Quantitative microscopy of nonlinear dielectric constant using a scanning evanescent microwave microscopy

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Quantitative characterization of dielectric nonlinearity in ferroelectric materials has been successfully performed using a scanning evanescent microwave microscope, and a key calibration coefficient for quantitative microscopy of nonlinear dielectric constant is derived. This unique technique has advantages of high spatial resolution and simultaneously accessing of other related properties such as dielectric constant and magnetoelastic coefficient. Samples of LiNbO3 single crystal and PbTiO3 thin film are measured, which demonstrates that this technique can access the nonlinear dielectric constant of a microregion with a sensitivity of $1.0 \times 10^{-21}$ F/V.

Dielectric nonlinearity is a key property of the ferroelectric materials in various important applications. Recent progresses in ferroelectric thin film deposition and artificial ferroelectric domain structures have attracted many attentions. Quantitative characterization of the ferroelectric microstructure is critical for material investigation and device development. Recently, several types of scanning probe microscopes have been developed to image ferroelectric structures. Among them, scanning evanescent microwave microscope (SEMM) has the advantages of submicron resolution, no destruction, and operating in the same frequency range as wireless communication. However, the lack of quantitative theory limited the applications of SEMM in the microscopy of ferroelectric microstructure. Although semiquantitative result has been achieved in the pioneering works through calibration, a deep understanding between the nonlinear dielectric constant and the microscopy response is obviously necessary. In this letter, we will report our analysis on the field distribution and its contribution to nonlinear signal. An analytic relationship between the microscopy signal and the nonlinear dielectric constant was obtained from the analysis. We also compared the theoretical result with experiments’ measurement and the agreement confirmed the theoretical analysis.

The schematic of our microscope is shown in Fig. 1. The microwave frequency is about 2.5 GHz. The measurement is carried out in the soft contact mode as described in Refs. 11 and 13.

For ferroelectric material, the effective dielectric constant can be expressed as

$$\varepsilon_{ij}^{\text{eff}} = \varepsilon_{ij} + \varepsilon_{ij}^{\text{mod}} E_k^1 + E_k^m,$$

(1)

where $E_k^m$ is the applied modulation field, $E^1$ is the microwave electric field in the sample generated by the tip, and $\varepsilon_{ij}$ and $\varepsilon_{ijk}^{\text{mod}}$ are the linear and nonlinear dielectric constants, respectively. Here, we ignored the high order nonlinear effects, as their contribution is much smaller than that of the first order at the applied voltage.

By rewriting the frequency perturbation in the tensor format and substituting in Eq. (1), the resonant frequency modulations can be expressed as

$$\frac{\Delta f}{f_0} = -\frac{1}{2W} \int \sum_{ij} E_i^0 \Delta \varepsilon_{ij} E_j^1 + E_j^0 \int \sum_{ij} \varepsilon_{ij}[(\varepsilon_{ij} - \varepsilon_0) \delta_{ij} + \varepsilon_{ijk}^{\text{mod}} E_k^m] E_j^1 \, dv,$$

(2)

where $f_0$ and $f_r$ refer to the resonant frequencies before and after perturbation, respectively. $E^0$ is the microwave electric field without sample perturbation, $\varepsilon_0$ is the dielectric con-

![FIG. 1. (Color online) System setup of SEMM for nonlinear dielectric measurement. A signal generator and a lock-in amplifier are integrated into the system.](https://appliedphysics.org/aps/journal/apl/2006/89/4/044102-1/fig1.jpg)
stant of the air, and $W$ is the microwave energy stored in the probe cavity.

Under the isotropic assumption $\varepsilon_{ik}^* = \varepsilon_{333} \delta_{ik}$, the first harmonic detected by lock-in amplifier will be

$$V_{\text{lock-in}} = S(\Delta f) \Omega = -\frac{Sf_0}{2W} \varepsilon_{333} \int 3 \sum_{i=0}^{3} E_i E_i^{\text{mod}} dV,$$

(3)

where $S$ is the sensitivity of the SEMM on the resonant frequency shift.

Following our previous work, we adopt the concept of average modulation field $\bar{E}^{\text{mod}}$, which is the electric field at the tip-sample contact point (the maximum electric field inside the sample) divided by a coefficient $C$:

$$\bar{E}^{\text{mod}} = \frac{1}{CR_0} \frac{\varepsilon_{33} + \varepsilon_0}{2} V_{\text{mod}},$$

(4)

so Eq. (3) can be written as

$$V_{\text{lock-in}} = -SAf_0 \frac{\varepsilon_{33} \varepsilon_0}{\varepsilon_{33}} \bar{E}^{\text{mod}} = -\frac{SAf_0}{CR_0} \frac{\varepsilon_{33} + \varepsilon_0 \varepsilon_{33}}{2\varepsilon_0 \varepsilon_{33}} V_{\text{mod}},$$

$$\frac{1}{C} = \frac{\varepsilon_{33} + \varepsilon_0}{AW \varepsilon_{33} + \varepsilon_0} \frac{R_0}{V_{\text{mod}}} \int 3 \sum_{i=0}^{3} E_i E_i^{\text{mod}} dV,$$

(5)

where $R_0$ is the tip radius, $V_{\text{mod}}$ is the modulation amplitude of the applied ac voltage, and $A$ is a constant determined by the system configuration. In this phenomenological expression, the coefficient $C$ is a key parameter for the quantitative microwave microscopy of the nonlinear dielectric constant. We will theoretically calculate it by integrating the electric field contributions inside the sample.

For bulk materials, the electric field inside the sample can be expressed as

$$E^1 = \frac{4\pi \varepsilon_0 R_0 V_0}{2\pi(\varepsilon_{33} + \varepsilon_0)} \sum_{m=1}^{\infty} \frac{1}{b_{m-1}} \varepsilon_y \varepsilon_z + (\varepsilon + R_0/m)e_z, \frac{x^2 + y^2 + (z + R_0/n)^2}{3/2},$$

$$E^{\text{mod}} = \frac{4\pi \varepsilon_0 R_0 V_0}{2\pi(\varepsilon_{33} + \varepsilon_0)} \sum_{m=1}^{\infty} \frac{1}{b_{m-1}} \varepsilon_y \varepsilon_z + (\varepsilon + R_0/m)e_z, \frac{x^2 + y^2 + (z + R_0/n)^2}{3/2},$$

(6)

where $e_x$, $e_y$, and $e_z$ are unit vectors along the directions of $x$, $y$, and $z$, respectively. $V_0$ is the open end peak voltage of the microwave probe cavity, $b = (\varepsilon_{33} - \varepsilon_0)/(\varepsilon_{33} + \varepsilon_0)$. Substituting Eq. (6) into Eq. (5), we have

$$\frac{1}{C} = \frac{\varepsilon_0^2 \varepsilon_{33}}{\pi(\varepsilon_{33} + \varepsilon_0)} \sum_{m=1}^{\infty} \frac{1}{b^m} + 2 \left[ J_1(m,n) + J_2(m,n) \right],$$

(7)

where $J_1$ and $J_2$ are two ellipsoidal integrations:

$$J_1(m,n) = \int \int \int \frac{x^3}{x^2 + y^2 + (z + 1/n)^2 2/3} \frac{1}{(z + 1/n)(\varepsilon + 1/m)^2} \frac{(z + 1/m)^2}{x^2 + y^2 + (z + 1)^2} \frac{3/2}{3/2} dV,$$

$$J_2(m,n) = \int \int \int \frac{1}{x^2 + y^2 + (z + 1/n)^2 2/3} \frac{(z + 1/m)^2}{x^2 + y^2 + (z + 1)^2} \frac{1}{3/2} dV.$$

(8)

Thus, the coefficient $C$ can be obtained by using numerical method according to Eqs. (7) and (8). The calculated result is shown in Fig. 2, which can be fitted as a function of the relative dielectric constant $(\varepsilon_{33}/\varepsilon_0)$ of the sample:

$$C = 38.9225 + 3.032 \frac{\varepsilon_{33}}{\varepsilon_0} - 0.9494 \left( \frac{\varepsilon_{33}}{\varepsilon_0} \right)^{1.5}$$

$$+ 0.0027 \left( \frac{\varepsilon_{33}}{\varepsilon_0} \right)^2,$$

(9)

For thin films, analytic expression of the electric field distribution similar to Eq. (6) is not available due to the complicated tip-film-substrate interaction. However, the electric field can still be obtained by integrating the contribution of every image charge and so does the coefficient $C$. Unfortunately, as too many image charges are involved in the integration, thin film can only be handled case by case at

FIG. 2. (Color online) Coefficient $C$ as a function of relative dielectric constant for bulk materials.
current stage. More efficient algorithm is still under development.

Shown in Fig. 3 is the nonlinear signal as a function of modulation voltage measured on a LiNbO$_3$ single crystal. The single crystal surface is $c$ cut, and the spontaneous polarization is perpendicular to the surface. As expected, the nonlinear dielectric signal increases linearly with the modulation voltage amplitude. The slope of the best fit $V_{\text{lock-in}}/V_{\text{mod}}$ gives a nonlinear dielectric constant of $-2.55 \times 10^{-19}$ F/V.

A dc bias voltage is also applied to the PbTiO$_3$ thin film by using a homemade voltage amplifier. The ac modulation voltage amplitude is set to 5 V and the dc bias voltage is scanned from −100 to 100 V. The butterfly-type hysteresis loop of the nonlinear signal as a function of the maximum dc electric field inside the film (the electric field at the tip-film contact point) is shown in Fig. 5. (Here the maximum electric field is used since the rotation, reorientation, or nucleation of ferroelectric domain mainly depends on the maximum field instead of the effective field.) From the figure, the coercive field is about 95 kV/cm, in approximate agreement with the value reported elsewhere (115 kV/cm).  

In summary, we have successfully performed quantitative characterization of dielectric nonlinearity with a scanning evanescent microwave microscope. This technique has the advantages of nondestruction and high spatial resolution. It can also be used to obtain other related properties simultaneously. These advantages enable spatially resolved mapping of ferroelectric domains and combinatorial screening of novel ferroelectric materials.

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