

## Thickness dependence of the effective dielectric constant in a thin film capacitor

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The static value of the effective dielectric constant in a thin film capacitor is simulated by means of the local field theory. The value of it shows a sharp decrease as the film thickness is decreased in an ultrathin film geometry. This phenomenon is due to the size effect intrinsic to a thin film structure and has nothing to do with the material aspect. The decrease is more remarkable for larger values of the bulk dielectric constant. It is recovered by inserting interface layers with larger atomic polarizability between the film and the capacitor electrode. © 1998 American Institute of Physics. [S0003-6951(98)01031-6]

As a higher integration of dynamic random access memory is pursued, the possibility of applying high dielectric materials such as (Ba, Sr)TiO<sub>3</sub> to a high density memory cell capacitor has been investigated with a great deal of interest.<sup>1-5</sup> It is frequently reported, however, that the dielectric constant of such a material decreases as the film thickness is decreased.<sup>1-3</sup> The reason for this phenomenon is usually ascribed to a degradation in film quality. The film quality degrades as the thickness is decreased, yielding a smaller dielectric constant. Or, low dielectric constant interface layers are formed adjacent to the electrode, and they dominate the capacitance as the film thickness is decreased.

In this study, we have simulated the effective dielectric constant of a thin film capacitor and have found a strong dependence of the effective dielectricity on the thickness of the material. The thickness dependence in our simulation has nothing to do with the film quality change, but is an intrinsic effect due to the presence of a film surface.

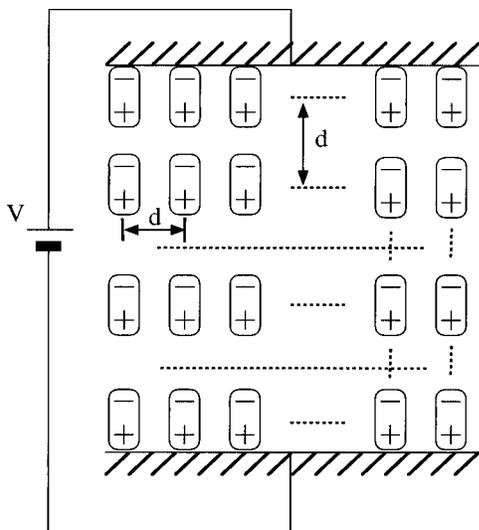


FIG. 1. The simulated capacitor structure. The dielectric film consists of atomic dipole moments arranged on lattice points of a simple cubic lattice with infinite extension along the  $x$  and  $y$  axis and with finite dipole layers along the  $z$  axis.

According to the Lorentz's local field theory,<sup>6</sup> the dielectric constant of the bulk material,  $K\epsilon_0$ , where  $\epsilon_0$  is the permittivity of a vacuum, is related to the atomic polarizability  $\alpha$  of the constituent dipole moment, by the Clausius-Mossotti (CM) relation

$$\frac{K-1}{K+2} = \frac{1}{3\epsilon_0} N\alpha, \quad (1)$$

where  $N$  is the number density of the dipole.

The effective dielectric constant of a thin film capacitor is simulated by means of the local field theory as follows. The sample structure consists of atomic dipole moments arranged on lattice points of a simple cubic lattice (periodicity  $d$ ) with an infinite extension along the  $x$  and  $y$  axis and with finite dipole layers ( $n$  layers) along the  $z$  axis (Fig. 1). The  $i$ th layer atomic dipole moment  $p(i)$  is equal to  $(4\pi\epsilon_0 d^3)\alpha(i)E_{\text{loc}}(i)$  in the linear approximation, where  $\alpha(i)$  and  $E_{\text{loc}}(i)$  are, respectively, the dimensionless atomic polarizability and the local field at the  $i$ th layer atomic site. All dipole moments in the sample yield the dipole field at each atomic site, and the dipole field as well as the field due to the capacitor plate charge constitutes the local field thereof. The contribution to the local field at an  $i$ th layer site from all of the  $j$ th layer dipole moments is expressed as  $(1/4\pi\epsilon_0 d^3)G_{ij}p(j)$ . The coupling factor  $G_{ij}$  is computed by

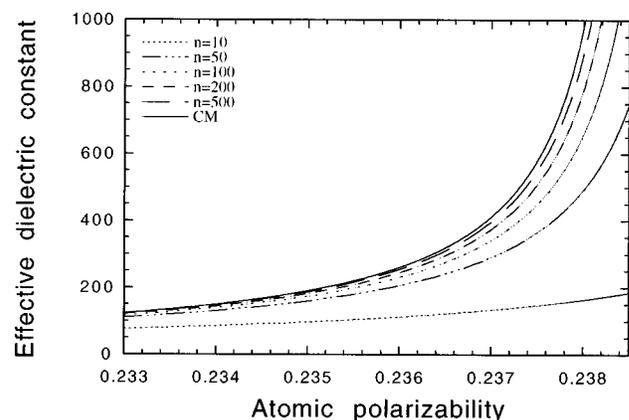


FIG. 2. The effective dielectric constant  $K_{\text{eff}}$  as a function of the uniform value of the dimensionless atomic polarizability,  $\alpha(i) \equiv \alpha_0$ . The parameter is  $n$ , the thickness of the sample. CM stands for the Clausius-Mossotti relation.

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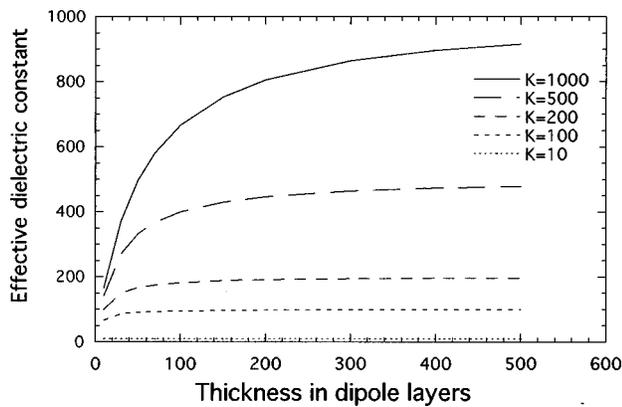


FIG. 3. The thickness,  $n$ , dependence of the effective dielectric constant  $K_{\text{eff}}$ . The parameter is the bulk dielectric constant.

summing up the contribution from all the dipole moments in the  $j$ th layer as  $G_{ij} = d^3 \sum (3 \cos^2 \theta - 1)/r^3$ ,<sup>6</sup> where the radial vector from one of the  $j$ th layer dipoles to the  $i$ th layer site has the magnitude  $r$  and forms an angle  $\theta$  with the dipole moment direction. The summation over the  $j$ th layer dipoles farther than a definite distance  $R$  from a  $j$ th layer site equivalent to the  $i$ th layer site can be approximated by an integration. A sufficiently large  $R$  is chosen so that  $G_{ij}$  does not depend on  $R$ . The local field  $E_{\text{L}}(i)$  is expressed as  $(Q/\epsilon_0)\tilde{E}_{\text{L}}(i)$  where  $Q$  is the charge density on the capacitor plate and is assumed to be equal to the electric flux density  $D(i)$  in our geometry, and the dimensionless local field  $\tilde{E}_{\text{L}}(i)$  is a solution of the simultaneous equation,

$$\sum_j [\delta_{ij} - G_{ij}\alpha(j)]\tilde{E}_{\text{L}}(j) = 1. \quad (2)$$

$\delta_{ij}$  equals unity for  $i=j$  and vanishes otherwise. The macroscopic polarization  $P(i)$  is given by  $p(i)/d^3$ , and the macroscopic field is equal to  $[D(i) - P(i)]/\epsilon_0$  at the  $i$ th layer site. The local dielectric constant  $K_{\text{L}}(i)\epsilon_0$  at the  $i$ th layer, which is defined by the ratio of the electric flux density and the macroscopic field thereof, is evaluated as  $K_{\text{L}}(i) = [1 - 4\pi\alpha(i)\tilde{E}_{\text{L}}(i)]^{-1}$ . The effective dielectric constant  $K_{\text{eff}}\epsilon_0$ , which is defined by the capacitance of the thin film capacitor  $C = Q/V \equiv K_{\text{eff}}\epsilon_0/(nd)$ , is expressed as

$$K_{\text{eff}} = \left[ 1 - \frac{1}{n} \sum_{i=1}^n 4\pi\alpha(i)\tilde{E}_{\text{L}}(i) \right]^{-1}. \quad (3)$$

Notice that  $K_{\text{eff}}$  is related to  $K_{\text{L}}(i)$  by  $K_{\text{eff}}^{-1} = [\sum_{i=1}^n K_{\text{L}}(i)^{-1}]/n$ .

Figure 2 shows the simulated effective dielectric constant  $K_{\text{eff}}$  as a function of the uniform atomic polarizability  $\alpha(i) \equiv \alpha_0$ . The parameter is  $n$ , the thickness of the sample. The bulk dielectric constant  $K$  given by the CM relation is also shown. It is clear that the effective dielectric constant increases to the CM result as the thickness increases. Figure 3 shows the thickness dependence of the effective dielectric

TABLE I. Examples of the value of  $K_{\text{eff}}$ .

$n$	$K=10$	$K=1000$
10	9.602	164.2
500	9.992	912.7
$\infty$	10.000	1000.0

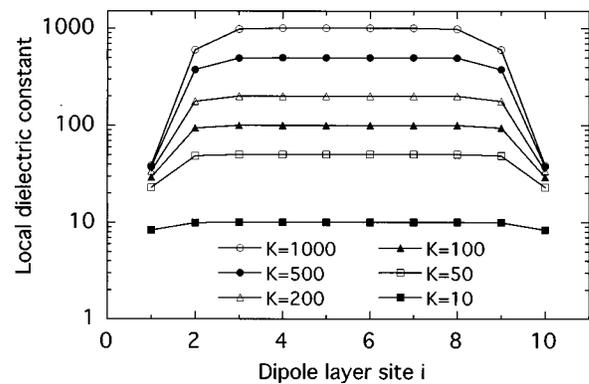


FIG. 4. The local dielectric constant distribution along the thickness direction. The thickness is  $n=10$ .

constant. The parameter is the atomic polarizability converted to  $K$  by the CM relation. The effective dielectric constant sharply decreases as the film thickness is decreased, and the decrease is more remarkable for larger  $K$  values. Some of the numerical values are summarized in Table I. Notice that the thickness dependence is almost negligible for samples with  $K \leq 10$ . Figure 4 shows the local dielectric constant distribution along the thickness direction for the case of  $n=10$ . It is clear that the local dielectric constant at the edge site has a reduced value due to the absence of an enforcing field effected by the nearest neighbor dipole in the adjacent layer, which fact yields a smaller effective dielectric constant in a thinner sample structure. The local dielectric constant has a bulk value deep in the film. Figure 5 shows that the decreased effective dielectric constant is recovered and is even enhanced by increasing the atomic polarizability of the edge layers. The parameter  $\beta$  is defined so that  $\alpha(1) = \alpha(n) = \beta\alpha_0$  as well as  $\alpha(i) = \alpha_0$  ( $2 \leq i \leq n-1$ ).

The effective dielectric constant of a thin film capacitor was simulated by means of Lorentz's local field theory. It was shown that the value decreases as the film thickness is reduced in ultrathin film geometry. This is due to the surface dipole moments suffering from different environmental conditions than the bulk dipole moments, and is due to the size effect intrinsic to the film structure. The decrease is more remarkable in samples with a larger bulk dielectric constant. The decreased value is recovered and is even enhanced by replacing the surface layers with layers having a larger atomic polarizability.

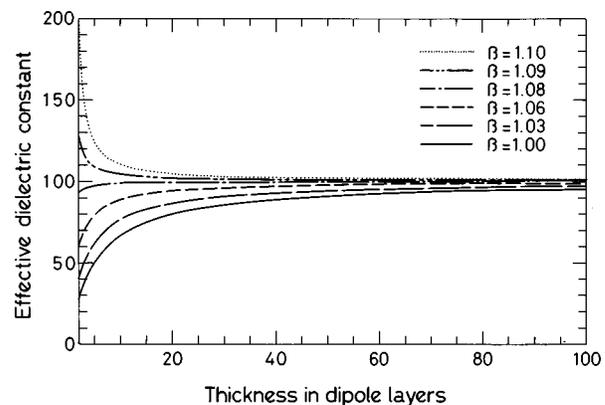


FIG. 5. The thickness dependence of the effective dielectric constant  $K_{\text{eff}}$ , where the atomic polarizability of edge layers is enhanced so that  $\alpha(1) = \alpha(n) = \beta\alpha_0$  and  $\alpha(i) = \alpha_0$  ( $2 \leq i \leq n-1$ ).  $K=100$ .

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<sup>6</sup>See standard textbooks, e.g., H. Fröhlich, *Theory of Dielectrics* (Clarendon, Oxford, 1958), or C. Kittel, *Introduction to Solid State Physics*, 7th ed. (Wiley, New York, 1996).