,426} with regard to understanding the fundamental physics of evaporation and bond breaking on surfaces under high field (see BOX 7). Modelling and simulations have brought many insights and understanding of the origins and influence of aberrations on APT data^{383,427-429}, yet it is critical that more is done in the future to advance our understanding of field evaporation physics, which in part underpins aberrations.

In contrast to TEM, there are no current ways to correct trajectory aberrations by implementing or improving ion optical devices. The limit originates from the field evaporation process and the influence of the atomic neighbourhood on the evaporation field – both locally in a random solid solution and on a mesoscale for a particle in a matrix. To better understand trajectory aberrations, a full simulation of an atom probe specimen of arbitrary composition and relevant size under high applied field with appropriate timescales would require linking time-dependent density functional theory with molecular dynamics, for example. This will surely become an active area of research as computational resources continues to improve in the future.

Intensive research continues to develop new reconstruction approaches to mitigate, and maybe eventually eliminate, the effects of the assumptions and approximations that

1042 underpin the state-of-the-art algorithms⁴³⁰. For instance, given its inherent simplicity, the
1043 point-projection algorithm described above is surprisingly robust. However, its limitations
1044 are well documented and can be prohibitive for certain analyses. Some of the most
1045 promising of these algorithms use complementary simulations of how the applied electric
1046 field evolves with the shape of the specimen to predict the path of the ions to the detector
1047 and ultimately to guide their reverse-projection back into the reconstructed image^{431–434}.
1048 Correlative experimental protocols also enable the determination of the actual shape of the
1049 emitter by using scanning-probe microscopy⁴³⁵ or electron tomography⁴³⁶. Combined with
1050 shape predictions of specimens, it may be possible to retrieve the necessary information
1051 from the APT experiment alone^{381,437}. Approaches are also being pursued to derive a
1052 specimen's shape from simulations^{428,431}. These new methods demonstrate increasing
1053 capability and viability and are likely to play a significant role in the future of APT. However,
1054 for now, the reverse-projection algorithm remains by far the most implemented model and
1055 imperfect data already provides much needed information.

1056 Finally, the data streams coming out of future APT experiments will be larger and integrate
1057 signals from simultaneous microscopies and spectroscopies, providing more information
1058 than human expertise alone can analyse. The field is poised to benefit from advances in
1059 artificial intelligence for automated analyses as well as to find features buried in noisy data.
1060 Forays have already been made across the data processing workflow by using machine
1061 learning approaches for identification of cluster or phases, patterns in detector maps and
1062 peaks in mass spectra^{146,175,438–440}. An added benefit will be the improved reproducibility of
1063 data analyses, which currently suffers from the lack of established standards.

1064 [H2] Liquids & soft matter

1065 Liquids are a new frontier for APT application, as controlled field evaporation of water-ice
1066 formed directly on field emitters had been reported^{441–444} without solutions. Adineh *et al.*⁴⁴⁵
1067 have proposed an approach to encapsulate liquids on a metal tip by using graphene,
1068 preventing sublimation. Qiu *et al.*⁴⁴⁶ used this encapsulation approach to study a solution
1069 containing a gold nanoparticle. Their results suggest the robustness of this strategy. The
1070 recent development of cryogenic FIB sample preparation and cryogenic UHV sample
1071 transfer offers additional potential for encapsulation in liquid solutions with controlled
1072 freezing rates. Several groups have analysed specimens from water layers on hydrated
1073 porous nanostructured materials kept frozen via a complete cold chain through to
1074 analysis^{124–126,130}, including challenging site-specific specimen preparation by lift-out at cryo-
1075 temperature via redeposition to weld the lifted-out wedge onto the support¹³⁰.

1076 Cryogenic FIB sample preparation also helps characterise beam-sensitive, organic-containing
1077 materials¹²⁹, which leads to how APT can be applied to biological materials beyond what has
1078 been achieved so far^{340,341,361,362,447,448}. Only careful fast cryogenic freezing can retain the
1079 original biological structure in a solid state. When cooled slowly, ice will crystallize and the
1080 associated volume expansion destroys the delicate structure of biological matter. However,
1081 when cooled fast enough, water can transform directly to a metastable vitreous ice phase
1082 that preserves the specimen's original structure. Further work is required to develop
1083 workflows that allow the preparation of vitreous biological matter, such as tissue, cell
1084 components, or solutions containing biological molecules.

[H2] Closing remarks

Going forward, there must be confidence in APT's ability to provide ground truth data and make absolute measurements of specimen shape with a high degree of certainty, and of chemical composition down to individual isotopes⁴³⁹. Improvements in the fundamental understanding of the field ion emission process are required for both of those aspects, and to determine oxidation states for the detected ions³²⁰. Modelling and simulation of the evaporation process needs to mature to where it can inform reconstruction so that experimental errors can be substantially reduced and, more importantly, accurately quantified. There must be developments in the areas of handling and analysis of heretofore "impossible" materials, which include materials as simple as pure water ice. Forays into novel areas must be accompanied by the development of standards to ensure the reliability and repeatability of data acquisition and interpretation beyond what is routinely done in the field. This is necessary to communicate with industries increasingly applying APT to solve technological problems such as by performing failure analysis of memory devices or batteries.

Figure Legends

Figure 1 – Microstructural features with their typical size and some analytical techniques used to analyse them. **a** Cartoon view of the typical microstructural features of an engineering material, from the atomic arrangement of a crystal lattice through to a range of crystalline defects, phases, grains, and up to imperfections at the surface. The presence and quantification of hydrogen, specifically at these features, is a crucial challenge (modified from Dr. Jazmin Duarte, MPIE). **b** Sensitivity and analysable feature size for some materials analysis techniques.

Figure 2 – Schematics of the atom probe instrumentation and of the field evaporation process. Schematic view, not to scale, of the inside of an atom probe ultra-high vacuum analysis chamber in a straight-flight-path instrument in **a** and on a reflectron-fitted instrument in **b**. **c-d**: Schematic depiction of the field evaporation and post-ionisation/dissociation processes in the case of **c**) a metal and **d**) a less-conducting material.

Figure 3: FIB-lift-out specimen preparation. **a** Four-step protocol for FIB lift-out specimen preparation, with specimen deposited on a support. **b** Protocol adapted for in-plane lift-out of targeted features to facilitate correlative analysis by transmission electron microscopy.

Figure 4: Specimen preparation techniques for non-bulk samples. **a** Electrophoresis of nanoparticles on a pre-electropolished platinum-rhenium needle; **b** transmission-electron microscopy images show that two layers of nanoparticles were deposited by increasing the pulsed voltage from 5 kV to 7 V for 6s; **c** APT of Ag(core)-Pd(shell) nanoparticles with 1:1 and 1:3 ratios⁹². **d** Nanoparticle picked up by the micromanipulator, deposited on a Si microtip, and protected by electron-beam assisted deposition¹⁰¹. **e** Inert gas condensation formation and deposition of Au-Cu nanoparticles on Si microtips, followed by metal film deposition and FIB-milling¹⁰⁶. **f** Diluted nanoparticle solution dropped on SEM stub, then covered by Cr film and electron-beam assisted deposited Pt, and a rotated image of the wedge⁸⁹. **g** Nanoparticles encapsulated using atomic-layer deposition, followed by FIB lift-out. **h** transmission-electron microscopy image of an APT specimen containing Pt nanoparticles sandwiched between atomically-deposited layers of ZnO¹¹⁵. **i** Electrophoresis of nanoparticles on a Cu plate, and electrodeposition of Ni (insets are the Cu plate after

1129 electrophoresis and electrodeposition, respectively). **j** transmission-electron microscopy
1130 image of the APT specimen reveals Pd nanoparticles (dotted circle) were capped in the Ni
1131 matrix and **k** corresponding APT analysis⁴⁵⁰. Parts b, c reprinted from ref 117, Springer Nature Limited.
1132 Part d adapted from ref 126, CC BY 4.0 (<https://creativecommons.org/licenses/by/4.0/>). Part e adapted with
1133 permission from ref 127, ACS. Part f adapted with permission from ref 114, Elsevier. Part h adapted with
1134 permission from ref 128, Elsevier. Parts j and k adapted with permission from ref 129, Elsevier.

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1136 **Figure 5 – Ranged mass spectrum and reconstruction protocol. a-b** Segments of an APT
1137 mass spectrum from the analysis of a 17-4PH steel sample. Mass spectrum highlights the
1138 detection of different charge-states, resolution of elemental isotopes and field evaporation
1139 of molecular ions. Data courtesy of G. Yeli. **c** Simplified schematic of the ion projection from
1140 the specimen to detector in a straight flight path atom probe instrument. The colourmap
1141 corresponds to typical isopotential estimated from two-dimensional finite element methods
1142 calculations, used as an illustration. **d** Comparison between projection models on an
1143 experimental pure-Al dataset. **e** Simple schematic of procedure to reconstruct real-space
1144 depth, i.e. z-coordinate, from top to bottom: the ion detected is assumed to be projected
1145 from a hemispherical surface, and for each subsequently detected ion an additional
1146 increment is added to the z-coordinate calculation to account for all preceding ions. Part d
1147 adapted with permission from ref 176, Cambridge University Press.

1148 **Figure 6: Examples of reconstruction and data visualisation and analysis from engineering**
1149 **alloys. a** reconstructed point cloud showing the elemental distribution within a
1150 nanocrystalline Al-alloy containing both precipitates and segregation of solutes to the grain
1151 boundaries⁴⁵¹, with close-ups of a single precipitate and different families of atomic planes.
1152 **b–f** reconstruction and analysis from an additively manufactured Ni-based alloy⁴⁵²: **b** point-
1153 cloud and isoconcentration surfaces highlighting the interfaces between the γ and γ' phases
1154 (in blue) and a grain boundary where B strongly segregates (in orange) **c** Composition in a 20
1155 nm-diameter cylinder across the grain-boundary calculated along the red arrow, enabling
1156 quantification of the B segregation and the γ and γ' phase compositions. **d** Individual γ'
1157 precipitate isolated from within the point-cloud, highlighted by the pink ellipse in **b**; **e**
1158 composition profile in the form of a proximity histogram from the isosurface delineating the
1159 precipitate shown in **d**; and **f** first nearest-neighbour distribution for Al showing a deviation
1160 from the random distribution indicative of the clustering/precipitation tendency readily
1161 visible from the atom map.

1162 **Figure 7: Spatial resolution of APT in the analysis of precipitates. a** Successive detection ion
1163 maps obtained for 10^6 detected ions during the analysis of a model 7XXX-series Al-alloy;
1164 more details on the materials can be found in ref.⁴⁵³. **b** Schematic view of the compression
1165 or divergence of trajectories associated to the presence of precipitates, and associated
1166 imaging error. **c** Size of precipitates reported by APT vs small-angle scattering, highlighting
1167 the presence of an effective spatial resolution limit below which microstructural features
1168 are detected but their size and composition can no longer be directly accurately measured.
1169 Part c adapted with permission from ref 1, Elsevier.

1170 Boxes

1171 [bH1] BOX 1: Field evaporation of non-conductors

1172 The main models of the field evaporation process were developed for metals. In a solid, the
1173 electric field is effectively screened by shifting the positive and negative charges (considered
1174 collectively) so as to generate an electric field of opposite direction and same magnitude to
1175 the external field. This is referred to as the Thomas-Fermi screening length, and is typically
1176 in the range < 0.1 nm in metals, which smaller than an interatomic distance and only atoms
1177 at the very surface are subjected to the most intense field^{454,455}. For semiconductor
1178 materials this field penetration depth was found to be much larger than for metals (**Error!**
1179 **Reference source not found**). For example, Tsong et al.⁴⁵⁶ reported one of
1180 approximately 2 nm in the near surface layers of silicon. This is because the number
1181 density of electrons for semiconductors ($10^{16}/\text{cm}^3$ ⁴⁵⁴) is much lower than the one for metals
1182 ($10^{23}/\text{cm}^3$ ⁴⁵⁷) and as a result, the screening is weakened. The field penetrates into the
1183 semiconducting surface causing the electron band structure to bend upwards, an effect
1184 known as band bending^{458,459}. For insulators, these effects can be even more prominent as
1185 the penetration is deeper⁶¹. In addition, APT using HV pulsing was not typically leading to
1186 appropriate experimental conditions for analysing semiconducting and insulating materials
1187⁴⁶⁰, which became more routine with the implementation of laser pulsing capabilities.
1188 However, additional complexities arise from the interaction between the laser pulse and the
1189 electric field, in particular because the specimen has dimensions that are smaller or
1190 comparable in size to the illuminating wavelength, which makes its absorption properties
1191 non trivial^{34,461,462}. Compared to metals, the field evaporation of semi-conductors must
1192 involve additional factors such as carrier diffusion, carrier recombination, thermal diffusion,
1193 field screening effect, and band bending^{462,463}. Since the specimen is kept at a low
1194 temperature, some of these effects may be minimized.

1195 The combination of a higher field evaporation temperature and a deeper penetration of the
1196 electrostatic field has consequences on the APT analysis. A deeper penetration of the field
1197 means that the bond-breaking between surface atoms will not take place exclusively for
1198 atoms on the topmost surface, but also slightly sub-surface⁴⁶⁴. A larger population of atoms
1199 under a high electrostatic field means that not only the most protruding atoms can be field
1200 evaporated. This prevents controlled sequential removal of atoms, which lowers the spatial
1201 resolution¹⁹⁴. This also facilitates the field evaporation of molecular ions^{460,465}. The
1202 detection of a high proportion of molecular ions is common in compound semiconductors
1203 such as chalcogenides^{466,467}, nitrides³⁹⁰, and oxides⁴⁶⁸, but are less often observed in
1204 monoatomic semiconductors such as silicon and germanium. Upon emission from the
1205 surface, these metastable molecular ions can undergo dissociative
1206 fragmentation^{64,65,390,468,469} that can cause species-specific losses and degrade the spatial
1207 resolution³⁹⁰.

1208 [NRMP-20-060-Gault-b1.ai]

1209

1210 [bH1] BOX 2: Practical considerations

1211 The rate at which ions are emitted by the specimen is controlled both by the electric field
1212 and the temperature via an Arrhenius-type relationship⁴⁷⁰. To a first approximation, and in
1213 agreement with experiments⁴⁷, the electric field required to cause the emission of ions at a
1214 given rate ϕ varies linearly with temperature close to the zero-barrier evaporation field. This
1215 is summarised in the diagram in panel **a**. The region coloured in blue corresponds to a
1216 continuous network of curves for increasing evaporation rates ϕ .

1217 For a specific experiment, the user can set the base temperature T_{base} , the direct current
1218 (DC) field, F_{DC} , and the desired rates at which ions are detected, which is directly related to
1219 the evaporation rate ϕ . These are the main experimental parameters that can be adjusted
1220 to control the field evaporation process. Let us assume a thought experiment performed in
1221 conditions corresponding to the large green circle. The pulsing mechanisms are such that
1222 either the field is temporarily increased at a constant temperature, or the temperature is
1223 pulsed at a constant field. Both pulsing modes are now commonly available by using high-
1224 voltage or laser pulses, respectively^{461,471,472}.

1225 An important difference between the pulsing modes is that the amplitude and duration of
1226 the thermal pulse depend on the geometry of the specimen and the thermophysical
1227 properties of the material^{35,462,471}. This can affect the accuracy of the measured mass, i.e.
1228 the mass peak width: see for instance the blue vs. yellow mass peaks in the cartoon view of
1229 a typical section of a mass spectrum in panel **b**. Such differences in width can also be
1230 observed between an instrument with a straight flight path and one fitted with a reflectron
1231 (see **Error! Reference source not found.**). The spatial distribution of the thermal pulse
1232 across the specimen is also responsible for a change in the specimen's shape⁴⁷³ that is
1233 associated to additional compositional inaccuracies^{474,475}.

1234

1235 [bH1] BOX 3: Correlative microscopy

1236 Common correlative microscopy experimental workflows involve ex-situ experiments, using
1237 for instance X-rays, electron microscopes, or scanning probe techniques. These often
1238 analyse a separate but representative sample of the same material, and then combine these
1239 data together in the interpretation of the material behaviour. Ongoing efforts aim to
1240 directly perform APT on specimens analysed by other techniques in-situ or nearly in-situ, or
1241 use such techniques directly on an APT specimen.

1242 Although limited in resolution, the SEM part of FIB/SEM systems allows easy access, prior to
1243 the lift-out, to perform backscattered electron (BSE) imaging, electron backscattered
1244 diffraction (EBSD), or electron-channelling contrast imaging to guide site-specific
1245 preparation. Transmission-Kikuchi diffraction (TKD) to provide crystal orientation during or
1246 at the end of the specimen preparation has also become popular^{223,476-478}. High-resolution
1247 SEM imaging can provide valuable information on the geometry of the final specimen⁵³, and
1248 more precise information can be gathered by using scanning-TEM. The correlation of TEM
1249 with APT has long been undertaken^{75,479,480}, often to obtain information on the specimen's
1250 outer shape that are beneficial to guide the data reconstruction process. Nowadays, TEM is
1251 also used to obtain an accurate composition and atomic-scale crystallographic information
1252 from the same region of interest in the material under investigation.

1253 The below figure showcases the application of three different electron microscopy
1254 techniques to APT specimens. Note that the specimens were cleaned using low voltage (2
1255 kV – 5 kV) ion milling to remove the electron-beam damage and carbon build-up prior to
1256 performing APT. Panel a shows the image quality (IQ) and colour map from TKD on
1257 specimens prepared from a grain boundary region in a chalcopyrite, a promising sulphide
1258 candidate material for solar cell applications³. The two grains are represented by two
1259 colours with different crystallographic orientations sharing a high-angle grain boundary (GB,
1260 blue line) with a misorientation of 26.4° about the [110] direction. In panel b, APT reveals
1261 co-segregation of sodium and carbon along the grain boundary. Nanoscale sodium- and

1262 carbon-rich clusters are also imaged. These segregations influence the local optoelectronic
1263 properties; hence, APT measurements are critical to establish structure-property
1264 relationships^{224,226}.

1265 With respect to TEM, panel c shows a dark field transmitted electron micrograph from a
1266 **twinning** diffraction spot, that highlights a twinned region in a needle specimen prepared
1267 from an L1₀-ordered ferromagnetic MnAl alloy^{269,270}. The corresponding distribution of
1268 aluminium atoms from APT is in panel d, along with a set of the isosurfaces. A 2D
1269 compositional map further shows the manganese segregation confined to the twin
1270 boundary and depletion outside the twinned region. These segregations and twinned
1271 structure are expected to directly influence the local magnetic domain structure.

1272 Finally, a brightfield image of a needle specimen from a creep-deformed CoNi-based
1273 superalloy is displayed in panel e. The dark contrast indicates the presence of stacking
1274 faults. A high-resolution high angle annular dark field (HAADF) image from the stacking fault
1275 region reveals a change in atomic structure at the fault with respect to the surrounding
1276 lattice⁵. Panel f is the corresponding distribution of cobalt and a confined linear region
1277 highlighted by an isosurface highlighting segregation of chromium to a partial dislocation²⁶¹.
1278 The 2D elemental composition map of Al (at. %) further shows a confined depletion of
1279 aluminium along a plane that terminates exactly at the partial dislocation. This plane
1280 corresponds to the SF plane, which is chemically distinct from the associated partial
1281 dislocation. The compositional profiles along the SF plane indicate an in-plane diffusion
1282 mechanism.

1283 [NRMP-20-060-Gault-b3.ai]

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1285 [bH1] BOX 4: Calibration

1286 A number of parameters involved in the reconstruction process, such as the image
1287 compression factor ξ , are required to further refine the spatial accuracy of the image. In
1288 many cases, this optimization of reconstruction parameters is grounded by ensuring that
1289 the reconstruction accurately reflects one or more known physical prominent characteristic
1290 of the specimen across a range of length-scales. This includes partial crystallographic
1291 information such as lattice planes that are sometimes available in the reconstructed data¹⁴²,
1292 as in Figure 9. It also includes specific microstructural features such as interfaces or
1293 precipitates, the morphology of which has been previously confirmed by complementary or
1294 correlative electron microscopy^{481,482} (see BOX 3), or the overall shape of the specimen
1295 before and/or after the APT experiment, usually measured by electron microscopy^{436,483}.
1296 Other approaches, for example atomic force microscopy, have also been used⁴³⁵. Not all of
1297 these approaches are applicable in every instance, and each has their own strengths and
1298 limitations.

1299 [bH1] BOX 5: Limitations to species detection

1300 There are several factors that can limit the detection of a species. First, there is a certain
1301 level of background, as shown in red or green in BOX 3 panel b. This background is related
1302 to the dark current of the MCPs that leads to a few counts per second per square
1303 centimetre. Additionally, ions can be created at the electrostatic field and not correlated to
1304 a time pulse, either resulting from field ionisation of residual gas atoms or field emitted
1305 from the specimen's surface, as illustrated in BOX 3 panel a and its associated discussion.

1306 These uncorrelated counts form a random uniform background in the time-of-flight
1307 spectrum. Due to the conversion from time-of-flight into mass-to-charge, this appears as a
1308 decaying signal in the mass spectrum, with the background at lower masses relatively more
1309 prominent than at higher masses. The level of background depends on the analysis
1310 conditions such as vacuum level, intensity of the electrostatic field, etc.

1311 Second, the number of peaks for a specific element depends on its isotopic distribution, and
1312 the different charge states in which it is detected, and hence on its ionisation energies and
1313 on the analysis conditions. The width of the peaks is related to the precision of the
1314 measurement of the time-of-flight, voltage, and flight distance, but also to the precision of
1315 the spread in the energy of the ions in voltage pulsing mode, or of delays in the time the ion
1316 forms during the pulse in laser pulsing mode. The latter effect significantly depends on the
1317 specimen geometry and thermal conductivity^{462,471}.

1318 Third, the single-particle detector is imperfect. In principle, it is operated in a mode where
1319 incoming ions of all energies above a certain threshold (typically 2 keV) will trigger a signal
1320 of similar amplitude. This enables the detection of light and heavy ions with the same
1321 efficiency and, since the MCPs have a limited open area (approx. 50 % – 90 %), the
1322 probability to detect an ion depends only on where it lands on the detector. Yet, these
1323 settings make it impossible to use the current generation of detectors for assessing charge
1324 states, which causes overlap between ions of different elements but with the same mass to
1325 charge ratio: for example, $^{14}\text{N}^+$ and $^{28}\text{Si}^{2+}$, $^{27}\text{Al}^+$ and $^{54}\text{Fe}^{2+}$, $^{14}\text{N}_2^+$, $^{28}\text{Si}^+$, and $^{56}\text{Fe}^{2+}$, etc. In the
1326 measurement of the composition, these overlaps can be deconvoluted based on the relative
1327 isotopic abundances¹³⁶, but the specific position of ions of each species with the same mass-
1328 to-charge ratio cannot be determined.

1329 In addition, if in theory the efficiency is not dependent on the element, some elements are
1330 more prone to be detected as part of multiple events⁵⁷. This makes their loss more likely
1331 due to pile-up effects – two ions with very close times of flight land nearly at the same
1332 detector position, making it impossible for the second ion to trigger a high-enough signal to
1333 be detected. This occurrence is in part caused by the response time of the MCPs, but also by
1334 the processing of the electric signals of the delay-line detector^{44,484}. Additional losses can
1335 originate from the dissociation of molecular ions that lead to the formation of low-energy
1336 ions or neutral atoms or molecules. Depending on where the dissociative event occurs along
1337 the ion flight, the daughter ions may not be accelerated sufficiently to trigger a detectable
1338 signal³⁹⁰. Daughter ions may not acquire the energy that is necessary to make the time-of-
1339 flight of these ions close enough to other ions from the same species, making it impossible
1340 to associate them to a specific range. These aspects are typically revealed by using Saxey's
1341 approach of a correlation histogram⁵⁸. Species-specific losses have been well documented in
1342 the case of $\text{C}^{60,485,486}$, $\text{B}^{219,487}$, $\text{N}^{68,390}$, and $\text{O}^{65,468,488}$, for instance.

1343 These aspects make the balancing act between peak height and background level rather
1344 subtle. A number of statistical criteria can be used to assess whether a detected peak is
1345 statistically significantly above the level of background, locally in a range of mass-to-charge
1346 ratios. The best thought-through criteria have been introduced in ref.^{23,366}. Estimates of the
1347 sensitivity will vary from analysis to analysis, from element to element, but those reported
1348 in the literature are typically in the range of atomic part-per-million^{41,366}.

1349 [bH1] BOX 6: Advances in atom probe detectors

1350 The demands on an atom probe detector are stringent, and further developments will be
1351 required to demonstrate fast readout speed, sufficient field of view, and reliable
1352 discrimination of multiple hits and isobaric overlaps. Different approaches are currently
1353 under investigations to solve or improve these problems. For example, Bacchi et al.⁴¹⁸ used
1354 secondary electron generation by ions crossing a thin carbon foil of 20 μm ⁴⁸⁹ to generate a
1355 kinetic energy proportional signal. By carefully monitoring the signal amplitude generated
1356 by the electron cloud impinging on the subsequent MCP, the system was able to distinguish
1357 overlapping signals at 32 Da in the case of Zn and O. A second approach by Kelly and Hunt
1358⁴⁹⁰ was to adapt a superconducting detector to the requirements of APT^{491,492}. The concept
1359 behind a superconducting detector is rather similar to a delay-line detector⁴⁹³. The detector
1360 is cooled such that electrons are coupled in Cooper pairs, which underpin superconductivity
1361 in solids. The localized heating associated to the impact of the ions emitted by the APT
1362 specimen can break these pairs and generate electrons that are collected at the end of the
1363 delay-line. These approaches bear great potential to reach 100 % detection efficiency, even
1364 if substantial technical obstacles must still be solved to increase the surface area and lower
1365 dead time, which is the time during which the detector is not operational following the
1366 detection of one or more ions.

1367 [bH1] BOX 7: Theory

1368 Beyond new models for FIM contrast interpretation, new forays are being made in the
1369 theoretical understanding of the field evaporation process. An aspect of experimental
1370 sciences that can sometimes get overlooked is their underpinning theoretical aspects. This is
1371 particularly true in APT. Field evaporation mechanisms were theorised early^{45,494,495} with
1372 some advances in the following decades^{46,470,496,497}, but focused almost exclusively on
1373 metals, when the field has now fast expanded into the analysis of non-conductors. While
1374 numerical simulations have been conducted to better understand some of the artefacts
1375 observed in APT³⁸³, the simulations are concerned with the ion trajectories rather than the
1376 field evaporation process itself.

1377 Ab initio calculations have been proposed as a tool to study field evaporation effects^{65,498–}
1378⁵⁰⁰, and there are current efforts to revive this activity. The initial forays are very promising
1379 with true modelling of the field evaporation process³⁸⁵, including the small roll-up-type
1380 motion that had been predicted and confirmed experimentally⁵⁰¹. In combination with
1381 molecular dynamics, ab initio approaches have also been used to estimate the stability of
1382 molecular ions^{288,390} and complete energetics during ion flight to identify dissociation
1383 channels under the influence of the electrostatic field^{59,65,502,503}.

1384 Much theory remains to be developed, in particular in connection with experimental
1385 observations. This handshake is crucial to guide the optimisation of the experimental
1386 conditions in order to maximise data quality. As already mentioned, the ideal APT detector
1387 would detect all ions, yet as long as low-energy neutrals atoms and molecules originating
1388 from the dissociations of molecular ions are created there will be species-specific losses that
1389 make reconstructing 100 % of the data impossible. Better predictions of the stability of field
1390 evaporated species are necessary, and so are, for instance, insights into surface migrations
1391 and roll-ups that limit the spatial resolution.

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The article was a collaborative effort and, even though we tried to be inclusive of all perspectives on various aspects of APT research, it reflects our experiences and some articles likely escaped our attention. Apologies to those forgotten – it was not intentional.

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Author contributions

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Competing interests

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No competing interests to declare.

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Peer review information

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Related links

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Software tools for APT analysis: <https://tinyurl.com/APM-SoftwareList>

1422

APT-HDF5 File Specification: https://docs.google.com/document/u/2/d/e/2PACX-1vRxcJ_xF_jiNS77CoeZdQdDXD8l2BebL-DoOBkDrAsGTrkArdjLHEMCXAifBieeS8pTO9j9xnstKxs/pub

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1426 Glossary (these terms will be defined for the reader after the peer
1427 review process)

1428 **Solutes:** atoms of a species different from the main constituent atoms, which correspond to
1429 the solvent in a mixture. Solutes, often called dopants in electronic materials, are added to
1430 modify the material's properties.

1431 **Microstructural imperfections:** irregularity in the arrangement of atoms in a crystal often
1432 modifying a material's physical properties. These include lattice defects as well as inclusions
1433 of isolated or clustered foreign atoms, second phases, or particles forming in a matrix of the
1434 main constituting element (solvent).

1435 **Vacancies:** atom missing on one of the crystal lattice sites forming a point defect.

1436 **Dislocation:** linear crystal defect typically associated with the plastic deformation of a
1437 material. There are two main types of dislocations, edge and screw. A single defect can
1438 exhibit both characters in different parts along the dislocation line. Mobile (glissile) and
1439 immobile (sessile) dislocations both exist. In the case of an edge dislocation, the addition of
1440 an extra half-plane of atoms in the structure results in a compressive stress on one side of
1441 the dislocation and a tensile stress on the other. Segregation of solute elements to the
1442 dislocation help reduce the free energy associated to these defects.

1443 **Stacking fault:** local change in the stacking sequence of atomic layers in a crystal.

1444 **Twin:** two crystals with a defined crystallographic relationship with each other, formed
1445 typically by a cooperative displacement of atoms along a specific plane referred to as a twin
1446 boundary, which can be caused by plastic deformation. The organisation of atoms on either
1447 side of the twin boundary can be such that they are mirror images of each other, or follow a
1448 specific rational twin law. Twin boundaries are often considered low-energy.

1449 **Grain boundaries:** most crystalline materials are made of an ensemble of individual crystals,
1450 referred to as grains, and the junction of two crystals is called a grain boundary. The local
1451 discontinuity of the atomic arrangement makes grain boundaries loci of interest for
1452 microstructure design. Segregation of solutes typically happens to minimise the system's
1453 free energy, and grain boundaries assist with heterogeneous nucleation of secondary
1454 phases, for instance. The grain boundary energy depends on the magnitude of the change in
1455 orientation between the two grains, as well as the crystallographic plane at the junction of
1456 the two grains.

1457 **Secondary phases and phase boundaries:** solids formed by a mixture of species can adopt
1458 one or more thermodynamic phases, which can sometimes coexist. The formation of such
1459 secondary phases can be hindered by the kinetics, often associated to lattice diffusion and
1460 thermal activation. The discontinuity in the crystal lattice introduced by the presence of this
1461 second phase forms a phase boundary. The difference in the lattice unit cell can make
1462 secondary phases only partially or completely incoherent with the host lattice. Often there
1463 exist a relationship in the crystalline orientation between the matrix and of the secondary
1464 phase particle.

1465 **Composition:** relative quantity of atoms of a species with respect to all atoms of all the
1466 detected species given in atomic percent (at. %).

1467 **Polarity:** here the polarity is the electrical polarity, which is used to represent the electric
1468 positive (+) or negative (-) sign of the electrical potential at the ends of an electrical circuit.

1469 **Field ionisation:** physical phenomenon whereby atoms or molecules can be ionised because
1470 of an intense electric field.

1471 **Field evaporation:** physical phenomenon whereby atoms constituting a material can be
1472 removed in the form of ions because of an intense electric field.

1473 **Projection optic:** in microscopy, the projection is the transfer of the image of an object onto
1474 a surface through an optical system that can contain lenses or mirrors, for instance.

1475 **Time-of-flight mass spectrometer:** spectrometer that exploits the proportionality of an ion's
1476 mass-to-charge ratio with its time-of-flight from a source to a particle detector to deduce
1477 the nature of atomic or molecular ions.

1478 **Reflectron:** electrostatic mirror that can be flat or concave helping to correct spread in time-
1479 of-flight associated to energy deficits by allowing to adjust the ions' flight distance
1480 proportionally to their incoming energy.

1481 **Delay-line detector:** type of particle detector where the particle impact location on the
1482 detector's surface is deduced from the difference in the arrival time of electrical signals at
1483 the two ends of a line, i.e. a wire. Delay-line detectors typically contain two or three lines to
1484 obtain the lateral and vertical coordinate of the impact position, the signals from the third
1485 line are used to disambiguate combinations of signals coming from multiple impacts.

1486 **Molecular ion:** as opposed to an atomic ion, a molecular ion contains more than one atom
1487 and has, overall, lost one or more electrons. Molecular ions are usually metastable, but
1488 some are sufficiently long-lived to be detected.

1489 **Microtip coupon:** support for lift-out specimen preparation, typically made of silicon
1490 processed by reactive-ion and/or chemical etching.

1491 **Local electrode:** conical micro-electrode implemented on the commercial Local Electrode
1492 Atom Probe (LEAP), positioned approx. 40 microns away from the specimen, and enabling a
1493 strong localised increase in the electric field at the apex of the specimen. The
1494 implementation of such micro-electrodes enabled mounting multiple specimens at once
1495 into the instrument and analyse them in succession.

1496 **Mass peak ranging:** definition of the lower and higher mass-to-charge values of each
1497 individual mass peak in the mass spectrum to associate the mass-to-charge to one element
1498 or a combination of atoms from one or multiple elements.

1499 **Image compression:** an atom-probe specific term, it describes the angular compression
1500 associated to the ion projection; that is, the ratio of the crystallographic angle to the imaged
1501 angle.

1502 **Stereographic and quasi-stereographic projection:** model of point-projection of a sphere
1503 onto a plane, which is bijective, and preserves angles but neither distances nor areas. The
1504 standard projection has the projection point and the projection plane diametrically
1505 opposed. In a quasi-stereographic projection this is not necessarily the case.

1506 **Voxelisation:** in atom probe, this corresponds to the conversion of the three-dimensional
1507 point cloud into an array or grid of volumetric elements containing a certain number of
1508 atoms of a certain size. Following voxelisation, the number of atoms of each defined species

1509 can be used to calculate a local composition, and is usually subject to a smoothing process
1510 termed delocalisation.

1511 **Isosurface**: three-dimensional surface representing points of a given threshold value of
1512 composition, concentration, or density within the 3D point cloud. The isosurface is built
1513 from the grid of voxels and hence subject to the delocalisation.

1514 **Iso-Concentration**: a concentration reports a quantity per unit volume expressed in at.nm^{-3} ,
1515 for instance, and is equivalent to a density. Due to trajectory aberrations and reconstruction
1516 issues, volume estimations from atom probe are typically not precise.

1517 **Interfacial excess**: the number of excess atoms of a certain species per unit area of an
1518 interface.

1519 Round robin experiments:

1520 **Rayleigh criterion**: shortest distance below which the diffraction-limited image of two point-
1521 sources can no longer be separated.

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ToC

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This Primer on atom probe tomography introduces the fundamentals of the technique and its experimental setup, describes recent developments in specimen preparation, highlights aspects of data reconstruction and analysis, and showcases various applications of atom probe tomography in the materials sciences, geosciences and biological sciences.

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