

Dielectric constant measurement sensitivity in electrostatic force and force gradient microscopy-based modes

Gheorghe Stan

Material Measurement Laboratory, National Institute of Standards and Technology, Gaithersburg, MD
20899, USA

Understanding the nanoscale electrostatic interaction between a conductive atomic force microscopy (AFM) probe and a dielectric film is central to the operation of various nanoscale dielectric microscopies and determination of dielectric properties of the film. There is no simple analytical description of the electrostatic interaction generated in the confined probe-sample geometry for neither the static nor dynamic AFM modes used in dielectric measurements. An accurate description of the involved physics is obtained only by means of a finite element analysis modeling of the system. However, the alternative to using numerical analysis is not very popular due to being slower and requiring relatively high computation resources. In this work we revised the contributions from different parts of the AFM probe to the probe-sample capacitance by both analytical and numerical methods. We tried to reconcile the two approaches and observed the differences as a function of geometry and material parameters. Under various noise levels, the efficiency of an analytical model was tested against finite element analysis that captures in detail the electrostatic interaction in AFM-based dielectric measurements. The investigation was performed in both spectroscopic force-distance curves and constant height scans with measurements for the deflection and frequency of the AFM probe. The obtained measurement sensitivities are relevant in selecting the optimal scanning mode and its operational parameters for a given film thicknesses and dielectric constants but also show the critical role of numerical analysis to the correct interpretation of the measurements.

Key words: nanoscale dielectric measurements; electrostatic force microscopy; finite element analysis

Gheorghe Stan – Material Measurement Laboratory, National Institute of Standards and Technology, Gaithersburg, Maryland 20899, United States; <https://orcid.org/0000-0001-5773-0328>; Email: gheorghe.stan@nist.gov

I. Introduction

A multitude of Atomic Force Microscopy (AFM) based techniques has emerged in the last decades for material property characterizations aiming to advance scientific endeavors and technological applications at the nanoscale. These include measurements for mechanical, electrical, chemical, and electro-optical properties that either individually, or in combination with each other, can provide a very localized characterization of the structure-property relationship of materials, structures, and devices of interest. Among them, several AFM techniques like electrostatic force microscopy (EFM) [1, 2, 3, 4, 5, 6], scanning capacitance microscopy (SCM) [7, 8, 9], nanoscale impedance microscopy (NIM) [10, 11], scanning microwave microscopy (SMIM) [12, 13], and scanning nonlinear dielectric microscopy (SNDM) [14, 15] are used for quantitative measurements of the local dielectric properties of dielectric materials and structures. The capabilities of these techniques to extract and quantify the dielectric contrast from the measured response were tested on common dielectric films [3, 5], fabricated nanostructures (nanotubes, nanowires, nanoparticles, embedded circuits) [13, 16, 17, 18], nanocomposites [19, 20], and organic materials (membranes, cells, viruses) [9, 21, 22]. As each method matures and extends its range of applicability, more complex modeling can be used to improve quantitative outputs.

Simple analytical models are usually preferred to interpret the measurements in converting the qualitative contrast into quantitative parameters. For example, in earlier dielectric EFM-based measurements, the approach was to consider only the main contribution to the electrostatic AFM probe-sample interaction, namely the one coming from the apex of the tip [8, 23]. Gradually, more complex shapes and geometries were considered to represent the actual AFM probe in modeling the surrounding electric field and induced film polarization. Among others, the AFM probe was approximated as a spherical apex with circular cone [24, 25], a spherical apex with a circular cone and crown disk on top of the cone [24, 26], a spherical apex with double angle cones [27], etc. In general, the analytical models used for dielectric EFM analysis rely on expressions derived for the capacitive interaction between conductive AFM probes and metallic substrates [28, 29, 30] and equivalent charge method [31, 33, 34]. Their applicability to dielectric EFM was confirmed in the limit of thin films and low dielectric constants through finite element modeling and recognized to be limited in the range of thick film [24]. This is because both the analytics and numerical modeling favor simplified versions of the AFM probe representation with 2D axial symmetry, omission of or limited modeling of the cantilever contribution, and no tilt of the probe. A full 3D modeling is needed once the tilt of the probe is included and factored to the probe-sample interaction [35]. The complexity associated with 3D numerical modeling is due to the multiscale contributions of various parts of the AFM probe to the electrostatic field going from few nanometers (apex) to tens of micrometers (cone) and hundreds of micrometers (cantilever).

In this work, we investigate the applicability limits of an analytical model in terms of dielectric constant, film thickness, probe-film separation, and noise. The analysis was performed against data provided by a full 3D modeling that realistically reproduces the size, shape, and orientation of a conductive AFM probe in EFM force-distance curve measurements. With integral 3D modeling we capture the actual electric potential distribution around the AFM probe as coming cohesively from all the parts (apex, cone, cantilever) rather than a superposition of independent contributions. With individual contributions it is not feasible to calculate the force and force gradient experienced by the cantilever during its bending under the electrostatic probe-sample interaction. To address that, the 3D finite element modeling performed in this work provided both the deflection and resonance frequency of the cantilever from the electromechanical response of the system. These can be directly related to either static or dynamic EFM measurements for dielectric characterizations.

The analysis was restricted to basic cases of dielectric EFM measurements under applied dc bias voltages in the form of force-distance ramps and constant height measurements. These configurations offer the most direct test vehicles for comparison to analytical models and correspond with many of the implemented measurement modes. Based on these configurations, we determine the measurement sensitivity as a function of tip-film separation and dielectric constant. The most explicit response is retrieved from force-distance ramps, on which analytical expressions can be tested to observe their applicability for the correct interpretation of the electrostatic tip-sample interaction. The analytical model used for comparison was the one derived from the capacitive interaction between conductive AFM probes and metallic substrates [28, 29, 30] due to its direct solution for the dielectric constant as a fit parameter. On the other hand, the model based on the equivalent charge method [31, 33, 34] was not examined here because that model works only for a geometry with axial symmetry (*i.e.*, the cantilever and its tilt are not included) and the fit procedure to extract the dielectric constant from expressions of the force or force-gradient are more complex.

In probing the accuracy of the analytics against modeling we also look at the effect of noise. Noise is inherent in measurements and, due to the high sensitivity of analysis to the convoluted dependence of the dielectric constant and geometrical parameters like tip radius and film thickness, the results can be significantly affected by noise level. In constant height measurements like those used in mapping, we evaluated the dielectric constant measurement sensitivity to common noise levels for several configurations. The results point to selections that can be done in terms of measurement configurations that optimize measurement sensitivity over a large range of dielectric constant values.

II. Dielectric Constant from Electrostatic Force Microscopy

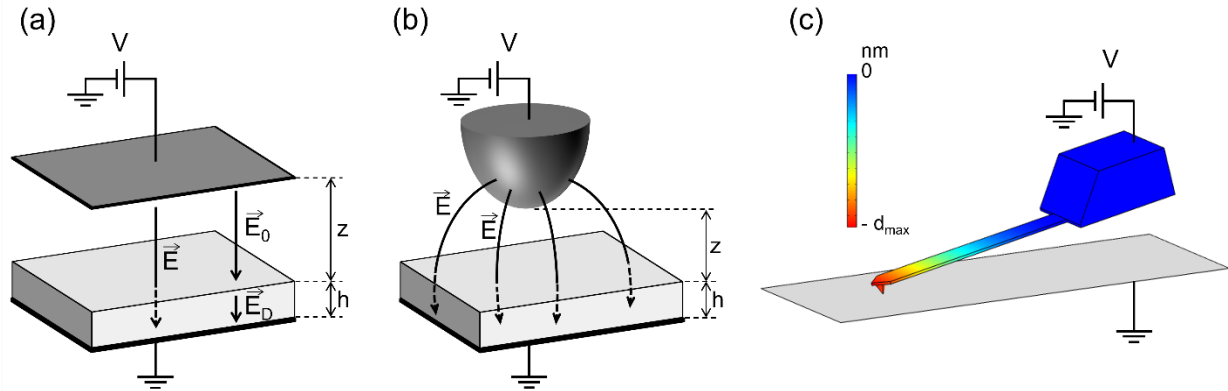


Figure 1. a) The electric field inside of a biased parallel-plate capacitor that is partially filled with a dielectric slab. b) The electric field between a conductive paraboloid and a dielectric film on top of a metallic flat substrate. c) The EFM configuration with a conductive AFM probe above a dielectric film on a metallic flat substrate. The AFM probe is tilted in the plane normal to the surface of the film. The color code shows the bending of the AFM cantilever under the electrostatic attraction between the AFM probe and the metallic substrate under an applied voltage.

The dielectric EFM configuration resembles that of a parallel-plate capacitor with a dielectric slab between its plates as shown in Figure 1a. In response to the voltage V applied on the metallic plates of the capacitor, the dielectric material develops a polarization field that reduces the strength of the electric field from $\vec{E}_0$ (in the empty space) to $\vec{E}_D = \vec{E}_0/k$ (within the dielectric), where k is the dielectric constant of the dielectric material (assuming here linear dielectric materials). It can be readily calculated that the capacitance C of the parallel-plate capacitor partially filled with a dielectric slab is that of two capacitors in series, one with the dielectric material and the other without,

$$C = \frac{\epsilon_0 A}{z+h/k} \quad (1)$$

where z is the separation between the top capacitor plate and the top surface of the dielectric slab (*i.e.*, the thickness of the empty space), h is the thickness of dielectric slab, A is the area of each of the capacitor plates, and ϵ_0 is the vacuum permittivity. However, the EFM configuration substantially deviates from this simple geometry with the top plate replaced by the paraboloid shape of the AFM tip, as shown in Figure 1b. Therefore, as expected, the expression given by (1) becomes a crude approximation. This is because the lines of the electric field generated between the tip and the metallic substrate are no longer straight and the induced polarization is not uniform as a function of position and across the film thickness. For EFM analysis we need to include these nonlinearities to the calculation of the net electric field or, equivalently, the electrostatic force between the two electrodes.

The electrostatic force F developed between a conductive AFM probe and a sample under an applied voltage V is proportional to the gradient of capacitance and the square of the voltage:

$$F = \frac{1}{2} \frac{\partial C}{\partial z} V^2. \quad (2)$$

While a simple expression for the force can be derived from (1) and (2) for the case of the parallel-plate capacitor in Figure 1a, a more complex derivation can be envisioned for the EFM configuration in Figure 1b. A starting point is that of the analytical model proposed by Hudelt *et al.* [28] for the capacitive force between a metallic paraboloid and a metallic surface (without any dielectric in between). In their model, the net force was calculated by integrating the contributions from infinitesimal regions of the tip and surface. These infinitesimal contributions are assumed to be like those of infinite dihedral plate capacitors. There are two contributions to the total force, one between the spherical apex of the tip and the metal substrate

$$F_{\text{Apex-M}}(z) = \pi \epsilon_0 V^2 \frac{R^2(1-\sin \theta)}{z[z+R(1-\sin \theta)]} \quad (3)$$

and one between the conical part of the tip and the metal substrate

$$F_{\text{Cone-M}}(z) = \frac{\pi \epsilon_0 V^2}{[\ln(\tan(\theta/2))]^2} \left[\ln \frac{H}{z+R(1-\sin \theta)} - 1 + \frac{R(\cos \theta)^2 / \sin \theta}{z+R(1-\sin \theta)} \right] \quad (4)$$

where z is the distance from the spherical apex to the substrate (same as in Figure 1b but without the dielectric), R is the radius of the tip apex, θ is the half angle of the cone, and H is the height of the tip (spherical apex and cone). It should be noted that the above expressions are derived for the axial symmetry around the paraboloid shape of the tip that is normally positioned above an infinite metallic substrate. A third contribution $F_{\text{Cantilever-M}}$ to the total probe-sample electrostatic interaction comes from the cantilever

of the AFM probe. A simple expression for that is obtained for a tilted and rigid rectangular cantilever that is part of a dihedral plane capacitor [29]

$$F_{\text{Cantilever-M}}(z) = \frac{\varepsilon_0 V^2}{2} \frac{wL(\sin \alpha)^2}{\alpha^2} \frac{1}{(z+H \cos \alpha)(z+H \cos \alpha+L \sin \alpha)} \quad (5)$$

with α being the tilt angle and L and w the length and width of the cantilever, respectively. A more complicated expression for $F_{\text{Cantilever-M}}$ is given in [30], where the bending of the cantilever under the action of the distributed electrostatic interaction along the cantilever was considered.

The expressions of the capacitive forces given by (3) and (4) were extended to the case when a dielectric film is present between an AFM tip and a metallic substrate by considering the screening effect of a dielectric material to the electric field of a capacitor described by (1). This is done by simply replacing the distance z by $z + h/k$, with z now being the distance from the apex to the top surface of the dielectric film and h the thickness of the dielectric film, as shown in Figure 1b. The extension was proposed first by considering only the apex-sample interaction [8, 23, 31, 36, 3]

$$F_{\text{Apex-D}}(z) = \pi \varepsilon_0 V^2 \frac{R^2(1-\sin \theta)}{(z+h/k)[(z+h/k)+R(1-\sin \theta)]} \quad (6)$$

to be the dominant contributor to the EFM measurements and later the cone contribution was added into the model analysis [24]

$$F_{\text{Cone-D}}(z) = \frac{\pi \varepsilon_0 V^2}{[\ln(\tan(\theta/2))]^2} \left[\ln \frac{H}{(z+h/k)+R(1-\sin \theta)} - 1 + \frac{R(\cos \theta)^2 / \sin \theta}{(z+h/k)+R(1-\sin \theta)} \right]. \quad (7)$$

Similarly, the expression given by (5) for $F_{\text{Cantilever-M}}$ can be extended to the case when a dielectric film is present to get $F_{\text{Cantilever-D}}$. The total probe-sample capacitive force is assumed to be a superposition response with contributions from the individual parts, $F_{\text{Probe-D}} = F_{\text{Apex-D}} + F_{\text{Cone-D}} + F_{\text{Cantilever-D}}$, and used to analyze the measurements to determine the dielectric constant, while all the other parameters are known. There are many works where each of these three individual contributions was examined and tried against various analytical expressions [8, 24, 32, 33]. In particular, it was found [8] that $F_{\text{Cantilever-D}}$ varies very little over the tip-sample separations used in electrostatic measurements for dielectric constants. Considering the contributions $F_{\text{Cantilever-D}} = F_0$ as constant, the $F_{\text{Probe-D}}$ becomes

$$F_{\text{Probe-D}}(z) = F_0 + \pi \varepsilon_0 V^2 \frac{R^2(1-\sin \theta)}{(z+\frac{h}{k})[(z+\frac{h}{k})+R(1-\sin \theta)]} + \frac{\pi \varepsilon_0 V^2}{[\ln(\tan(\theta/2))]^2} \left[\ln \frac{H}{(z+h/k)+R(1-\sin \theta)} - 1 + \frac{R(\cos \theta)^2 / \sin \theta}{(z+h/k)+R(1-\sin \theta)} \right]. \quad (8)$$

In practice, the constant F_0 can be eliminated through a differential analysis by reporting $F_{\text{Probe-D}}(z)$ at any given tip-film separation z to a $F_{\text{Probe-D}}(z_{\text{max}}) \cong F_0$ that is measured at a large distance $z = z_{\text{max}}$ [5, 8]. Furthermore, some of the discrepancies attributed to the approximative description provided by (8) when applied to an actual AFM probe are mitigated by considering the tip radius R as a fit parameter that can be obtained from measurements over a metallic substrate (*i.e.*, $h = 0$ in equation (8)) [24]. In the subsequent analysis presented in this work, we will use the integral form of (8) with both F_0 and R as fit parameters extracted from measurements over a metallic substrate rather than the difference $F_{\text{Probe-D}}(z) - F_{\text{Probe-D}}(z_{\text{max}})$ as in the aforementioned differential analysis. This is because the determination of F_0 from measurements at a given large tip-film separation was found to be affected by the inherent noise of the measurements, which in turn added to the uncertainty of the resolved dielectric constant of the films.

The entire AFM probe (including the base of the cantilever) from the numerical analysis, which will be discussed in detail later, is shown in Figure 1c. Here we have a conductive AFM probe, tilted with respect to the sample, and with the tip apex brought in proximity to the sample. The sample is a dielectric film, tens of nanometers thick, on top of a metallic electrode. Under an applied bias voltage, the electrostatic interaction brings the tip further down to the sample as the cantilever is free to bend (refer to the color code showing the mechanical bending of the cantilever under bias in Figure 1c). Thus, it is desirable to include the cantilever in the EFM analysis as the cantilever contributes to both the static configuration and electrostatic response of the probe. However, these additional contributions that include the detailed shape, tilt, and bending of the cantilever are difficult, if not impossible, to capture in an analytical model that treats the AFM probe as sum of different parts of simpler geometry [33]. They can be integrally addressed, however, in an electro-mechanical numerical analysis that models the entire body (apex, cone, and cantilever) as shown in Figure 1c. Besides the static response of the cantilever, another benefit of the numerical modeling is the characterization of the cantilever's dynamic parameters such as resonance frequency and phase. Similar to the case of static EFM, a dynamic analysis for an AFM probe subjected to electrostatic interaction becomes very complex analytically once the above requirements are to be included. Therefore, the numerical model is preferable. In the following discussion of the EFM modeling, the static EFM will be analyzed in terms of force that relates to the cantilever deflection (bending) and the dynamic EFM in terms of force-gradient that relates to the change in the resonance frequency of the cantilever. Alternatively, as suggested by the relationship given by (2) and adopted in many previous works [3, 5, 8, 23, 31, 36, 24], the analysis for dielectric EFM can be performed in terms of the capacitance gradient $\partial C / \partial z$ for force and $\partial^2 C / \partial z^2$ for force-gradient measurements. In this work, the response analysis for dielectric EFM was observed in terms of force and resonance frequency of the cantilever, which are easier to relate to the operational configuration of the AFM.

III. Finite Element Analysis for 3D Dielectric EFM

We used the AC/DC and Structural Mechanics modules of the COMSOL Multiphysics 6.0 software (COMSOL, Inc., Burlington, MA) to model the three-dimensional (3D) electrostatic interaction between a conductive AFM probe and a dielectric film supported by a metallic substrate. The probe-sample system is highlighted in Figure 1c, with the AFM probe made of a fixed base, cantilever, cone, and spherical apex above a dielectric film. The AFM probe is tilted at 11° in a vertical plane that is normal to the surface of the film, as in the actual AFM configuration. The Structural Mechanics module provided the simulation for the bending of the cantilever under the attractive electrostatic force. The AC/DC module provided the calculation of the electric field generated between the biased electrodes separated by air and the dielectric film. The base of the cantilever and the film are immovable in mechanics while an electro-mechanical force coupling solves simultaneously for the displacement and integral electrostatic response of the system. The EFM operation was obtained by applying a bias voltage V between a conductive AFM probe and the bottom of a dielectric film at various probe-film separations. Under the applied voltage V , the cantilever bends downward towards the sample and reaches an equilibrium position where the bending force, $k_c d$, balances the attractive capacitive force, $F_{\text{Probe-D}}$, with k_c being the cantilever spring stiffness and d the bending deflection at the end of the cantilever. The bending of the cantilever from its initial state is allowed by a moving mesh of the air space in between cantilever and the dielectric film.

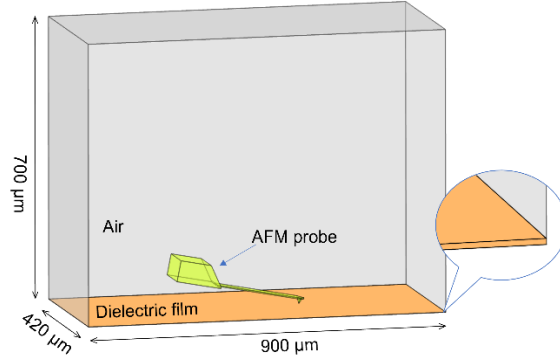


Figure 2: The 3D structure modeled in COMSOL Multiphysics includes a dielectric film (orange), an AFM probe (bright green), and air (gray).

The modeled 3D structure comprising a dielectric film, an AFM probe, and surrounding air is shown in Figure 2. The largest air domain encircling the AFM probe was in the form of a rectangular box (length = 900 μm , depth = 420 μm , and height = 700 μm) with dimensions several times larger than the cantilever dimensions to include all capacitive fringes and the decay of the generated electric field. The AFM probe was made of Si (Young's modulus of 170 GPa and Poisson's ratio of 0.28 for the mechanics) and selected as the biased electrode at the applied voltage V with respect to a grounded electrode (the backside of the dielectric film). Equivalently, the modeled AFM probe corresponds to a metal-coated Si probe commonly used in EFM measurements. The cantilever was modeled as a rectangular bar of length 225.0 μm , width 35.0 μm , and thickness 2.8 μm , with a triangular end shape that has a half-opening angle of 40°. The position of the tip along the cantilever was at 215 μm from the base, the tip height was 13 μm , the half-opening angle of the cone was 25°, and a tip radius of 50 nm. The spring constant of the cantilever was determined to be 3.0 N/m with the first two resonance frequencies in air at 78.4 kHz and 492.6 kHz, respectively.

The electrostatic interaction was solved throughout the space shown in Figure 2, based on the Poisson's equation

$$\nabla \cdot \vec{D} = 0, \quad (9)$$

with $\vec{D}$ being the electric displacement, $\vec{D} = \epsilon_0 k \vec{E}$, $\vec{E}$ the electric field, and k the relative permittivity or dielectric constant ($k = 1$ in air and $k \neq 1$ in the dielectric film); $\vec{E} = -\nabla U$, with U the electric potential. The AFM probe was considered an electrode with electric potential equal to the applied bias, $U = V$, while the back of the dielectric film was grounded, $U = 0$. The other boundary conditions for equation (9) were in the form of the continuity equation, $\vec{n} \cdot (\vec{D}_{\text{air}} - \vec{D}_{\text{film}}) = 0$, at the air-film interface, and zero charge, $\vec{n} \cdot \vec{D} = 0$, at the outside facets of the box in Figure 2 (except the back of the film that was grounded); $\vec{n}$ is the unit normal vector on the interface.

Overall, an unstructured tetrahedral mesh (Figure 3) with about 820000 elements was generated over the 3D geometry of the modeled AFM probe (base, cantilever, and tip), dielectric film, and the air space between them, providing a solution for 4000000 degrees of freedom. The mesh density was increased within and around the apex of the AFM probe to detail the most relevant region for the EFM interaction (refer to Figure 3e), with the minimum element size down to 1 nm. Mesh refinement was performed to assure variations less than 0.1 % in the calculated cantilever bending under the applied bias.

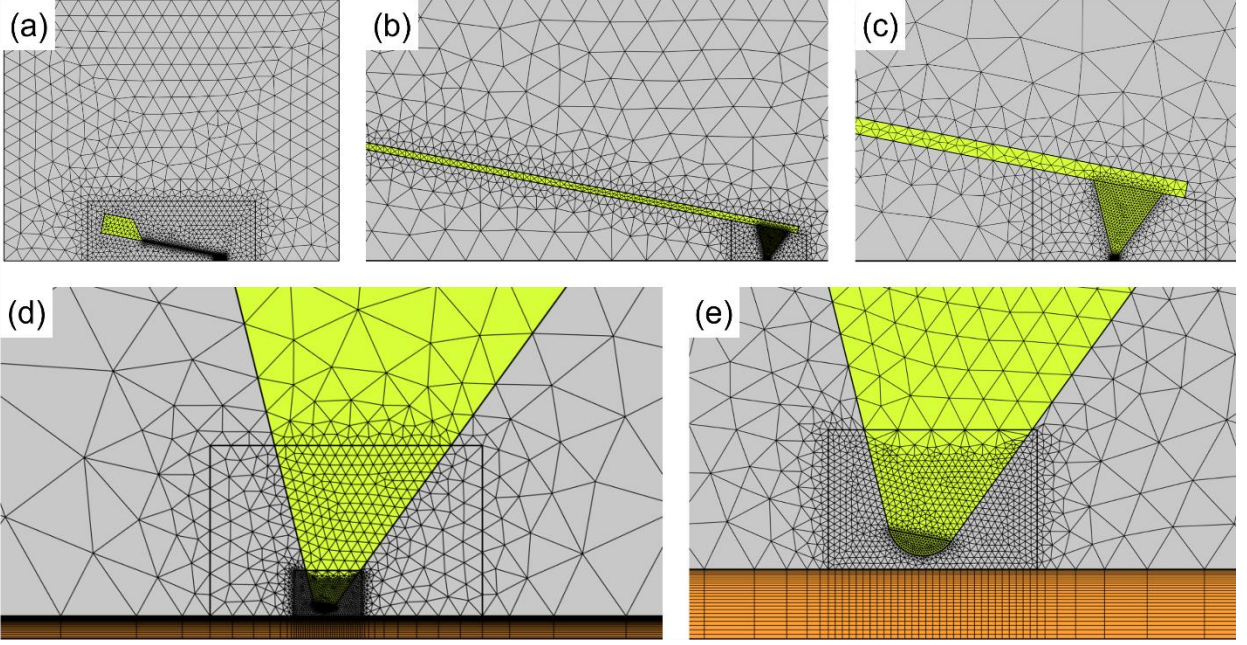


Figure 3: a) to e) Sequential zoom-in images of the regions meshed in COMSOL Multiphysics for the electrostatic interaction between a conductive AFM probe (bright green) and a dielectric film (orange). They are all from the vertical middle section of the rectangular box shown in Figure 2; the image size in a) is $900\ \mu\text{m} \times 700\ \mu\text{m}$. The most enlarged view is e) showing the tip radius of 50 nm, film thickness of 100 nm, tip-film separation of 19 nm, and details of the dense mesh around the tip and within the film; the 100 nm film thickness serves as the scale bar of the image.

In previous works, the numerical modeling featured a simplified version of the AFM probe. Under the imposed 2D axial symmetry constrain, the AFM probe was modeled as a single- or double-angle cone terminated by a spherical apex and with the cantilever reduced to a crown disk [24, 26, 27], with the symmetry axis of the cone normal to the film surface. However, by modeling the entire 3D geometry rather than its 2D counterpart, the model employed here is capable of including the effects introduced by the tilt of the cantilever [35], the geometrical features of the conical tip (*i.e.*, a tapered cross-section) [27], and the cantilever itself (*i.e.*, effective crown disk) [26]. Moreover, with the entire tilted cantilever included in a 3D model, the resultant electric field around the tip has a more accurate resemblance to the actual one. This is because the asymmetry introduced by the tilt of the cantilever enhances some contributions to the electric field from some parts of the cone and cantilever while diminishing others. For the sake of simplicity, we modeled the cantilever as a rectangular bar with triangular end shape and the tip as a circular cone terminated by a spherical apex. The circular cone shape of the tip was chosen to facilitate a straightforward comparison with the discussed analytical model (equation 8) and it does not represent an actual probe. A detailed imaging of the probe (*e.g.* by scanning electron microscopy) at different orientations and magnifications would be necessary [37] for a faithful representation of the shape and dimensions of a probe used in measurements. In early 3D finite element EFM modeling various approximations (cone, tetrahedron, or pyramid) for the tip were considered [38]. However, the shape of an actual tip deviates from any of these simple, regular shapes being necessary a combination of few various 3D shapes for a realistic representation [37]. Additionally, it is important to have a 3D integral representation of the AFM probe in modeling to observe the collective tip-sample interaction rather than the superposition of contributions from

individual parts as done in early 3D modeling [38]. An example of the calculated electric potential around the entire AFM probe is shown in Figure 4 for the vertical cross section bisecting the system shown in Figure 2 along the symmetry plane of the AFM probe. The non-symmetric field distribution around the probe can be seen in the enlarged views shown in Figures 4b and 4c.

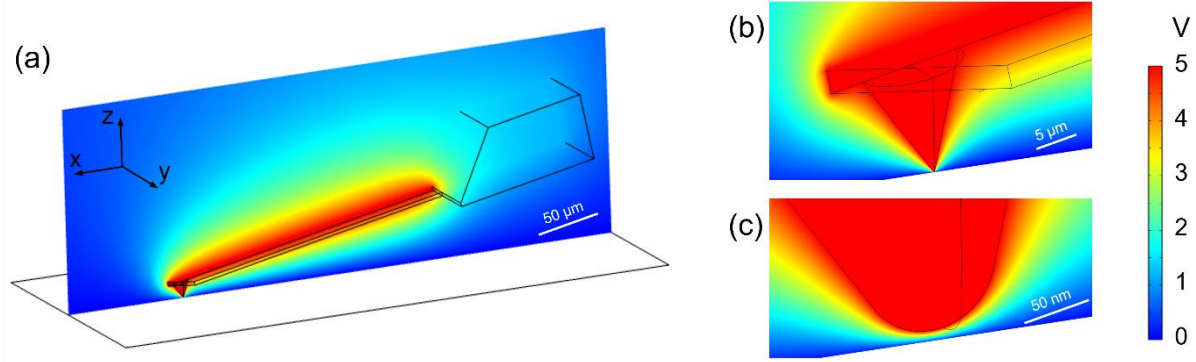


Figure 4: a) Vertical cross-section of the electric potential around a conductive cantilever above a dielectric film ($k = 50$, $h = 100$ nm) at a tip-film separation of 7.5 nm and under an applied bias of 5 V. The cross-section corresponds to the inner rectangular shown in Figure 3a (length = 450 μm , depth = 210 μm , height = 160 μm). Successive zoom-in images around the end of the cantilever and conical tip in b) and the apex of the tip in c). All the scale bars are in the xz -plane.

In simulations, films of various dielectric constants and thickness were considered with the results in terms of force (from the deflection of the cantilever end) versus tip-film separation at a given dc bias, similar to basic dielectric EFM measurements [3]. The calculated curves were then fitted by the analytical model given by equations (8) to extract the dielectric constant and probe the performance of the analytical model versus modeling. In addition, because here the mechanics of the cantilever was observed in modeling, we also retrieved the resonance frequency of the cantilever as function of the tip-film separation. In the general case, when the modeling is to be compared to experimental measurements, the dielectric constant will be obtained by solving an inverse problem in which the dielectric constant is adjusted in modeling until the force-distance curve at a given bias voltage matches the one from measurements.

IV. Numerical Versus Analytical Results for Dielectric EFM

The dielectric constant is a quantitative parameter that relates induced polarization to the electric field generated between the capacitor terminals. In the simple case of a parallel plate capacitor (refer to Figure 1a), the electric field and polarization are uniform, parallel fields along a direction perpendicular to the capacitor plates. This allows for a simple analytical dependence of the capacitance C to the dielectric constant k , as expressed in (1). However, in the case of EFM with one plate replaced by the AFM probe, the generated electric field and the induced polarization are neither parallel fields nor uniform. The analytical model given by (3) and (4) is for the non-parallel field of an EFM capacitor without a dielectric and its extension in (6) and (7) to the dielectric EFM assumes uniform polarization within the dielectric film. In Figures 5 and 6 we look explicitly at the electric field and polarization response of two dielectric films of dielectric constants $k = 4$ and $k = 50$ and thicknesses $h = 10$ nm and $h = 100$ nm in a typical

EFM configuration. The purpose of this exercise is to get a sense of how much the use of the analytical model is justified as a function of dielectric constant and film thickness.

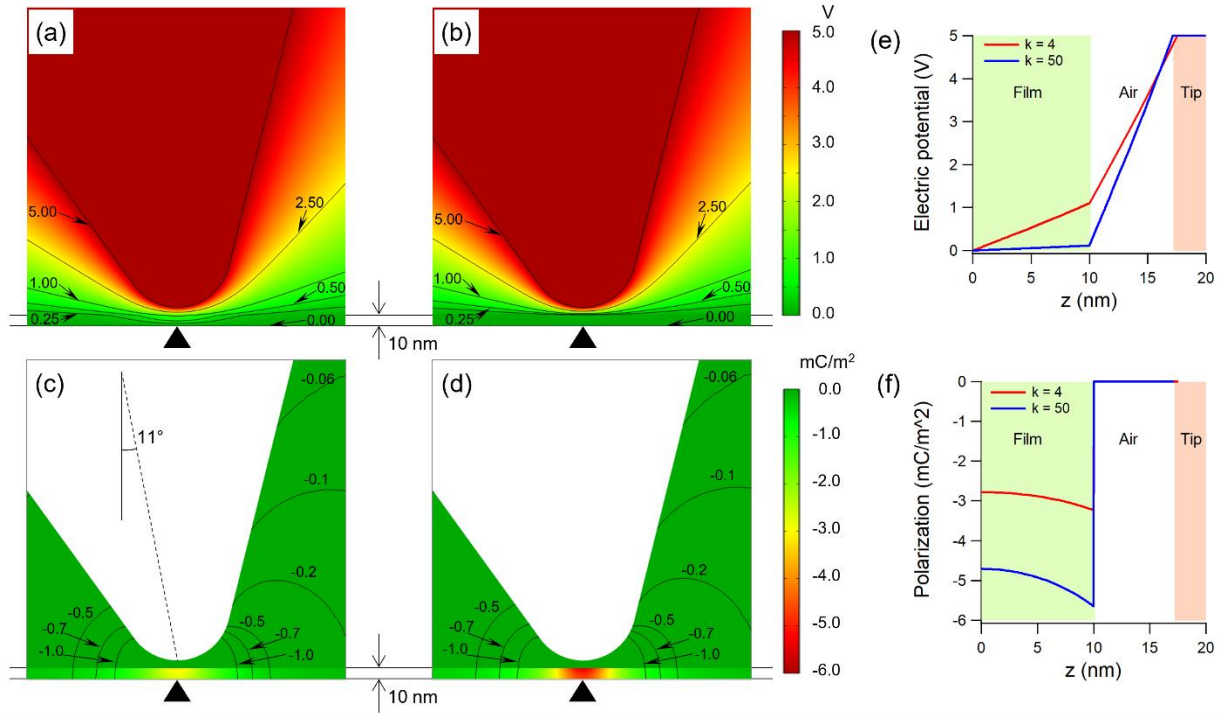


Figure 5: Electrostatic interaction between a conductive tip apex of radius 50 nm at 7.5 nm above a 10 nm thick dielectric film; the tip was biased at 5 V and the bottom of the dielectric film was grounded. a) and b) Surface map and contours of the electric potential (color bar at the right) for films of dielectric constant of 4 and 50, respectively. c) and d) Surface map of the z-component of the film polarization (color bar at the right) and contours of the z-component of the electric displacement field (contour labels in mC/m² units) for films of dielectric constant of 4 and 50, respectively. e) Electric potential profiles along the vertical direction (z-axis) from the bottom of the film (z = 0 nm) to the apex of the tip, at the location marked by the black triangle at the bottom of the film in a) and b). f) z-polarization component profiles along the vertical direction (z-axis) from the bottom of the film (z = 0 nm) to the apex of the tip, at the location marked by the black triangle in c) and d) at the bottom of the film. The tilt of the tip with respect to the normal to the film surface is shown in c). The area shown in a) to d) is 300 nm x 300 nm. The film thickness is marked by horizontal lines extended outside the bottom of the maps.

The dielectric EFM configuration analyzed in Figures 5 and 6 was that as described above, with the AFM probe biased at + 5 V and the dielectric film on top of a grounded electrode. Figures 5a and 5b show the electric potential maps and contours around the tip apex and within the dielectric film while Figures 5c and 5d show the map of the z-component of the film polarization and contours of the z-component of the electric displacement field for films of dielectric constant of 4 (Figures 5a and 5c) and 50 (Figures 5b and 5d). As can be seen from the electric potential maps, the electric field penetrates further inside the $k = 4$ dielectric film than the $k = 50$. The decay of the electric potential along the vertical line at the closest distance (location marked by triangles in Figures 5a and 5b) is shown in Figure 5e for the two

dielectric constants. In both cases, the dependence is linear inside the films, decreasing from 1 V and 0.1 V at the top surface of the film to 0 at the bottom of the film for the $k = 4$ and $k = 50$ films, respectively. The initial position of the tip was 10 nm from the top surface. This is measured from the apex of the tip and due to the tilt of the AFM probe the shorter tip-film separation is reduced by about 1 nm. Furthermore, upon the applied bias the equilibrium position is reached at even a shorter tip-film separation, decreased by roughly 1.5 nm, as indicated at the right of Figure 5e by the position of the orange strip; these positions are slightly different for each dielectric constant due to the difference in the magnitude of the capacitive force. As shown in Figures 5c and 5d, the electric field lines are almost perpendicular to the biased tip surface and to the grounded back of the dielectric film. Because the dielectric film is thin the lines inside the film are nearly parallel and the induced polarization almost uniform across the film. The polarization profiles along the vertical line at the shortest tip-film separation distance are shown in Figure 5f. It can be seen that the polarization is not entirely constant across the film but has a small variation. Under these circumstances, for thin dielectric films, we assume that the extension of the analytical model (equations (8)) should be valid.

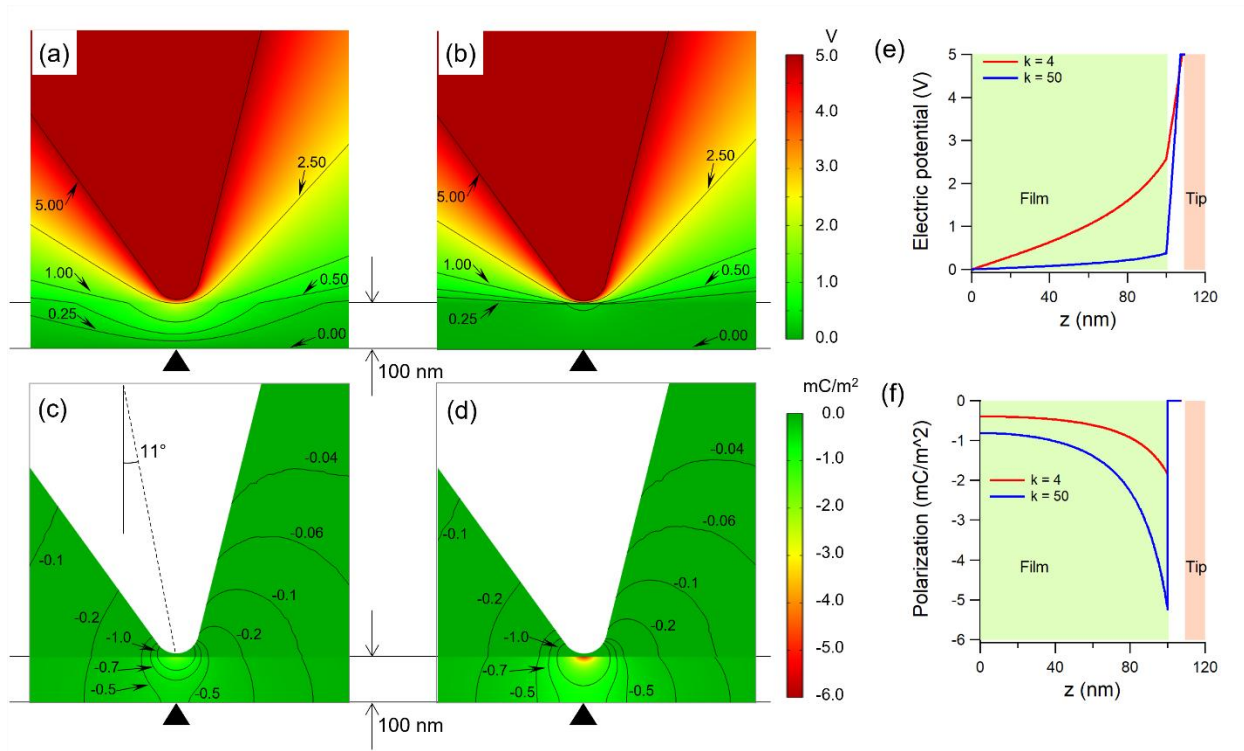


Figure 6: Electrostatic interaction between a conductive tip apex of radius 50 nm at 7.5 nm above a 100 nm thick dielectric film; the tip was biased at 5 V and the bottom of the dielectric film was grounded. a) and b) Surface map and contours of the electric potential (color bar at the right) for films of dielectric constant of 4 and 50, respectively. c) and d) Surface map of the z-component of the film polarization (color bar at the right) and contours of the z-component of the electric displacement field (contour labels in mC/m² units) for films of dielectric constant of 4 and 50, respectively. e) Electric potential profiles along the vertical direction (z-axis) from the bottom of the film ($z = 0$ nm) to the apex of the tip, at the location marked by the black triangle at the bottom of the film in a) and b). f) z-polarization component profiles along the vertical direction (z-axis) from the bottom of the film ($z = 0$ nm) to the apex of the tip, at the location marked by the black triangle in c) and d) at the bottom of the film. The tilt of the tip with respect to the normal to the film surface is shown in c). The area shown in a) to d) is 700 nm x 700 nm. The film thickness is marked by horizontal lines extended outside the bottom of the maps.

The above analysis was also performed for dielectric films of dielectric constants $k = 4$ and $k = 50$ and a thickness of 100 nm with results presented in Figure 6. Similar to the case of the thin film, for the 100 nm thick film, the electric potential (Figures 6a and 6b) penetrates deeper inside the film of $k = 4$ than $k = 50$. However, as can be seen in Figure 6e, in both cases the decay of the electric potential across the film is not linear. This correlates with a more pronounced parabolic bending of the electric field lines within the film (refer to the electric displacement contours in Figures 6c and 6d). As a result, the polarization of the dielectric is highly non-uniform inside the film as shown by the color gradient in the maps of Figures 6c and 6d and the strong non-linear profiles of the electric displacement field across the thickness of the film (curves in Figure 6f). This analysis suggests that for thick dielectric films, and especially for ones with higher dielectric constants, the analytical approximations given by (8) might not provide a realistic description. In Table I is shown the relative variation of the polarization across the thickness of the films discussed here. Additionally, the relative variations of the polarization across the 100 nm film for a tip-film separation of 30 nm are added in the last two lines of Table I. As can be seen, these relative variations are attenuated by about 20% as the tip-separation was increased from 10 nm to 30 nm, which suggests that the analytical model might be a better description for measurements with greater tip-film separations.

Table I: Variation in the z-polarization across the dielectric film (the difference between the top and bottom sides of the film) at the closest tip-film separation; data in rows 1 through 4 are based on the curves shown in Figures 5f and 6f.

Film thickness h (nm)	Dielectric constant k	Initial tip-film separation (nm)	z-polarization variation dP_z (%)
10	4	10	14
10	50	10	17
100	4	10	79
100	50	10	85
100	4	30	59
100	50	30	63

The most common type of dielectric AFM measurement is based on the electrostatic force response under an applied voltage bias, either in quasi-static [3, 22, 25] or amplitude-modulation [5, 16, 22, 26] methods at a fixed frequency. Alternatively, the measurements could be performed in terms of force gradient, in which case, frequency-modulation methods are to be adopted [2, 4, 39]. Due to the commonality of force-based measurements, we will look primarily to the biased force-distance response of a conductive AFM probe above dielectric films on metal substrates. For the probe-film system discussed above, these force-distance ramps were calculated in COMSOL Multiphysics under a fixed bias of 5 V for a metal substrate and dielectric films of dielectric constants 5, 10, 20, 35, and 50. For each dielectric constant, two film thicknesses, 10 nm and 100 nm, were analyzed. Given the fact that noise is inherent to the actual measurements, some amount of noise was added to the calculated AFM deflection afterwards. In general, the deflection of an AFM cantilever is measured in air with a thermal noise from 10 pm to 100 pm. For dielectric measurements, a deflection noise was reported of about 100 pm in dc biased EFM [3] and in the range of 4 pm to 25 pm in amplitude modulation EFM (inferred from the reported gradient capacitance

noise) [21, 41]. In this work, we considered three different values for the added noise of 10 pm, 50 pm and 100 pm. The statistics of actual measurements was mimicked by generating a set of 4096 (64×64) curves from each calculated curve by adding random noise, within the given level to the calculated values. In practice, this will correspond to a low-resolution scan over the area of interest. The 4096 generated curves were divided into 64 rounds of measurements with 64 measurements per round. In each round of measurements, the 64 generated measurements at a given tip-film separation were used to calculate the mean and the standard deviation of the values used for fitting. The analytical model given by equations (8) was then used to do a non-linear model fit with standard deviations as weights (Mathematica 13.2 from Wofram Research Inc.); the adjusted R^2 (adjusted for the number of model parameters) of each fit was better than 0.9999. For each force versus tip-film separation data set, the average fit value of the dielectric constant k_i and its fit uncertainty σ_i were determined, with index i counting the measurement round. Several fit examples are shown in Figure 7 and will be discussed later. A statistical analysis was further performed on the fit results of all 64 rounds of measurements for a given thickness and dielectric constant. These rounds of measurements correspond to experimental trials, when a set of different film thicknesses and dielectric constants would be measured successively to insure the repeatability of results. The weighted mean and its standard deviation from all these measurement rounds were calculated as

$$k = (\sum k_i / \sigma_i^2) / (\sum 1 / \sigma_i^2) \quad (9)$$

and

$$\sigma = 1 / \sqrt{\sum 1 / \sigma_i^2}, \quad (10)$$

respectively. First, we performed fits over a metallic substrate and determined that a nominally $R = 50$ nm is best approximated by the analytical model (8) as (41.725 ± 0.032) nm, (41.747 ± 0.037) nm, and (41.717 ± 0.048) nm for the noise levels of 10 pm, 50 pm, and 100 pm, respectively, which are about 20 % less than the actual R , consistent to what is reported in practice [21, 24]. The parameter F_0 was found to be (-0.659 ± 0.01) nN for any of the considered noise levels. With these two determined parameters, the analytical model (8) was then used to fit the curves of force versus probe-film separation over the dielectric films. The results of this analysis are presented in Table II for all the film thicknesses, dielectric constants, and noise level considered in this work.

Table II: Fit results from the force versus tip-film separation for the tilted AFM probe over the 10 nm and 100 nm thick dielectric films of dielectric constants of 4, 10, 20, 35, and 50. The fits were performed over an extended range of tip-film separation z from 13 nm to 150 nm. Three different noise levels of 10 pm, 50 pm, and 100 pm were considered for each of the analyzed cases.

Thickness (nm)	Theoretical value k	Fit value k , average $\pm$ standard deviation		
		10 pm noise	50 pm noise	100 pm noise
10	4	3.809 ± 0.014	3.805 ± 0.016	3.759 ± 0.021
	10	9.920 ± 0.082	9.828 ± 0.091	9.615 ± 0.117
	20	20.716 ± 0.346	20.639 ± 0.403	18.284 ± 0.423
	35	38.920 ± 1.202	35.867 ± 1.217	30.465 ± 1.272
	50	59.174 ± 2.805	53.838 ± 2.911	42.135 ± 2.686
100	4	5.219 ± 0.008	5.214 ± 0.09	5.230 ± 0.012
	10	14.692 ± 0.040	14.691 ± 0.043	14.714 ± 0.053
	20	31.403 ± 0.140	31.468 ± 0.151	31.305 ± 0.177
	35	57.513 ± 0.317	57.061 ± 0.351	57.533 ± 0.460
	50	84.669 ± 0.610	84.625 ± 0.708	82.943 ± 0.896

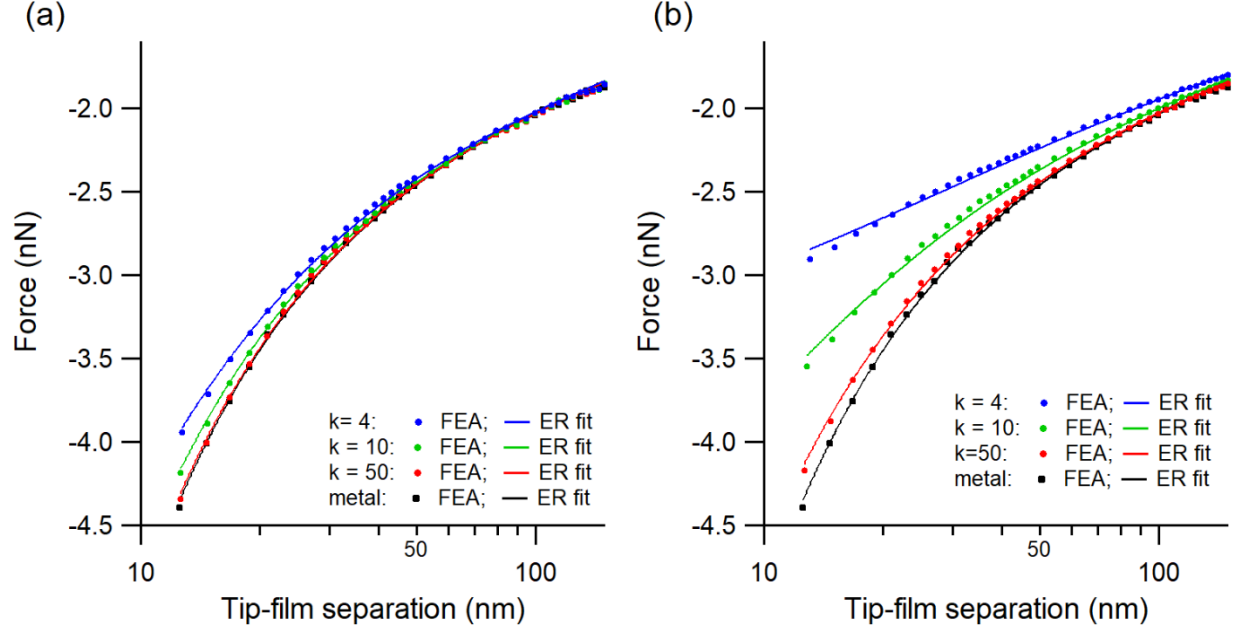


Figure 7: The electrostatic response in terms of force versus tip-film separation between the studied conductive AFM probe and a metal substrate and three dielectric films ($k = 4, 10$, and 50) on a metal substrate. The thickness of the dielectric films is 10 nm in a) and 100 nm in b). The markers are average values of numerical calculations with added noise of 10 pm (see text for details) and the continuous lines are fits of these values by the analytical model described in the text. The error bars associated with the standard deviation uncertainties (see Table II) are omitted from the plots as they are within the size of the markers. The tip-film separation is the distance z specified in Figure 1. ER fit means that the fit was performed over the entire range of data.

A few selected fits of the force versus tip-film separation under 5 V bias from a round of measurements are shown in Figure 7 for 10 nm and 100 nm dielectric films for an added noise of 10 pm. The tip-film separation is the distance z marked in Figure 1 between the apex of the tip and the top surface of the film. This distance is the final equilibrium distance that considers the bending of the cantilever due the electrostatic probe-sample interaction. As can be seen, the fits follow the data reasonably well for the 10 nm films in Figure 7a over the entire range (ER) of z from 13 nm to 150 nm. The obtained dielectric constants from these fits (refer to Table II) are in good agreement with the input values up to about 20 and start to show an increased deviation above 20 (e.g., about 20% off for $k = 50$). This is because, as k increases, the ratio h/k in equation (8) decreases and approaches the limit of the metallic substrate (refer to the curve $k = 50$ in Figure 7a). For the 100 nm thick films, the ratio h/k is larger, hence, the curves of different k in Figure 7b separate more from each other and from that of the metallic substrate. However, the fits produced by the analytical model (8) do not show a good track over the entire range of z for the thick films. This observation correlates with the previous observation that the induced polarization in the thick films is not uniform across the film thickness, a behavior that is not captured by the model used here.

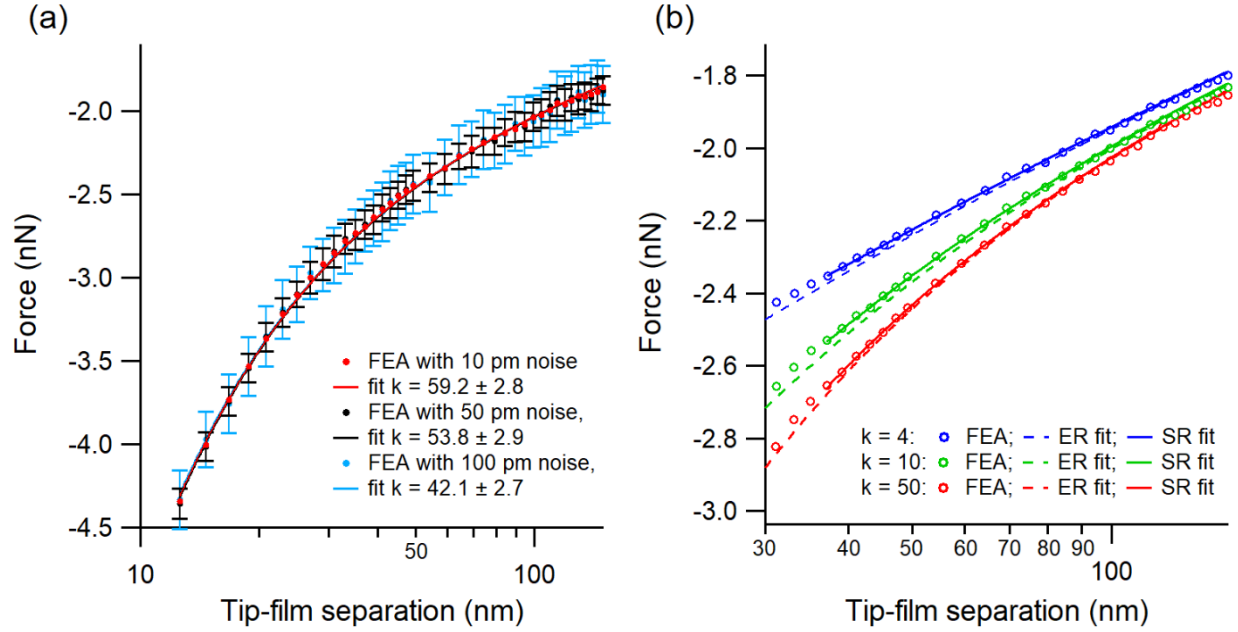


Figure 8: a) The electrostatic response in terms of force versus tip-film separation between the studied conductive AFM probe and a 10 nm thick dielectric film ($k = 50$) on a metal substrate. The fit results (continuous line and obtained k values) were observed for various noise amounts added to the numerical calculations. b) The entire range (ER) fits on 100 nm dielectric films from Figure 7b are shown here in comparison to shorter range (SR) fits at higher tip-film separations. The ER fits are with dashed lines and SR fits with continuous lines.

From the above analysis, two aspects were identified as main factors affecting the data analysis by a simple analytical model as the one described in equation (8): One is the noise level present in measurements and the second is the neglect of the field gradients across the thick films. As expected, the noise will have a greater effect on the analysis of thin films because there is little separation among curves of different k for the thin films (refer to Figure 7a). Figure 8a shows an example for the 10 nm thick film of $k = 50$ curves with different amounts of noise added. With the increase in noise the accuracy of the fit is increasingly altered, with k going from about 60 for 10 pm noise to about 40 for 100 pm noise; the uncertainty remains the same due to the increased number of measurements and rounds of measurements. It is interesting to observe that in fact, for the 50 pm noise, the determined k approaches the actual value due to the relaxation introduced by noise into the fit. This is merely coincidental (with larger noise the fit is inaccurate) and adding noise to the measurements is not the solution across different thicknesses.

The second aspect related to the inability of the analytical model to correctly describe the probe-film capacitive force for thick dielectric films was previously addressed [24, 26] by using approximative solutions in terms of equivalent geometries (*e.g.*, replace the cantilever contribution by that of a crown disk). It is interesting to observe that despite mediocre fitting by the analytical model the curves for the 100 nm thick films (Figure 7b) do show good separability, meaning good dielectric constant sensitivity, even at greater z tip-film separations. This contrasts with the curves for the 10 nm films (Figure 7a) that strongly overlap each other at greater distances. Because of that we can restrict the fit to a shorter range (SR) at higher z values from 37 nm to 150 nm. These new fits do show better data fitting in Figure 8b (refer to the continuous lines) in comparison to those over the entire range of z values (refer to the dashed lines in Figure 8b and continuous lines in Figure 7b). The fit results for the 100 nm films of all considered dielectric

constants and noise levels are given in Table III. This improvement in the fit at larger tip-film separation for thick films correlates with the smaller variation in the polarization across the film that was mentioned in Table I for a z separation of 30 nm. However, the downside of operating at larger tip-film separation is the decrease in lateral resolution [3]. The lower limit of the SR fit (*i.e.*, 37 nm) was decided by gradually changing the minimum tip-film separation until an improvement in fit was observed.

Table III: Fit results from the force versus tip-film separation for the tilted AFM probe over the 100 nm thick dielectric films of dielectric constants of 4, 10, 20, 35, and 50. The fits were performed over a shorter range (SR) of tip-film separation z from 37 nm to 150 nm. Three different noise levels of 10 pm, 50 pm, and 100 pm were considered for each of the analyzed cases.

Thickness (nm)	Theoretical value k	Fit value k , average $\pm$ standard deviation		
		10 pm noise	50 pm noise	100 pm noise
100	4	4.767 ± 0.005	4.757 ± 0.010	4.749 ± 0.018
	10	12.003 ± 0.025	11.944 ± 0.045	11.954 ± 0.084
	20	23.064 ± 0.096	22.889 ± 0.162	22.419 ± 0.289
	35	39.069 ± 0.266	38.329 ± 0.430	35.773 ± 0.688
	50	53.047 ± 0.492	51.668 ± 0.779	46.456 ± 1.165

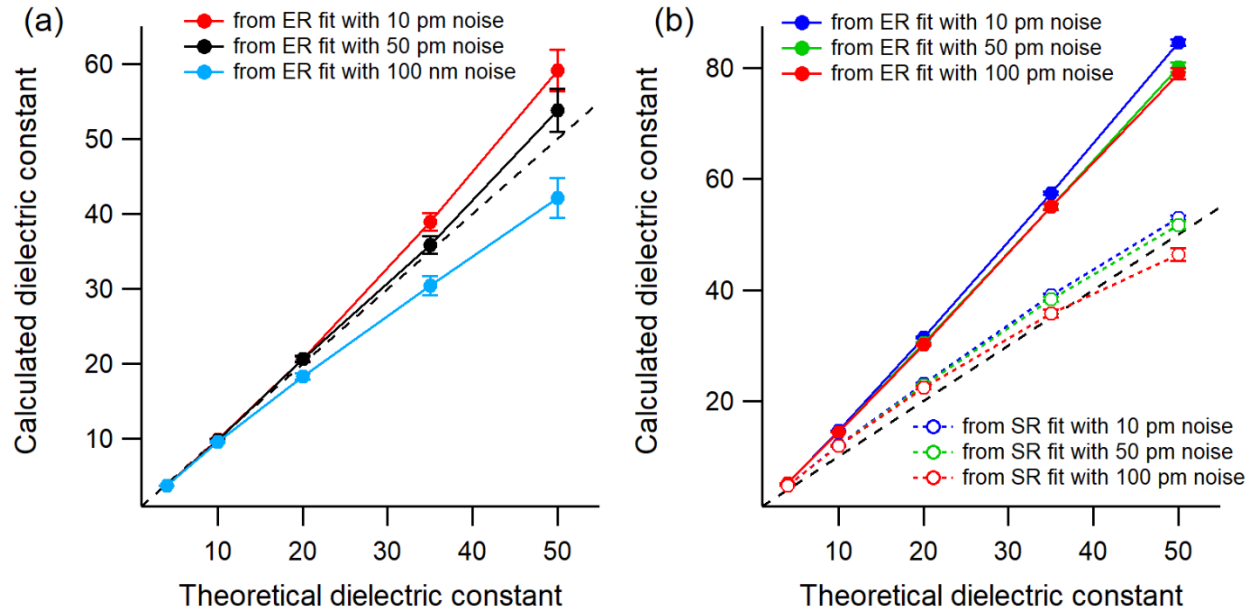


Figure 9: a) Dielectric constants obtained from ER fits for the 10 nm thick dielectric films; the results for the 10 pm noise come from the curves shown in Figure 7a. b) Dielectric constants obtained from ER and SR fits for the 100 nm thick dielectric films; the results for the 10 pm noise come from the curves shown in Figures 7b and 8b, respectively. The dashed line is the bisector line of the plot between theoretical and calculated values. All the plotted values and uncertainties are given in Tables II and III.

All the fit results given in Tables II and III are plotted in Figure 9 for the two thicknesses and various dielectric constants and noise levels. From Figure 9a we can say that the analytical model is a good representation for thin films with dielectric constant less than 10 disregarding the noise level of the measurements. With the increase in the dielectric constant above 20, the fit starts to lose accuracy and the noise level further affects the determined dielectric constant. The fit results for the 100 nm films are shown in Figure 9b. In this case, we can say that the analytical model is not a good description for any dielectric constant above 4 and the noise has a smaller influence than that observed on thin films. The fitting is improved by restricting the z range at larger tip-film separations. However, the analytical model is still not reproducing the actual dependences although the determined dielectric constants come quite close, within few percent, to the nominal k values.

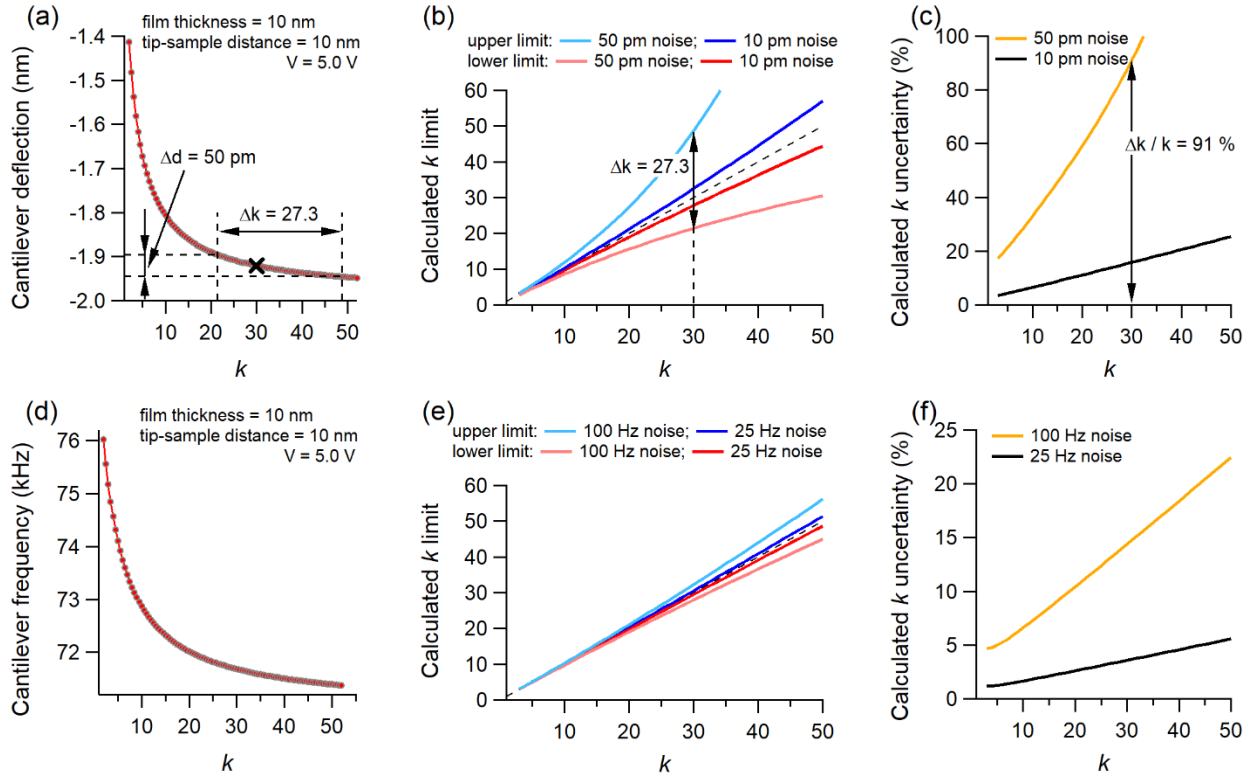


Figure 10: Deflection and frequency sensitivities for resolving the dielectric constant from EFM measurements at 5 V bias over a 10 nm dielectric film, with the probe apex initially at 10 nm distance from the film surface: a) The deflection cantilever versus dielectric constant; b) Upper and lower dielectric constant limits calculated for noise thresholds of 10 pm and 50 pm noise around each dielectric constant value in a); c) The relative difference (in percent) between the upper and lower branches from b) for the noise thresholds of 10 pm and 50 pm; d) The resonance frequency of the cantilever versus dielectric constant; e) Upper and lower dielectric constant limits calculated for noise thresholds of 25 Hz and 100 Hz noise for the resonance frequency around each dielectric constant value in a); f) The relative difference (in percent) between the upper and lower branches from e) for the noise thresholds of 25 Hz and 100 Hz, respectively.

The above analysis for EFM spectroscopy pointed to the limited applicability of the analytical model to extract the dielectric constant from measurements. However, the results can be greatly improved with a more complex analytical model or by using numerical modeling to realistically capture the geometry and physics of the system. It is thus relevant to evaluate the measurement sensitivity independently of an

analytical model used for analysis in both spectroscopy and mapping configurations. In spectroscopy, the measurement sensitivity is expected to be higher because the response over a range of z separations is measured. However, the spatial resolution will be moderate if the measurements are to be assembled into a volume mapping. On the other hand, mapping at a fixed tip-sample separation, either in amplitude or frequency modulation, can produce high spatial resolution maps over the scan area [4, 21, 22, 40, 41]. In the following, independent of the model analysis, we evaluate the measurement sensitivity for dielectric constant determination from force and force-gradient EFM measurements at a fixed tip-film separation in the presence of various noise levels. Both the deflection and resonance frequency of the cantilever discussed in the following are from the finite element calculations in COMSOL Multiphysics. After the equilibrium position of the AFM probe under an applied bias was found by a Stationary study (from the coupled Solid Mechanics and AC/DC modules), the resonance frequency was determined as the frequency of the first eigenmode of the system as calculated by the Eigenfrequency study of the Solid Mechanics module.

With the same system described above for spectroscopic modeling, the deflection and resonance frequency of the AFM probe were calculated for a given initial tip-film separation of 10 nm as a function of the dielectric constant of the film. The initial separation was the same for all the simulations but the final equilibrium z separation was dependent on the dielectric constant through the capacitive force acting on the AFM probe. The EFM responses are shown in Figures 10a and 10b in terms of deflection and resonance frequency of the cantilever for the case of a 10 nm thick film with the dielectric constant up to 50 under a bias voltage of 5 V. As can be seen, the resonance frequency shows a steeper variation over the same range of dielectric constants than the deflection, so a greater sensitivity is expected in terms of force-gradient than force. Quantitatively this is determined by comparing the response to various noise threshold levels. For the deflection, we selected the noise levels of 10 pm and 50 pm, and for the frequency the two thresholds were selected at 25 Hz and 100 Hz. The 25 Hz is normally achieved in resonance frequency tracking in air [37] and the 100 Hz was selected as a higher limit. These thresholds were used to calculate the crossing levels around each of the k values and construct the upper and lower limit branches shown in Figure 10b for deflection and Figure 10e for resonance frequency. The difference between these two branches gives the measurement sensitivities or uncertainties in the determined k plotted in Figures 10c and 10f. An example of how the uncertainty $\Delta k/k$ was calculated is shown across the Figures 10a to c for $k = 30$ with noise of 50 pm in deflection. For example, from the data given in Figure 10c, we can see an uncertainty of 25 % at $k = 50$ for a noise level of 10 pm and 90 % at $k = 30$ for a noise level of 50 pm. On the other hand, with the resonance frequency the uncertainties are substantially reduced to about 6 % with 25 Hz noise and 22 % with 100 Hz noise for $k = 50$ (refer to Figure 10f).

The same analysis was performed for the 100 nm thick films with the results presented in Figure 11. We expect greater sensitivities for the 100 nm than the 10 nm thick films based on the range of values over the same intervals of k values: The change in the deflection is 1 nm in Figure 11a for the 100 nm films compared to 0.6 nm in Figure 10a for the 10 nm thick films and the change in resonance frequency is 6 kHz in Figure 11d for the 100 nm films compared to 5 kHz in Figure 10d for the 10 nm films. Indeed, a reduced separation between the limited branches shown in Figures 11b and 11e was obtained for each of the considered noise thresholds. In Figure 11c, based on the calculated deflection variations for a nominal $k = 50$, we have an uncertainty of about 7 % with a noise level of 10 pm and 34 % with a noise level of 50 pm. On the other hand, the uncertainties in the resonance frequency are substantially reduced to about 2 % with 25 Hz noise and 7 % with 100 Hz noise for the same $k = 50$ (refer to Figure 11f). It is interesting to see that there is an increase in uncertainty with the increase in noise at small dielectric constants for both deflection and frequency. These results show an increased sensitivity for the 100 nm films compared to the 10 nm films and they correlate with the discussed separability of the curves in Figure 7 for the two film thicknesses.

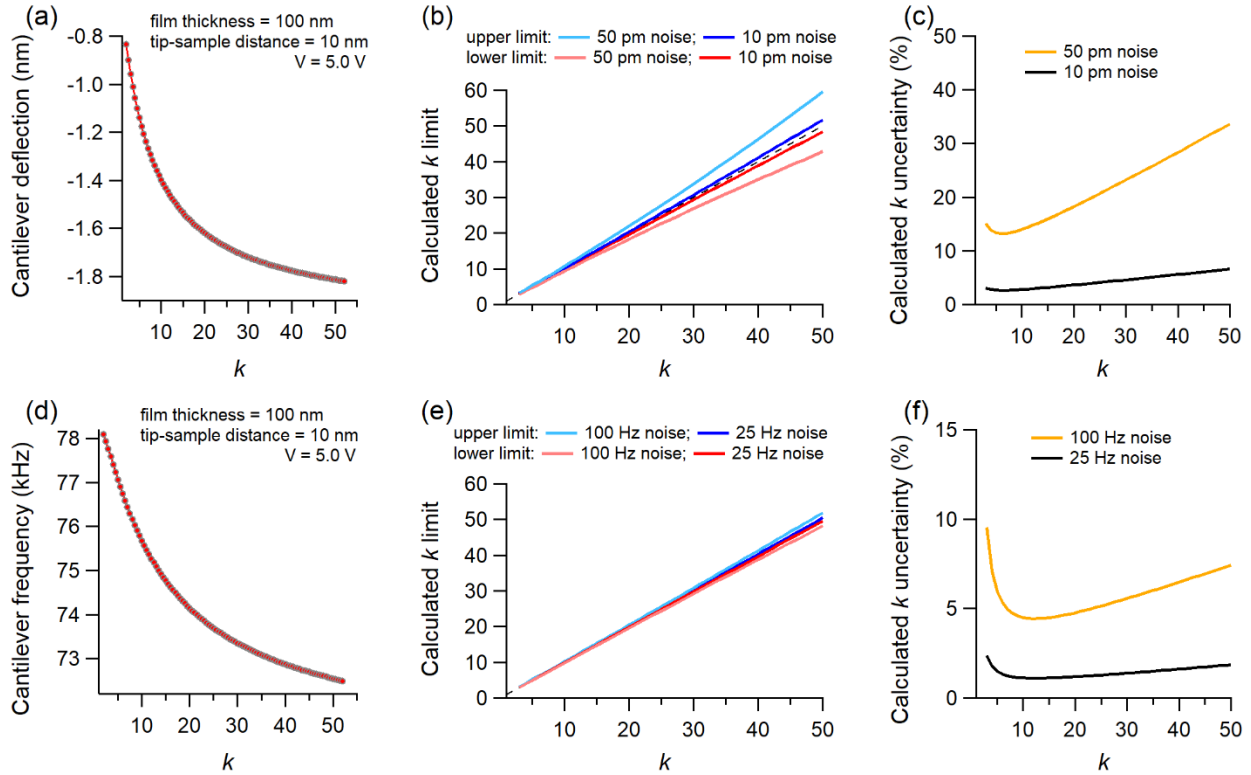


Figure 11: Deflection and frequency sensitivities for resolving the dielectric constant from EFM measurements at 5V bias over a 100 nm dielectric film, with the probe apex initially at 10 nm distance from the film surface: a) The deflection cantilever versus dielectric constant; b) Upper and lower dielectric constant limits calculated for noise thresholds of 10 pm and 50 pm noise around each dielectric constant value in a); c) The relative difference (in percent) between the upper and lower branches from b) for the noise thresholds of 10 pm and 50 pm; d) The resonance frequency of the cantilever versus dielectric constant; e) Upper and lower dielectric constant limits calculated for noise thresholds of 25 Hz and 100 Hz noise for the resonance frequency around each dielectric constant value in a); f) The relative difference (in percent) between the upper and lower branches from e) for the noise thresholds of 25 Hz and 100 Hz, respectively.

The observed trend of increasing uncertainty with the increase in the dielectric constant was previously confirmed in measurements. Thus, for the dielectric constants in the 2.5 to 10 range, the uncertainty was found in [21] to increase from 3 % at $k = 2.8$ to 10 % at $k = 9.4$. Similarly, in [42] the uncertainty for the determined dielectric constants was found to increase from 8 % at $k = 2.5$ to 20 % at $k = 6.2$. In both cases, the measurements were performed by EFM force modulation, so the comparison is with the results presented in Figures 10c and 11c. Thus, in [21], the samples measured were nanoparticles of diameters about 30 nm and the measurement noise was about 4 pm (the lowest reported measurement noise for EFM-based dielectric constant measurements), so the results are more aligned with the curve of lower noise in Figure 10c. On the other hand, in [42], the samples were in the form of films with thickness about 500 nm and the measurement noise was presumably higher than 10 pm, so the observed uncertainties are at the level of the curve of higher noise in Figure 11c. It is therefore remarkable that the modeling results in terms of uncertainties go along with the measurements, both qualitatively and quantitatively, in terms of

dielectric constant (increase with the increase in the dielectric constant) and measurement noise (increase with the increase in noise).

It is however worth pointing out that the constant tip-film separation sensitivities discussed above are for relatively small separations, with the tip at 10 nm above the film surface. With the tip close to the surface, both the measurement sensitivity and spatial resolution are increased. In practice, it is common to operate with the tip at relatively larger distances from the film surface, in the 20 nm to 50 nm range [3, 4, 16, 21, 22, 43, 44]. As an example, we repeated the above analysis for 100 nm thick films with the tip initially at 30 nm above the film surface. The results are shown in Figure 12 and are to be compared to those in Figure 11 (initial tip-surface separation of 10 nm). For the same noise thresholds, the limit branches around any given k spread wider in both deflection (Frequency 12b) and frequency (Frequency 12e) from the diagonal of their plots. Thus, the uncertainties shown in Figures 12c and 12f are consistently greater than their counterparts in Figures 11b and 11f, respectively. This indicates a reduction in the measurement sensitivity with the increase in the tip-film separation, in addition to the observed degradations due to the increase in measurement noise and dielectric constant. In some cases, the uncertainty due to all these factors, including the use of a simple model rather than that of a detailed numerical modeling, can be so large that it severely affects the accuracy of the results, even showing an inverse contrast in the determined dielectric constants. Thus in [19], the dielectric constants of 100 nm coats of Al_2O_3 and SiO_2 on polystyrene particles were determined to be 9.8 and 17, respectively. While 9.8 compares well with the nominal value for the dielectric constant of Al_2O_3 , the 17 value is much higher than the dielectric constant of SiO_2 , which is around 3.9.

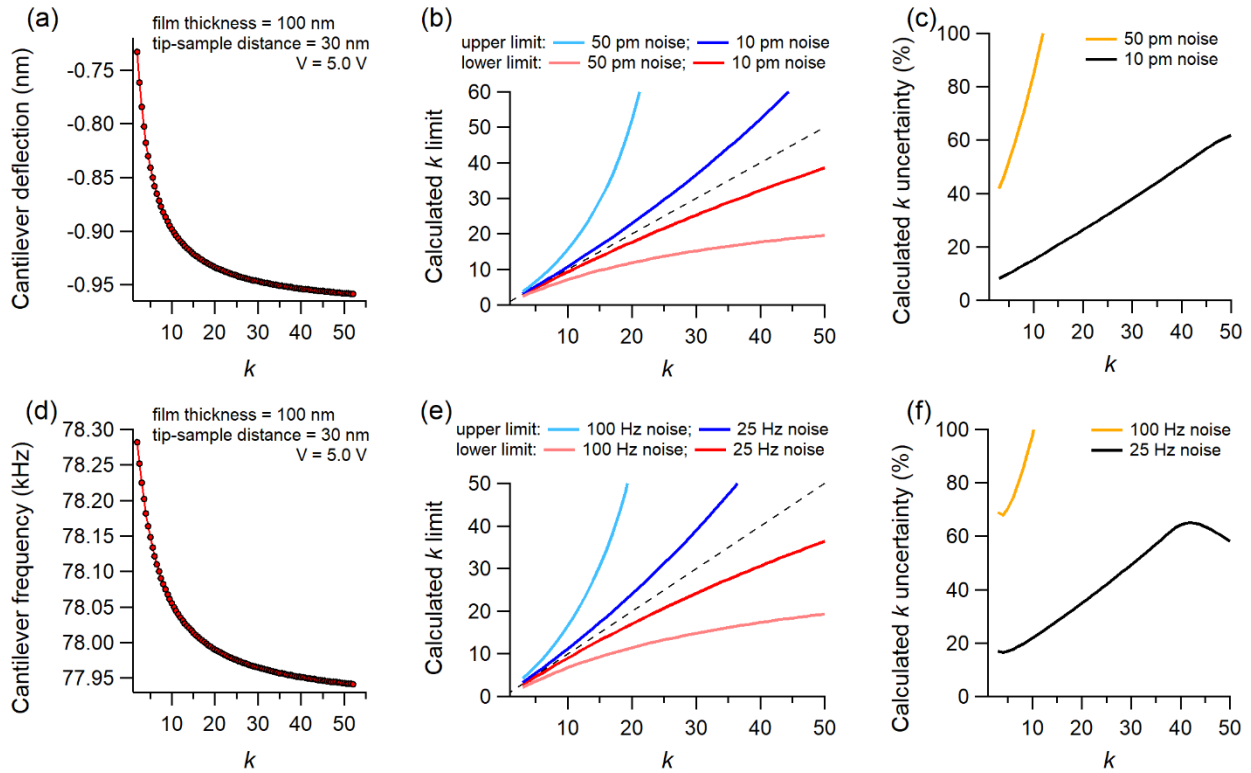


Figure 12: Deflection and frequency sensitivities for resolving the dielectric constant from EFM measurements at 5V bias over a 100 nm dielectric film, with the probe apex initially at 30 nm distance from the film surface: a) The

deflection cantilever versus dielectric constant; b) Upper and lower dielectric constant limits calculated for noise thresholds of 10 pm and 50 pm noise around each dielectric constant value in a); c) The relative difference (in percent) between the upper and lower branches from b) for the noise thresholds of 10 pm and 50 pm; d) The resonance frequency of the cantilever versus dielectric constant; e) Upper and lower dielectric constant limits calculated for noise thresholds of 25 Hz and 100 Hz noise around each dielectric constant value in a); f) The relative difference (in percent) between the upper and lower branches from e) for the noise thresholds of 25 Hz and 100 Hz, respectively.

In both cases discussed above, force-distance ramps and force or frequency response at constant tip-sample separation under a dc bias, numerical calculations were developed to mimic realistic EFM configurations. Through examples on various film thicknesses and dielectric constants, we demonstrated here that the finite element analysis is more inclusive and, implicitly, should perform better than an analytical model. Currently, there is no comprehensive analytical model to accurately describe the electrostatic interaction between an AFM probe and dielectric films of arbitrary thickness and dielectric constant. The alternative to that is to use a 3D finite element modeling as described in this work with the probe shape and dimensions replicating those of the actual probe and the exact tip-sample configuration of the measurement. The dielectric constant then would be determined through an inverse problem by minimizing the difference between the calculated and measured force or frequency versus distance curves. Besides the applications to homogeneous films discussed here, the 3D modeling of EFM measurements might be the only solution in the case of materials or structures containing dielectric heterogeneity at the nanoscale.

V. Conclusions

In this work we modeled in detail the electrostatic force microscopy system between a conductive AFM probe and dielectric films of various thicknesses and dielectric constants. The main objective was to compare the efficiency of a widely used analytical model against a realistic 3D modeling case with all parameters known. With defined dielectric constants and all the geometrical parameters considered known, we used the model to back calculate the dielectric constant of films by fitting the force response as a function of tip-film separation. From such spectroscopic analysis, it was concluded that the analytical model performs well for thin films of dielectric constants smaller than 10 but shows large errors for thick films of any dielectric constant. For thick films, the analysis is significantly improved for spectroscopic measurements performed at relatively large distances from the film surface. Independent of the applied model analysis, it was found that for constant height measurements frequency modulation will always provide a superior sensitivity to amplitude modulation in determining the dielectric constant. The conclusion here is that an analysis based on numerical calculations that replicate the actual geometry of the system could greatly increase the accuracy of dielectric EFM measurements.

Disclaimer

Certain commercial equipment, software, instruments, and materials are identified in this document. Such identification does not imply recommendation or endorsement by the National Institute of Standards and Technology, nor does it imply that the products identified are necessarily the best available for the purpose.

Conflict of Interest

The author has no conflicts to disclose.

Data Availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

References

1. A.V. Krayev and R.V. Talroze, Electric force microscopy of dielectric heterogeneous polymer blends, *Polymer* **45**, 8195–8200 (2004).
2. P. S. Crider, M. R. Majewski, Jingyun Zhang, H. Oukris, and N. E. Israeloff, Local dielectric spectroscopy of polymer films, *Appl. Phys. Lett.* **91**, 013102 (2007).
3. G. Gramse, I. Casuso, J. Toset, L. Fumagalli, and G. Gomila, Quantitative dielectric constant measurement of thin films by DC electrostatic force microscopy, *Nanotechnology* **20**, 395702 (2009).
4. C. Riedel, R. Arinero, Ph. Tordjeman, G. L  v  que, G. A. Schwartz, A. Alegria, and J. Colmenero, Nanodielectric mapping of a model polystyrene-poly(vinyl acetate) blend by electrostatic force microscopy, *Phys. Rev. E* **81**, 010801(R), (2010).
5. L. Fumagalli, G. Gramse, D. Esteban-Ferrer, M. A. Edwards, and G. Gomila, Quantifying the dielectric constant of thick insulators using electrostatic force microscopy, *Appl. Phys. Rev.* **96**, 183107 (2010).
6. M. J. Cadena, S. H. Sung, B. W. Boudouris, R. Reifengerger, and A. Raman, Nanoscale mapping of dielectric properties of nanomaterials from kilohertz to Megahertz using ultrascale cantilevers, *ACS Nano* **10**, 4062–4071 (2016).
7. D. T. Lee, J. P. Pelz, and B. Bhushan, Scanning capacitance microscopy for thin film measurements, *Nanotechnology* **17**, 1484–1491 (2006).
8. L. Fumagalli, G. Ferrari, M. Sampietro, and G. Gomila G, Dielectric-constant measurement of thin insulating films at low frequency by nanoscale capacitance microscopy, *Appl. Phys. Lett.* **91**, 243110 (2007).
9. L. Fumagalli, G. Ferrari, M. Sampietro, and G. Gomila, Quantitative nanoscale dielectric microscopy of single-layer supported biomembranes, *Nano Lett.* **9**, 1604 (2009).
10. R. Shao, S. V. Kalinin, and D. A. Bonnell, Local impedance imaging and spectroscopy of polycrystalline ZnO using contact atomic force microscopy, *Appl. Phys. Lett.* **82**, 1869 (2003).
11. L. S. C. Pingree and M. C. Hersam, Bridge-enhanced nanoscale impedance microscopy, *Appl. Phys. Lett.* **87**, 233117 (2005).
12. K. Lai, M. B. Ji, N. Leindecker, M. A. Kelly, and Z. X. Shen, Atomic-force-microscope-compatible near-field scanning microwave microscope with separated excitation and sensing probes, *Rev. Sci. Instrum.* **78**, 063702 (2007).
13. K. Lai, W. Kundhikanjana, M. A. Kelly, Z. X. Shen, Nanoscale microwave microscopy using shielded cantilever probes, *Appl. Nanosci.* **1**, 13 (2011).
14. Y. Cho, S. Kazuta, and K. Matsuura: Scanning nonlinear dielectric microscopy with nanometer resolution, *Appl. Phys. Lett.* **72**, 2833 (1999).
15. Y. Cho, Scanning nonlinear dielectric microscopy, *J. Mater. Res.* **26**, 2007–16 (2011).
16. W. Lu, D. Wang, and L. Chen, Near-static dielectric polarization of individual carbon nanotubes, *Nano Lett.* **7**, 2729–33 (2007).

17. M. Fuhrmann, A. Musyanovych, Ronald Thoelen, and H. Moebius, Determination of the dielectric constant of non-planar nanostructures and single nanoparticles by electrostatic force microscopy, *J. Phys. Commun.* **6**, 125005 (2022).
18. M. Descoteaux, J. P. Sunnerberg, C. Staii, Quantitative characterization of dielectric properties of nanoparticles using electrostatic force microscopy, *AIP Advances* **10**, 115118 (2020).
19. D. E. Khoury, R. Arinero, J. -C. Laurentie, M. Bechelany, M. Ramonda, and J. Castellon, Electrostatic force microscopy for the accurate characterization of interphases in nanocomposites, *Beilstein J. Nanotechnol.* **9**, 2999–3012 (2018).
20. M. Houssat, C. Villeneuve-Faure, N. L. Dignat, and J. -P. Cambronne, Nanoscale mechanical and electrical characterization of the interphase in polyimide/silicon nitride nanocomposites, *Nanotechnology* **32**, 425703 (2021).
21. L. Fumagalli, D. Esteban-Ferrer, A. Cuervo, J. L. Carrascosa, and G. Gomila, Label-free identification of single dielectric nanoparticles and viruses with ultraweak polarization forces, *Nat. Mater.* **11**, 808–16 (2012).
22. D. Esteban-Ferrer, M. A. Edwards, L. Fumagalli, A. Juárez, and G. Gomila, Electric polarization properties of single bacteria measured with electrostatic force microscopy, *ACS Nano* **8**, 9843–9 (2014).
23. G. Gomila, J. Toset, and L. Fumagalli, Nanoscale capacitance microscopy of thin dielectric films, *J. Appl. Phys.* **104**, 024315 (2008).
24. G. Gomila, G. Gramse, and L. Fumagalli, Finite-size effects and analytical modeling of electrostatic force microscopy applied to dielectric films, *Nanotechnology* **25**, 255702 (2014).
25. L. Fumagalli, G. Gramse, D. Esteban-Ferrer, M. A. Edwards, and G. Gomila, Quantifying the dielectric constant of thick insulators using electrostatic force microscopy, *Appl. Phys. Lett.* **96**, 183107 (2010).
26. G. Gramse, G. Gomila, and L. Fumagalli, Quantifying the dielectric constant of thick insulators by electrostatic force microscopy: effects of the microscopic parts of the probe, *Nanotechnology* **23**, 205703 (2012).
27. L. Fumagalli, M. A. Edwards, and G. Gomila, Quantitative electrostatic force microscopy with sharp silicon tips, *Nanotechnology* **25**, 495701 (2014).
28. S. Hudlet, M. Saint Jean, C. Guthmann, and J. Berger, Evaluation of the capacitive force between an atomic force microscopy tip and a metallic surface, *Eur. Phys. J. B* **2**, 5-10 (1998).
29. J. Colchero, A. Cil, and A. M. Baro, Resolution enhancement and improved data interpretation in electrostatic force microscopy, *Phys. Rev. B* **64**, 245403 (2001).
30. E. Bonaccorso, F. Schonfeld, and H. -J. Butt, Electrostatic forces acting on tip and cantilever in atomic force microscopy, *Phys. Rev. B* **74**, 085413 (2006).
31. G. M. Sacha, E. Sahagún, J. J. and Sáenz, A method for calculating capacitances and electrostatic forces in atomic force microscopy, *J. Appl. Phys.* **101**, 024310 (2007).
32. M. Labardi, J. Barsotti, D. Prevosto, S. Capaccioli, C. M. Roland, and R. Casalini, Extended model for the interaction of dielectric thin films with an electrostatic force microscope probe, *J. Appl. Phys.* **118**, 224104 (2015).
33. S. Belaidi, P. Girard, and G. Leveque, Electrostatic forces acting on the tip in atomic force microscopy: Modelization and comparison with analytic expressions, *J. Appl. Phys.* **81**, 1023 (1997).
34. C. Riedel, R. Arinero, Ph. Tordjeman, M. Ramonda, G. Lévéque, G. A. Schwartz, D. G. de Oteyza, A. Alegria and J. Colmenero, Determination of the nanoscale dielectric constant by means of a double pass method using electrostatic force microscopy, *J. Appl. Phys.* **106**, 024315 (2009).
35. M. Azib, F. Baudoin, N. Binaud, C. Villeneuve-Faure, G. Teyssedre, F. Bugarin, and S. Segonds, Sensitivity analysis of the electrostatic interaction between the atomic force microscopy probe and a thin dielectric film with 3D-localized charge cloud, *J. Appl. Phys.* **125**, 045305 (2019).
36. L. Fumagalli, G. Ferrari, M. Sampietro, and G. Gomila, Quantitative Nanoscale Dielectric Microscopy of Single-Layer Supported Biomembranes, *Nano Letters* **9**, 1604-1608 (2009).
37. G. Stan, C. V. Ciobanu, and S. W. King, Resolving the subsurface structure and elastic modulus of layered films via contact resonance atomic force microscopy, *ACS Appl. Mater. Interfaces* **14**, 55238–55248 (2022).
38. A. Boularas, F. Baudoin, G. Teyssedre, and C. Villeneuve-Faure, 3D modeling of electrostatic interaction between atomic force microscopy probe and dielectric surface: impact of tip shape and cantilever contribution, *IEEE Transactions on Dielectrics and Electrical Insulation* **23**, 705 (2016).
39. O. Cherniavskaya, L. Chen, V. Weng, L. Yuditsky, and L. E. Brus, Quantitative Noncontact Electrostatic Force Imaging of Nanocrystal Polarizability, *J. Phys. Chem. B* **107**, 1525-1531 (2003).

40. M. Van Der Hofstadt, R. Fabregas, M. C. Biagi, L. Fumagalli, and G. Gomila, Nanoscale dielectric microscopy of nonplanar samples by lift-mode electrostatic force microscopy, *Nanotechnology* **27**, 405706 (2016).
41. H. Lozano, R. Fabregas, N. Blanco-Cabra, R. Millán-Solsona, E. Torrents, L. Fumagalli, and G. Gomila, Dielectric constant of flagellin proteins measured by scanning dielectric microscopy, *Nanoscale* **10**, 19188 (2018).
42. R. I. Revilla, X. -J. Li, Y. -L. Yang, and C. Wang, Comparative method to quantify dielectric constant at nanoscale using atomic force microscopy, *J. Phys. Chem. C* **118**, 5556–5562 (2014).
43. A. Dols-Perez, G. Gramse, A. Calò, G. Gomila and L. Fumagalli, Nanoscale electric polarizability of ultrathin bilayers on insulating substrates by electrostatic force microscopy, *Nanoscale* **7**, 18327 (2015).
44. R. Fabregas and G. Gomila, Dielectric nanotomography based on electrostatic force microscopy: A numerical analysis, *J. Appl. Phys.* **127**, 024301 (2020).