Mater.Phys.Mech.1 (2000) 45-48

MODELING OF SIZE EFFECT ON DIELECTRIC RESPONSE OF THIN FERROELECTRIC FILMS

O.G. Vendik and S.P. Zubko

Electronics Department, Electrotechnical University,
Prof. Popova 5, St. Petersburg 197376, Russia
Fax: +7 812 234-4809,

Received: September 30, 1999

Abstract. The size effect in thin film sandwich structures is considered. Three types of boundary conditions for dynamic polarization at interface electrode-ferroelectric layer are formulated. The type of boundary conditions depends on matching crystal lattices of electrodes and ferroelectric layer. A model describing the dependence of dielectric permittivity on biasing field, temperature, and thickness of the thin ferroelectric film is proposed. The results of the simulation are in good agreement with experiments.

The dielectric nonlinearity of ferroelectrics allows to use these materials as the basis of electrically tunable devices. Thin film ferroelectrics are used at microwaves. If the thickness of the film is comparable with the correlation radius the size effect appears in the film, i.e. the dielectric permittivity of film depends on its thickness. The size effect is related with the spatial distribution of polarization inside the ferroelectric film and type of the boundary conditions for polarization. The size effect results in a decrease in the polarization and therefore, in reduced dielectric permittivity and tunability. Through the selection of electrode material one can control the size effect on dielectric response of the thin ferroelectric films.

The dielectric permittivity of ferroelectric bulk material is a function of biasing field and temperature. The model of dependence of dielectric permittivity on biasing field and temperature is based on Ginsburg-Devonshire expansion of free energy density in the power series over the order parameter. For ferroelectrics the order parameter is a spontaneous polarization [1-4].

The phenomenological model of the dielectric permittivity of a bulk sample is presented by the following equation:

$$\varepsilon^{-1} = \varepsilon_{00}^{-1}\left\{\left[(\xi^2+\eta^3)^{1/2}+\xi\right]^{2/3} + \left[(\xi^2+\eta^3)^{1/2}-\xi\right]^{2/3} - \eta\right\} \quad (1)$$

where

$$\xi(U_B) = \sqrt{\xi_S^2 + \xi_B^2}\,, \quad (2)$$

$$\eta(T) = \frac{\theta_F}{T_C}\sqrt{\frac{1}{16}+\left(\frac{T}{\theta_F}\right)^2} - 1, \quad (3)$$

$$\xi_B = \frac{U_B}{E_N h}, \qquad E_N = \frac{2D_N}{\varepsilon_0(3\varepsilon_{00})^{3/2}}, \quad (4)$$

ε_{00} is the permittivity of a free space, h is the thickness of ferroelectric film included in sandwich capacitor represented in Fig. 1, U_B is a biasing voltage.

Model parameters: ε_{00} is the analogue to Curie-Weiss constant; T_C is the effective Curie temperature; θ_F is the effective Debye temperature of the sublattice oscillations causing ferroelectric polarization; E_N is the

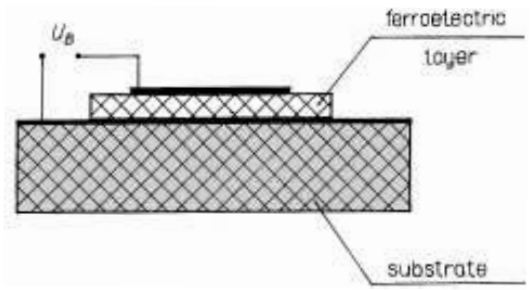

Fig. 1. Sandwich capacitor as a simplified general structure.

Corresponding author: S.P. Zubko, e-mail: SPZubko@eltech.ru

© 2000 Advanced Study Center Co. Ltd.

Table 1. Model parameters for single crystal $SrTiO_3$.

	T_C (K)	ε_{00}	E_N(kV/cm)	θ_F(K)	ξ_S
single crystal $SrTiO_3$ [5]	42	2081	19.3	175	0.018

normalizing electric field; ξ_S is the statistical dispersion of the biasing field characterizing the quality of material. The numerical values of model parameters for bulk material [5] are presented in Table 1.

The dependence of dielectric permittivity of the film on its thickness should be included in the model. In order to take into account the size effect, it is necessary to solve a second-order differential equation with respect to polarization. This equation is a consequence of Ginsburg-Devonshire expansion [6]:

$$-2\lambda_1 \frac{d^2P(x)}{dx^2} + \frac{D(x)}{\varepsilon(T)} + \frac{D^3(x)}{D_N^2} = \varepsilon_0 E(x), \qquad (5)$$

$$\varepsilon(T) = \frac{\varepsilon_{00}}{\eta(T)} \qquad (6)$$

where *P(x), D(x), E(x)* are polarization, displacement, and electric field; λ_1 is a correlation parameter; *x* axis is directed normally to the electrode-ferroelectric film interface.

The numerical values of the correlation parameter determined from the experimental data on inelastic neutron scattering on soft mode of ferroelectric crystal are presented in Table 2 [7,8]. The film thickness above which the size effect appears is a correlation radius of ferroelectric polarization which can be extracted from the equation (5).

Table 2. Numerical values of correlation parameter.

λ_1	$SrTiO_3$	$KTaO_3$	$BaTiO_3$*
μm^2	$1.15\cdot10^{-7}$	$5.37\cdot10^{-7}$	10^{-9}

* Preliminary estimation

$$r_c(T) = \sqrt{2\lambda_1 \varepsilon(T)}. \qquad (7)$$

For different crystal structures of electrodes three different types of boundary conditions at electrode-ferroelectric interface can be realized. The spatial distribution of polarization corresponding to a certain type of boundary conditions induces the size effect in the film.

There are three types of boundary conditions for dynamic polarization [9]:

(I) Zero boundary conditions:

$$P_{ac}(x)|_{x=\pm h/2} = 0 \qquad h_{eff} = h$$

(II) Intermediate boundary conditions:

$$\left(P_{ac}(x) \pm b\frac{dP_{ac}(x)}{dx}\right)|_{x=\pm h/2} = 0 \qquad h < h_{eff} < \infty$$

(III) Free boundary conditions:

$$\frac{dP_{ac}(x)}{dx}|_{x=\pm h/2} = 0 \qquad h_{eff} \to \infty.$$

The boundary conditions of the type (I) are refered to a sandwich capacitor with electrodes made of a normal metal. The polarization distribution provided by these boundary conditions causes the size effect. The boundary conditions of the type (II) are refered in a sandwich capacitor with $YBa_2Cu_3O_{7-x}$ (YBCO) electrodes. In this case polarization can partially penetrate inside the electrodes. To allow the better understanding of that, the effective film thickness h_{eff} was introduced. Free boundary conditions (type III) are realized in structures with $SrRuO_3$ (SRO) electrodes. The spatial distribution of polarization inside the ferroelectric layer for three types of boundary conditions is shown in Fig. 2.

Having solved equation (5) with zero boundary conditions, one can derive the expression for dynamic polarization taking into account the size effect:

$$P_{ac}(x) = P_{ac}(0)\left[1 - \frac{ch(\alpha x)}{ch(\alpha h/2)}\right],$$

$$P_{ac}(0) = \frac{Q_{ac}}{S}\left[1 - \frac{1}{\varepsilon(T)} - 3\left(\frac{Q_{dc}}{D_N S}\right)^2\right]$$

where Q_{ac} and Q_{dc} are the alternating and direct current components of the charge at the electrodes respectively; *S* is the area of the electrodes.

The inverse effective dielectric permittivity of a thin ferroelectric film is:

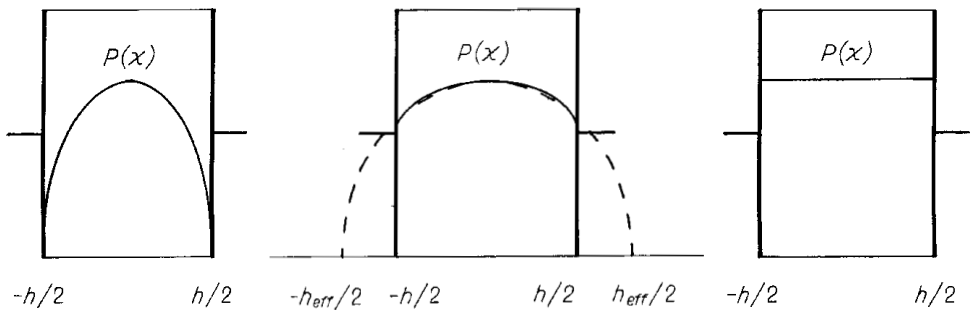

Fig. 2. Spatial distribution of dynamic polarization inside ferroelectric layer in the case: a — zero boundary conditions; b — intermediate boundary conditions; c — free boundary conditions.

$$\varepsilon^{-1} = \varepsilon_{00}^{-1}\left\{\left[(\xi^2+\eta^3)^{1/2}+\xi\right]^{2/3}+\left[(\xi^2+\eta^3)^{1/2}+\xi\right]^{2/3} - \eta + a^2\right\} \quad (8)$$

where the parameter of size effect is $a = \sqrt{2\varepsilon_{00}/\alpha h_{eff}}$; $\alpha = 1/\sqrt{2\lambda_1}$.

The numerical values of the model parameters obtained for various sandwich structures are presented in Table 3. Fig. 3 shows good agreement between experimental and model dependencies calculated using model parameters from Table 3. Curves in Fig. 3 illustrate influence of size effect on dielectric permittivity. In the ferroelectric capacitors with YBCO electrodes dynamic polarization partially penetrates inside electrodes and influence of the size effect in this case is attenuated.

Experimental [13] and model dependencies of dielectric permittivity of thin film $Ba_{0.12}Sr_{0.88}TiO_3$ (BSTO) included in sandwich capacitors with Pt and SRO electrodes are presented in Fig. 4. At room temperature

Table 3. Model parameters for thin ferroelectric films.

Capacitor	$h(\mu m)$	$h_{eff}(\mu m)$	ε_{00}	$T_C(K)$	$\theta_F(K)$	$E_N(kV/cm)$	ξ_S	a	
YBCO/STO/Au [10]	0.250	0.230	4265	34	152	6.0	2.3	16	(I)
Pt/STO/Ni [11]	0.046	0.037	4143	40	152	8.7	1.5	7	(I)
YBCO/STO/YBCO [12]	0.800	2.300	3090	35	152	6.5	0.8	1.2	(II)
SRO/BSTO/SRO [13]	0.020	∞	4400	42	175	12	1.0	0	(III)
SRO/BSTO/Pt [13]	0.020	0.37	4400	42	175	12	1.0	3	(II)

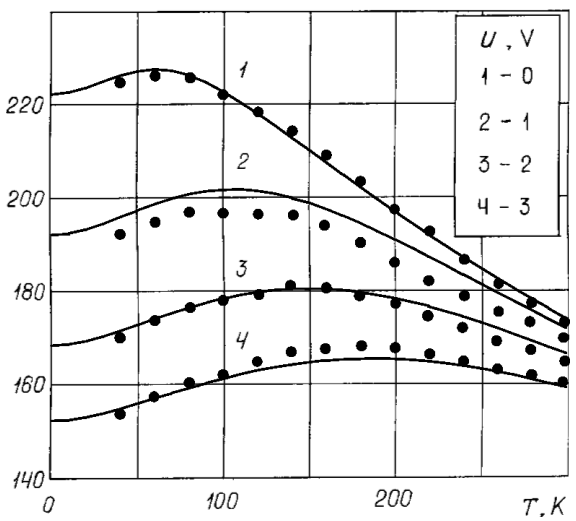

Fig. 3. Experimental (points) [10] and model dependencies (solid lines) of effective dielectric constant of $SrTiO_3$ film on temperature and biasing field.

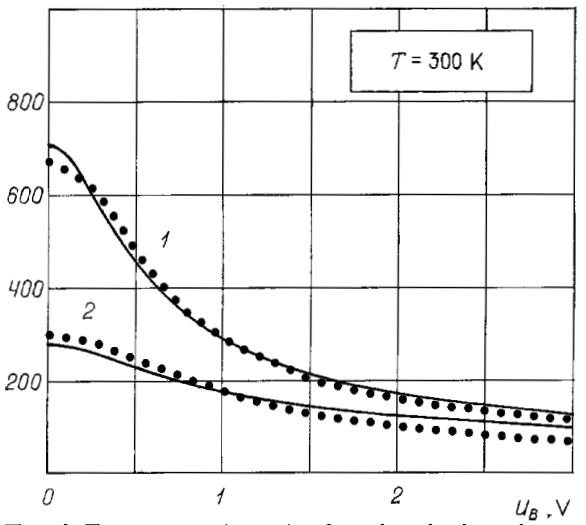

Fig. 4. Experimental (points) [13] and model dependencies (solid lines) of effective dielectric constant on biasing voltage: 1 — $SrRuO_3/Ba_{0.12}Sr_{0.88}TiO_3/SrRuO_3$; 2 — $SrRuO_3/Ba_{0.12}Sr_{0.88}TiO_3/Pt$.

effective permittivity of the film in capacitor with SRO electrodes is higher than permittivity of single crystal in about 2.5 times. One can conclude that in the case of good matching of crystal lattices of electrodes and ferroelectric film the size effect is suppressed and free boundary conditions for dynamic polarization are realized. Tunability of such capacitor is more better than tunability of the capacitor with Pt electrode.

Thus interface between thin ferroelectric film and electrodes determines distribution of polarization inside ferroelectric layer and therefore variation of dielectric characteristics of sandwich capacitor. By chosen electrodes one can suppress size effect in thin film structures.

REFERENCES

[1] V.L. Ginsburg // *Zh. Eksp. Theor. Fiz.* **19** (1949) 36.
[2] A.F. Devonshire // *Phil. Mag.* **40** (1949) 1040.
[3] O.G. Vendik // *Fiz. Tverd. Tela* **14** (1972) 989.
[4] O.G. Vendik and S.P. Zubko // *J. Appl. Phys.* **82** (1997) 4475.
[5] K. Bethe // *Philips Research Report*, Supplement, No. 2 (1970) 1.
[6] O.G. Vendik and L.T. Ter-Martirosyan // *Fiz. Tverd. Tela* **36** (1994) 3343.
[7] O.G. Vendik and I.G. Mironenko // *Fiz. Tverd. Tela* **16** (1974) 3445.
[8] S.P.Zubko // *Tech. Phys. Lett.* **24** (1998) 839.
[9] O.G. Vendik, S.P. Zubko and L.T. Ter-Martirosayn // *Appl. Phys. Lett.* **73** (1998) 37.
[10] F.A. Miranda, C.H. Mueller, G.A. Koepf and R.M. Yandrofski // *Supercond. Sci. Technol.* **8** (1995) 755.
[11] S. Komatsu and K. Abe // *Jpn. J. Appl. Phys.* **23** Pt. 1 (1995) 3597.
[12] A.T. Findikoglu, C. Doughty and S.M. Anlage // *Appl. Phys. Lett.* **63** (1993) 3215.
[13] M. Izuha, K. Abe and N. Fukushima // *Jpn. J. Appl. Phys.* **36**, Pt.1 (1997) 5866.