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MICROMETROLOGY IN PURSUIT OF QUANTUM RADIATION STANDARDS

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Abstract – With the recent redefinition of the SI base units in terms of constants of nature, the race is on to maximize achievable precision by developing primary standards based on quantum metrology, thereby to realize the Quantum SI. For the becquerel (Bq) and gray (Gy), the derived SI units for activity and absorbed dose, respectively, this is especially challenging because production and detection of ionizing radiation is difficult to manage at the atomic scale, and quantum coherences are easily lost in the cascade of inelastic processes that accompany the slowing of energetic particles in matter. This would not preclude a secondary role for quantum metrology in the pursuit of single-event detection and resolution of radiation effects at microscopic scales, where we are beginning to apply new transition-edge sensors, silicon photonics, and link to quantum electrical standards. Here we summarize recent results for microcalorimetry in activity and dosimetry measurements and discuss their potential as next-generation standards.

Keywords: radioactivity, dosimetry, metrology, decay energy spectrometry, calorimetry, transition edge sensors

1. INTRODUCTION

The goal to make the SI traceable to constants of nature finally was achieved a couple of years ago after a prolonged effort to eliminate physical artifacts from the calibration chain for each of the base units. An important co-development is the practical implementation of primary standards based on quantum effects, like entanglement and squeezing, which carry the prospect of baseline precision at the Heisenberg limit. Independent of these exciting developments are recent advances to make chip-scale standards that are field-deployable, which if based on quantum metrology would culminate in zero-chain traceability of measurements to constants of nature with quantum precision. How all this may unfold in the realization of each of the various units of the SI will depend on the degree to which quantum metrology can be tailored to meet diverse measurement needs.

Applications in ionizing radiation pose a unique challenge and opportunity for the development of relevant quantum metrology as well as the articulation of calibration needs that might exploit the potential of quantum metrology for measuring radiation interactions at the atomic scale. However, any attempt to marry quantum metrology with radiation physics must overcome two big obstacles: the fundamental stochasticity that governs radiation processes like nuclear decay, production of photons and secondary electrons in ionizing collisions and the like, and the corresponding energy transfers in those interactions, which are several orders of magnitude above the spectrum of elementary excitations exploited in quantum metrology. If such obstacles seem insurmountable, the unlikely suitors may nevertheless be coaxed along by exigencies of industry in pursuit of zero-chain traceability for radiation metrology and measurements at the scale of nanofabricated devices and biological cells.

At NIST, industry needs have spawned projects to provide traceable metrology for activity and dose at those microscopic scales. While none of these projects yet encroaches on territory in radiation physics where quantum coherences prevail, they do present opportunities for use of quantum metrology to improve measurement resolution that may eventually yield quantum standards to realize SI units of activity and dose.

2. OVERVIEW OF RADIATION STANDARDS

Present and historic realizations of the SI for ionizing radiation have most-often used detectors based on ionization of gas, semiconductor excitation, liquid scintillation, or calorimetry.[1, 2] These systems are used to realize the becquerel and the gray with relative combined standard uncertainties of one to a few parts in 10^3.

For activity, non-calorimetric realization of the SI unit for activity of a radionuclide, becquerel (s^{-1}), is done in counting mode – that is, counting individual decay events. Key challenges to metrology are identifying the nuclide that decayed, determining the efficiency of the counter, and accounting for dead-time and spurious counts. These challenges are often approached by using multiple detectors of one or more type, either separately for spectroscopic identification of radionuclides and/or together in a coincidence-mode to determine counting efficiency[3, 4]. To date, realizations of the becquerel using calorimetry have been limited to measuring average energy of high-activity sources, rather than the heat signature of individual decay events. This is typically done through isothermal calorimetry, where the major concerns are with heat defect (e.g. chemical reactions) and in knowing the average decay energy from other methods[5].

An important consideration is that in radionuclide metrology, the measurand is often the massic activity (Bq/g) of a solution. An ancillary requirement for activity standards is to know the mass of the aliquot being measured, such that one can infer the activity of other samples of the same material. These measurements of small aqueous masses require special techniques to overcome limitations of evaporation, balance stability, and source morphology.[6]

For dosimetry, realization of the gray (J/kg) is typically achieved with ionization chambers or water calorimeters in a Co-60 reference beam. In both cases, the dose is averaged over a volume typically on the order of 1 mL. The former is operated in an average current mode and the major considerations are knowing the average energy per electron-ion pair created and determining the effective mass of the ionization region. In water calorimetry, the major considerations are heat defect due to radiolytic reactions of water, thermal perturbations due to non-water materials near the point of measurement and heat transport[7].

An important additional consideration for standard reference dosimetry is uniformity of the dose field over the detection volume. This contrasts with many applications in medicine and manufacturing, which are increasingly making use of dose fields with very sharp gradients to deliver large doses with high spatial selectivity to desired targets. This is critically important in applications like external-beam radiotherapy, where it is desirable to deliver dose to irregular tumor volumes while sparing surrounding healthy tissue, or in sterilization of surgical instruments, where it is desirable to kill bacteria on surfaces without damaging the underlying material. For such applications, dose distributions may change appreciably over a span of 50 μm within an irradiated medium. Current primary standards are not suitable for calibrating dosimeters for use in such fields, and traceability becomes problematic.

However, with sufficient miniaturization of radiation metrology, it may become possible to realize local beam uniformity required for absolute dosimetry in these non-standard fields and thereby create a pathway for establishing direct traceability. It might also begin to expose microscopic spatial variations in dose, due to track physics and related quantum effects, that present opportunities to apply quantum metrology.

2.1. Recent Developments

2.1.1 Radionuclide Activity

The rapidly developing field of quantum information science has driven innovation in superconducting devices and supported advancements in related sensor technology and user-friendly cryogenic systems. Ultra-cold sensors are not only of interest for radiation measurements due to their lower thermal noise, but in that they enable novel sensor platforms. One such platform is metal-foil Transition Edge Sensors (TES). These sensors typically operate at ≈ 100 mK. Thus, for radiation sensing they have advantages of both high gain at the transition temperature and low thermal noise. As such, TES have reached resolving powers $> 3 \cdot 10^3$ [8]. For measuring radioactive decay, TES have been used for Decay Energy Spectrometry (DES) in which a radionuclide sample is embedded in a metal or superconducting foil, which acts as an energy absorber and is thermally connected to a TES. In this way, the TES-DES system can theoretically achieve 4π solid angle, similar to liquid scintillation, but with a factor of 1000 improvement in resolving power. For α -decay radionuclides, 100 % quantum efficiency is possible and if secondary x-ray or γ-rays are absorbed, the DES spectrum converges to a single peak at the Q-value (difference in rest mass between parent and progeny). A milestone experiment was the measurement of the isotopic ratio of Pu-239 and Pu-240 by DES, which agreed well with mass spectrometery results [9].

Efforts are underway to realize the Bq using TES-DES. This amounts to calorimetry but is a radical departure from classic radionuclide calorimetry or even "cryogenic microcalorimetry" (which was microwatt calorimetry at 8 K)[10]. Here, instead of measuring average power from a "hot" source (typically several gigabecquerels of activity), the radioactive decay products from each single decay event are thermalized in a small absorber and that individual heat pulse (~ 1 pJ) counted and measured. This requires numerous innovations. First the TES must be made fast enough to operate at necessary count rates without introducing undue pulse pileup or dead-time, while having a large enough absorber to stop the decay products. This is achievable through optimal design of the detector system, operated in electro-thermal feedback mode. Figure 1 shows a Geant4 Monte Carlo simulation illustrating the effect of Au absorber thickness on DES spectral shape. For a thin (15 μm) absorber, a significant secondary peak due to escape of the 59 keV γ-ray is present. Increasing the thickness to 1000 μm eliminates the escape peak, resulting in a single peak at the Q-value, while somewhat degrading the energy resolution. Here, the energy resolution at 15 μm for a TES operating at a temperature of T = 120 mK was taken from Hoover et al [9]. For the thicker foil, thermal noise $\approx \sqrt{kT^2C}$ for the additional Au mass was added in quadrature, where k is the Boltzmann constant and C is the heat capacity. By adequately capturing all the energy of each decay event, realization of the becquerel becomes a "simple" matter of counting the decays in the Q-value peak over a known period. In practice, the usual considerations of dead-time and pulse-pileup need to be considered[11].

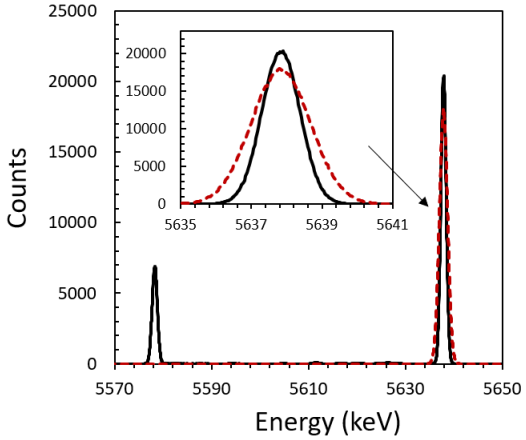

Fig. 1. Geant4 Monte Carlo simulation of Am-241 DES spectra for Au absorber of thickness 15 μm (solid black) or 1000 μm (dotted red). In the latter case, the γ-ray escape peak is absent and the activity is equal to the count rate in the peak located at the Q-value (right

peak). Inset shows degradation in energy resolution due to increased thermal mass for thicker foil.

To measure massic activity, the gravimetric source preparation must also be improved. Because TES energy resolution is ultimately limited by thermal mass, the absorber foils are typically only a few millimeters across. This limits the amount of aqueous solution that can be deposited to about 1 μL (1 mg). Dispensing 1 mg of solution with the desired relative uncertainty of less than 0.1 % is a significant challenge. Recent work has demonstrated 0.3 % uncertainty using a manual method [12] and ongoing research leveraging the redefinition of the kilogram in the SI to realize mass by electrostatic force has potential to reduce that uncertainty by an order of magnitude [13].

Decay Energy Spectrometry provides a new tool to study the interaction between ionizing radiation and matter. Many ionizing radiation measurements in radioactivity as well as dosimetry rely on radiation interaction models to interpret and make corrections to experimental results. These models are generally well-understood at higher energies, but accuracy rapidly degrades below 100 keV, and the models stop at 100 eV. To understand the interactions and potential damage to these systems requires extending these models 5 orders of magnitude to 10 meV. Doing so would not only enable design of next-generation DES sensors, but be a tool to study quantum decoherence effects due to ionizing radiation that would adversely impact development of fault-tolerant quantum computers [14, 15].

2.1.2 Radiation Dosimetry

Similar to the field of radioactivity, in dosimetry there is also a push towards smaller calorimeters. However, dosimetry applications are not amenable to ultra-low temperature measurements of single events. There are two major reasons for this. First, in typical dosimetry applications, the flux of incoming energetic particles is too high for a TES to resolve in time. Secondly, unlike radioactivity, in which an aliquot of a radioactive sample can be taken from a material of interest and embedded into a detector, which is then cooled, in dosimetry, the need is to calibrate beams of radiation at room temperature.

Nonetheless, the move by industry to low energy X-ray and electron beams that only penetrate < 100 µm into material as well as non-uniform higher energy beams, similarly motivates research into micron-scale calorimeters. At NIST, we are developing chip-scale dosimeters based on photonic resonator devices with the goal of resolving spatial variations of absorbed dose at the level of individual components on a silicon wafer or bacteria. The photonic platform uses telecommunications-wavelength components, making it cost-effective. Our studies on photonic thermometers have demonstrated radiation-insensitive thermo-optic coefficients, making them suitable for calorimetry [16, 17].

We have demonstrated calorimetry using a 1.8 MeV electron beam of current about 2 µA pulsed with a duty cycle of 50 % and a half-period of 30 s, as shown in Fig. 2. The beam was incident on a chip containing a photonic crystal cavity of dimensions 0.7 µm × 30 µm × 0.22 µm, having resonant wavelength of ≈ 1550 nm. The chip was exposed to multiple 30 s exposures at fixed beam current, which was varied from 0.25 mA to 5 mA (representative waveforms shown in Figure 2). Resonance wavelength was monitored with a wavemeter and scanning laser. The photonics apparatus is described in [16]. The resonant wavelength shift $\Delta\lambda$ during irradiation cycles was determined by extrapolating pre- and post-irradiation drift segments to the midpoint of the irradiation cycle, and measuring the difference.

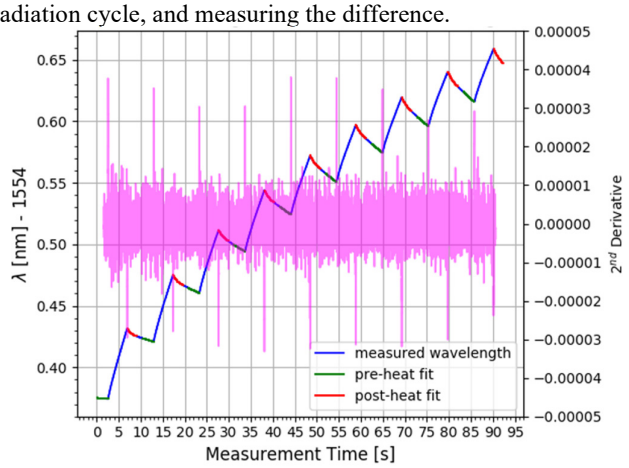

Fig 2. Resonance wavelength (λ) of photonic calorimeter sensor during irradiation cycles. Differential heating was determined by extrapolating pre-heating (green) and post-heating (red) data to midpoints, determined by 2nd derivative (pink).

The average $\Delta\lambda$ for 9 cycles was 73.3 pm with a standard deviation of 1.2 %. Absorbed dose was determined from $\Delta\lambda$, the known opto-thermal coefficient of the device, and the heat capacity of Si. The absorbed dose was validated using standard alanine dosimeters located at the same vertical distance from the beam entrance as the chip. The average absorbed dose measured with the alanine pellets will in general be different from that measured by the silicon sensor due to the dose gradient within the thicker alanine pellet, and the difference between the alanine and silicon stopping powers. In order to determine the alanine-to-sensor correction factor, a Monte Carlo computation of the experimental apparatus was performed. The Monte Carlo results show that the average absorbed dose to the alanine pellets only differs from the silicon sensor by about 4 %, despite the large dose gradient and stopping power difference. Alanine and chip runs were normalized by the electron current in a Faraday cup. The dose-to-charge ratio measured for the photonic device was on average 1.94 % higher than that for alanine. Although these results are preliminary, and a full uncertainty assessment is still underway, the agreement is within 2 × the between-cycle standard deviation for the photonic device at a single beam current, which is encouraging.

These photonic calorimeters have the potential to serve as calibrators for on-chip dosimeters (photonic or otherwise). Although they would not be quantum standards, they could be calibrated traceable to NIST temperature standards and thus enable SI-traceable realization of the gray in the field.

3. CONCLUSIONS

The calorimeters described here represent shrinking of spatial scales by more than a factor of 1000 from classical approaches. These technologies enable new measurement capabilities, such as characterization of individual radioactive decay events with 100 % quantum efficiency and exquisite energy resolution and absolute micrometer-scale dosimetry of

industrial electron beams. Further, the low noise floor and high energy resolution of DES make this a tool for extending the knowledge of radiation interactions at low energies while photonic calorimeters can readily be fabricated into arrays to study hard-to-approach questions in dosimetry at material interfaces and within microscopic structures.

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