Hysteresis Behavior and Pyroelectric Properties of Multi-Surface Ferroelectric Thin Films

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Using the transverse Ising model, we investigate the dielectric properties and the hysteresis behavior of multi-surface ferroelectric (MSF) thin films in the framework of the effective field theory based on the probability distribution technique that accounts for the self-spin correlation functions. The effects of the exchange interactions and the transverse field on the longitudinal and the transverse polarizations, the dielectric susceptibility, the pyroelectric coefficient, and the hysteresis loops are studied. A number of interesting phenomena have been found.

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I. INTRODUCTION

Ferroelectric thin films have attracted much attention both experimentally [1–3] and theoretically [4–8], because of their physical properties and possible technological applications, such as high dielectric capacitors, non-volatile memories with low switching voltage, infrared sensors, electro-optical devices, surface acoustic wave, delay lines, pyroelectric sensors, optical shutters, and modulators [9–15]. Experimentally, with the development of characterization technology for nanostructured systems, many researchers have invested in their studies. Indeed, Wang et al. [16] and Molotskii et al. [17] have analyzed the ferroelectric thin films by atomic force microscopy (AFM). They have revealed that AFM can induce an electric field and generate ferroelectric domains in ferroelectric thin films. Moreover, by using scanning force microscopy, Shvartsman et al. [18] have studied the asymmetric nanoscale switching and nonlinear local piezoelectric deformation in ferroelectric thin films.

Theoretically, a good candidate for describing the H-bonded ferroelectric systems is generally believed to be the transverse Ising model (TIM) within the framework of the pseudo-spins theory. The TIM was first applied by de Gennes [19] to study the order and disorder ferroelectric phase transitions. The model assumes ferroelectrics to consist of pseu-
dospins with exchange interactions in a transverse field. Despite its simplicity, the TIM is successful in studying the microscopic properties of ferroelectrics under many approximations [20–32], as well as, the Green function method [33–38], the effective field theory [39], the Monte Carlo simulations [40], the Ginzburg–Landau phenomenological theory [41], and the transfer-matrix method [42]. Using the mean field approximation (MFA), Wang et al. [22–24] and Sy [25–27] have studied the surface size effects on the ferroelectric phase transitions and pyroelectric properties of a ferroelectric bilayer and superlattice. Lu [28] has investigated in detail under the usual MFA the effects of the surface modification on the critical behavior in multiple-surface-layer ferroelectric thin films. Taking into account surface transition layers (STLs), Sun et al. [29] have studied the phase transformation and pyroelectric properties of ferroelectric thin films by employing the same approximation. The distribution functions representing the intra- and inter-layer couplings between the two nearest neighbors pseudo-spins are introduced to characterize STLs. Compared with the results obtained by the traditional treatments for the thin films using only the single surface transition layer (SSL), they have shown that the STL model reflects a more realistic and comprehensive situation of films.

Furthermore, Essaoudi et al. [30] and Oubelkacem et al. [31] have studied the effects of surface transition layers on the phase diagrams, the pyroelectric, and the dielectric properties of ferroelectric thin films using the TIM. Recently, Cui et al. [43] have discussed the influence of the surface transition layer on the switching time and coercive field under a step electric field. They have shown that the surface transition layer plays a crucial role in the properties of reversal polarization in a ferroelectric thin film. Yang et al. [44], using the MFA and Green function methods, have studied in detail the dependence of the layers and the average polarizations of the multi-surface ferroelectric thin film on the transverse field and on the three different exchange interaction parameters.

Our aim in this paper is to study the surface effects on the dielectric properties, hysteresis loops, and pyroelectric behaviors of the MSF thin film. This system is described by the transverse Ising model, using the effective field theory with the probability distribution technique, which is believed to give more exact results than those of the standard MFA [45]. In Section II, we outline the theoretical formulation. In Section III, the numerical results for the layers and average polarizations, the pyroelectric coefficient, the dielectric susceptibility, and the hysteresis loops are presented. In Section IV, a brief conclusion is given.

II. FORMALISM

The geometric structure of the MSF thin film under study is shown in Fig. 1. We consider a ferroelectric thin film with pseudo-spin sites on a simple cubic lattice composed of \( L \) layers in the \( z \)-direction. The system is described by the Ising Hamiltonian in the presence of the external electric and transverse fields as follows:

\[
H = - \sum_{\langle ij \rangle} J_{i,j} S_{i,z} S_{j,z} - 2 \mu E \sum_i S_{i,z} - \sum_{i,n} \Omega_n S_{i,x},
\] (1)
where $S_{i,z}$ and $S_{i,x}$ denote the $z$ and $x$ components of a quantum spin $\vec{S}_i$ of magnitude $S = \frac{1}{2}$ at site $i$. $J_{i,j}$ is the exchange interaction constant between the spins on the neighboring sites $i$ and $j$. $\mu$ is the dipole moment of site $i$, $E$ is the external electric field, and $\Omega_n$ is the transverse field. We allow $\Omega_n$ to be different in the surface layers; $\Omega_n = \Omega_s$ for $n$ in the surface layers and $\Omega_n = \Omega$ otherwise. We introduce three different exchange interaction constants: $J_s$, $J_a$, and $J_e$ are the surface, the intra-layer, and inter-layer exchange interactions, respectively. The method used is the effective field theory, fully described in [45], that employed the probability distribution technique to account for the single-site spin correlations. Following that procedure and for a fixed configuration of neighboring

FIG. 1: Schematic illustration of the MSF thin film.
spins, we find that the layer longitudinal and transverse polarizations are given by

\[ p_{n,z} = \langle S_{n,z} \rangle = \left\langle f_z \left( \sum_j J_{i,j} S_{j,z} + 2 \mu E, \Omega_n \right) \right\rangle, \tag{2} \]

\[ p_{n,x} = \langle S_{n,x} \rangle = \left\langle f_x \left( \sum_j J_{i,j} S_{j,z} + 2 \mu E, \Omega_n \right) \right\rangle, \tag{3} \]

where \( n \) is the layer index. The functions \( f_z \) and \( f_x \) are given by

\[ f_z \left( \sum_j J_{i,j} S_{j,z} + 2 \mu E, \Omega_n \right) = \frac{\sum_j J_{i,j} S_{j,z} + 2 \mu E}{2 \sqrt{\left( \sum_j J_{i,j} S_{j,z} + 2 \mu E \right)^2 + \Omega_n^2}} \times \tanh \left( \frac{\beta}{2} \sqrt{\left( \sum_j J_{i,j} S_{j,z} + 2 \mu E \right)^2 + \Omega_n^2} \right), \tag{4} \]

\[ f_x \left( \sum_j J_{i,j} S_{j,z} + 2 \mu E, \Omega_n \right) = \frac{\Omega_n}{2 \sqrt{\left( \sum_j J_{i,j} S_{j,z} + 2 \mu E \right)^2 + \Omega_n^2}} \times \tanh \left( \frac{\beta}{2} \sqrt{\left( \sum_j J_{i,j} S_{j,z} + 2 \mu E \right)^2 + \Omega_n^2} \right), \tag{5} \]

with \( \beta = \frac{1}{k_B T} \) and \( T \) is the temperature of the system. In Eqs. (2) and (3), \( \langle \cdots \rangle \) indicates the usual canonical ensemble thermal average for a given configuration, and the sum runs over all nearest-neighbors of the spin \( \vec{S}_i \). To perform the thermal averaging on the right hand side of Eqs. (2) and (3), we follow the general approach described in Ref. [45]. In the spirit of the effective field theory, multi-spin-correlation functions are approximated by products of single spin averages. We then take advantage of the integral representation of the Dirac delta distribution, in order to write Eqs. (2) and (3) in the following form:

\[ p_{n,z} = \int d\omega f_\alpha(\omega, \Omega) \frac{1}{2\pi} \int dt \exp(i\omega t) \prod_j \langle \exp(\langle it J_{i,j} S_{j,z} \rangle) \rangle. \tag{6} \]

Now, we introduce the probability distribution of the spin variables (for details see Ref. [45]):

\[ P_r(S_{n,z}) = \frac{1}{2} \left[ (1 - 2p_{n,z}) \delta \left( S_{n,z} - \frac{1}{2} \right) + (1 + 2p_{n,z}) \delta \left( S_{n,z} + \frac{1}{2} \right) \right]. \tag{7} \]
Using the six previous equations, we get the following equations for the layer polarizations:

For \( n = 1 \)

\[
p_{1,\alpha} = 2^{-N-N_0} \sum_{\mu_1=0}^{N} \sum_{\mu_2=0}^{N_0} \left\{ C_{\mu_1}^N C_{\mu_2}^{N_0} (1 - 2p_{1,z})^{\mu_1} (1 + 2p_{1,z})^{N-\mu_1} \right\}
\]

\[
(1 - 2p_{2,z})^{\mu_2} (1 + 2p_{2,z})^{N_0-\mu_2} f_\alpha (y_1 + 2\mu E, \Omega_s) \},
\]

with

\[
y_1 = \frac{1}{2} J_s [N + N_0 - 2(\mu_1 + \mu_2)].
\]

For \( n = 2 \)

\[
p_{2,\alpha} = 2^{-N-2N_0} \sum_{\mu_1=0}^{N} \sum_{\mu_2=0}^{N_0} \sum_{\mu_3=0}^{N_0} \left\{ C_{\mu_1}^N C_{\mu_2}^{N_0} C_{\mu_3}^{N_0} (1 - 2p_{2,z})^{\mu_1} (1 + 2p_{2,z})^{N-\mu_1} (1 - 2p_{1,z})^{\mu_2} \right\}
\]

\[
(1 + 2p_{1,z})^{N_0-\mu_2} (1 - 2p_{3,z})^{\mu_3} (1 + 2p_{3,z})^{N_0-\mu_3} f_\alpha (y_2 + 2\mu E, \Omega_s) \},
\]

with

\[
y_2 = \frac{1}{2} [J_s (N - 2\mu_1) + J_s (N_0 - 2\mu_2) + J_e (N_0 - 2\mu_3)].
\]

For \( n = 3 \)

\[
p_{3,\alpha} = 2^{-N-2N_0} \sum_{\mu_1=0}^{N} \sum_{\mu_2=0}^{N_0} \sum_{\mu_3=0}^{N_0} \left\{ C_{\mu_1}^N C_{\mu_2}^{N_0} C_{\mu_3}^{N_0} (1 - 2p_{3,z})^{\mu_1} (1 + 2p_{3,z})^{N-\mu_1} (1 - 2p_{2,z})^{\mu_2} \right\}
\]

\[
(1 + 2p_{2,z})^{N_0-\mu_2} (1 - 2p_{4,z})^{\mu_3} (1 + 2p_{4,z})^{N_0-\mu_3} f_\alpha (y_3 + 2\mu E, \Omega) \},
\]

with

\[
y_3 = \frac{1}{2} [J_a (N - 2\mu_1) + J_e (N_0 - 2\mu_2) + J_a (N_0 - 2\mu_3)].
\]

For \( 3 < n < L - 2 \)

\[
p_{n,\alpha} = 2^{-N-2N_0} \sum_{\mu_1=0}^{N} \sum_{\mu_2=0}^{N_0} \sum_{\mu_3=0}^{N_0} \left\{ C_{\mu_1}^N C_{\mu_2}^{N_0} C_{\mu_3}^{N_0} (1 - 2p_{n,z})^{\mu_1} (1 + 2p_{n,z})^{N-\mu_1} (1 - 2p_{n-1,z})^{\mu_2} \right\}
\]

\[
(1 + 2p_{n-1,z})^{N_0-\mu_2} (1 - 2p_{n+1,z})^{\mu_3} (1 + 2p_{n+1,z})^{N_0-\mu_3} f_\alpha (y_n + 2\mu E, \Omega) \},
\]

with

\[
y_n = \frac{1}{2} J_a [N + 2N_0 - 2(\mu_1 + \mu_2 + \mu_3)].
\]

For \( n = L - 2 \)

\[
p_{L-2,\alpha} = 2^{-N-2N_0} \sum_{\mu_1=0}^{N} \sum_{\mu_2=0}^{N_0} \sum_{\mu_3=0}^{N_0} \left\{ C_{\mu_1}^N C_{\mu_2}^{N_0} C_{\mu_3}^{N_0} (1 - 2p_{L-2,z})^{\mu_1} (1 + 2p_{L-2,z})^{N-\mu_1} (1 - 2p_{L-3,z})^{\mu_2} \right\}
\]

\[
(1 + 2p_{L-3,z})^{N_0-\mu_2} (1 - 2p_{L-1,z})^{\mu_3} (1 + 2p_{L-1,z})^{N_0-\mu_3} f_\alpha (y_{L-2} + 2\mu E, \Omega_s) \},
\]

(16)
with
\[
y_{L-2} = \frac{1}{2} [J_a (N - 2\mu_1) + J_a (N_0 - 2\mu_2) + J_e (N_0 - 2\mu_3)].
\] (17)

For \( n = L - 1 \)
\[
p_{L-1,\alpha} = 2^{-N-2N_0} \sum_{\mu_1=0}^{N} \sum_{\mu_2=0}^{N_0} \sum_{\mu_3=0}^{N_0} \{ C_{\mu_1}^N C_{\mu_2}^{N_0} C_{\mu_3}^{N_0} (1 - 2p_{L-1,z})^{\mu_1} (1 + 2p_{L-1,z})^{N-\mu_1} (1 - 2p_{L-2,z})^{\mu_2} \\
(1 + 2p_{L-2,z})^{N_0-\mu_2} (1 - 2p_{L,z})^{\mu_3} (1 + 2p_{L,z})^{N_0-\mu_3} f_\alpha (y_{L-1} + 2\mu E, \Omega) \},
\] (18)

with
\[
y_{L-1} = \frac{1}{2} [J_s (N - 2\mu_1) + J_e (N_0 - 2\mu_2) + J_s (N_0 - 2\mu_3)].
\] (19)

For \( n = L \)
\[
p_{L,\alpha} = 2^{-N-N_0} \sum_{\mu_1=0}^{N} \sum_{\mu_2=0}^{N_0} \sum_{\mu_3=0}^{N_0} \{ C_{\mu_1}^N C_{\mu_2}^{N_0} (1 - 2p_{L,z})^{\mu_1} (1 + 2p_{L,z})^{N-\mu_1} \\
(1 - 2p_{L-1,z})^{\mu_2} (1 + 2p_{L-1,z})^{N_0-\mu_2} f_\alpha (y_L + 2\mu E, \Omega) \},
\] (20)

with
\[
y_L = \frac{1}{2} J_s [N + N_0 - 2(\mu_1 + \mu_2)].
\] (21)

In these equations, \( C_k^l \) are the binomial coefficients: \( C_k^l = \frac{l!}{k!(l-k)!} \), \( N \) and \( N_0 \) denote, respectively, the coordination numbers in the parallel- and inter-planes. For the case of a simple cubic lattice which is considered here, one has \( N = 4 \) and \( N_0 = 1 \).

We have thus obtained the self-consistent equations (8)–(21) for the layer longitudinal polarizations \( p_{n,z} \) that can be directly solved by numerical iterations. The average longitudinal and transverse polarizations of the system are defined by
\[
\bar{P}_{av,z} = \frac{1}{L} \sum_{n=1}^{L} p_{n,z},
\] (22)
\[
\bar{P}_{av,x} = \frac{1}{L} \sum_{n=1}^{L} p_{n,x}.
\] (23)

The dielectric properties are important in practice and, in particular, the longitudinal dielectric susceptibility is an interesting physical quantity which describes how the longitudinal polarization responds to an external electric field \( E \). The phase transition is usually predicted by the abnormal behavior of the longitudinal dielectric susceptibility at the critical temperature.
The longitudinal dielectric susceptibility of the \( n \)th layer is given by

\[
\chi_{n,z} = \frac{\partial p_{n,z}}{\partial E} \bigg|_{E \to 0}.
\]  

(24)

The average longitudinal dielectric susceptibility of the MSF thin film \( \chi_z \) is determined from

\[
(1 + \chi_z)^{-1} = \frac{1}{L} \sum_{n=1}^{L} (1 + \chi_{n,z})^{-1}.
\]  

(25)

The pyroelectric coefficient of the \( n \)th layer is defined by

\[
Pyro_n = -\frac{\partial p_{n,z}}{\partial T}.
\]  

(26)

The average pyroelectric coefficient of the MSF is given by

\[
Pyro_{avg} = -\frac{1}{L} \sum_{n=1}^{L} Pyro_n.
\]  

(27)

III. RESULTS AND DISCUSSION

In this section, we are interested in the investigation of the layer and the average longitudinal and transverse polarizations, the dielectric susceptibility, the pyroelectric coefficient, and the hysteresis loops of the spin-\( \frac{1}{2} \) MSF thin film. These quantities depend on the temperature, the exchange interactions (\( J_s, J_a, \) and \( J_e \)), and the transverse fields (\( \Omega \) and \( \Omega_s \)). In our calculations and for simplicity, we take the reduced parameter \( J_a \) as the unit of the energy, and we consider the MSF thin film with two top and two bottom surface layers and \((L - 4)\) bulk layers (Fig. 1). Due to the symmetry of the system, we are interested in the polarization of the three top layers.

The interest of surface magnetism systems comes from the unique physical phenomenon related to the reduced dimensionality at the surface. Indeed, the system can be ordered in the surfaces before being so in the bulk, when the ratio of the surface exchange interaction to the bulk one (\( J_s/J_a \)) is larger than a critical value. Surface magnetic order, higher than the bulk one, has been obtained experimentally [46, 47]. While, the weak pseudo-spin interactions in surface transition layers lead to the reduction of the system transition temperature [48].

Figs. 2a–2d, show the temperature dependence of the layer and the average longitudinal and transverse polarizations, the dielectric susceptibility, and the pyroelectric coefficient for two values of the surface exchange interaction (\( J_s/J_a = 0.3 \) solid lines and \( J_s/J_a = 3.0 \) dashed lines)). The selected parameters in these figures are: \( \Omega/J_a = \Omega_s/J_a = 1.0, J_e/J_a = 1.0 \), and \( L = 10 \). In Fig. 2a, we depict the average longitudinal polarization (\( P_{av,z} \)) and the layer longitudinal polarizations (\( p_{1,z}, p_{2,z}, \) and \( p_{3,z} \)) of the MSF system. For
the weak surface exchange interaction, the polarization of the first layer $p_{1,z}$ is less than that of the second layer $p_{2,z}$, which is also lower than that of the third layer $p_{3,z}$. However, for the strong surface exchange interaction, the situation is reversed. Indeed, the polarization of the first and the second layers, $p_{1,z}$ and $p_{2,z}$, are greater than that of the third layer $p_{3,z}$. On the other hand, the average longitudinal polarization and the critical temperature of the system increase with the value of $J_s/J_a$. This is an interesting result; the largest layer polarization appears on the second surface layer, because its ligand-pseudospin number is greater than the first surface layer and its exchange interaction is stronger than the other inner layers. By comparing the polarizations (solid lines) and (dashed lines), we can see that when $J_s/J_a$ increases from 0.3 to 3.0, the spontaneous polarization of each layer becomes larger, and the polarizations of the two surface layers $p_{1,z}$ and $p_{2,z}$ increase more remarkably. Such a result is easy to understand, since the change of the surface exchange interaction $J_s$ should have a larger effect on the polarization of the surface layers. This result is in agreement with that obtained by Yang et al. [44]. In Fig. 2b, we plot the variation of the average and the layer transverse polarizations with the temperature for the two different values of $J_s/J_a$. We remark that the curves show a cusp near the transition temperature which increases with the value of $J_s/J_a$. For the weak surface exchange interaction, the first layer polarization $p_{1,x}$ is greater than that of the second one $p_{2,x}$, which is also greater than that of the third one $p_{3,x}$. Moreover, for the strong surface exchange interaction, the third layer transverse polarization $p_{3,x}$ is larger than those of $p_{1,x}$ and $p_{2,x}$. The average transverse polarization decreases with an increase of $J_s/J_a$.

The effects of the ratio $J_s/J_a$ on the pyroelectric coefficient and on the dielectric susceptibility of the system are given in Figs. 2c and 2d, for the same values of the parameters used in Fig. 2a. We remark that the curves present a peak at the critical temperature which increases with an increase of $J_s/J_a$. In Fig. 2e, we depict the surface exchange interaction effect on the ferroelectric hysteresis loops of the system which are obtained by changing cyclically the values of the external applied electric field. At the fixed temperature $T/J_a = 0.2$, the polarization curves are symmetric for both positive and negative values of the electric field. The hysteresis loops of the two surface layers become larger than those of the third layer with an increase of $J_s/J_a$.

In Figs. 3a and 3b, the layer and the average longitudinal and transverse polarizations are depicted as functions of the temperature for two values of the inter-layer exchange interaction ($J_e/J_a = 0.3$ (solid lines) and $J_e/J_a = 3.0$ (dashed lines)) and for $\Omega/J_a = \Omega_s/J_a = 1.0$, $J_s/J_a = 1.0$, and $L = 10$. We remark that when the inter-layer exchange interaction increases, the average and the layer longitudinal polarizations increase (Fig. 3a). However, the average and the layer transverse polarizations decrease (Fig. 3b), which leads to an increase of the critical temperature with $J_e/J_a$. In Figs. 3c and 3d, we display the temperature dependencies of the layer and the average susceptibilities and pyroelectric coefficient for the same values of the inter-layer exchange interaction $J_e/J_a = 0.3$ and 3.0. For the weak inter-layer exchange interaction, a round peak appears before the critical temperature. However, for the strong inter-layer exchange interaction, the last round peak disappears. Our results are in agreement with those obtained by Chen et al. [49], where they have shown that the effect of the intra-layer interactions is more than that of the inter-layer
(a) The three layers and the average longitudinal polarizations as functions of the temperature for different values of the surface exchange interaction ($J_s/J_a = 0.3$ (solid lines), $J_s/J_a = 3.0$ (dashed lines)). The parameters adopted for the calculations are: $\Omega/J_a = \Omega_s/J_a = 1.0$, $J_e/J_a = 1.0$, and $L = 10$.

(b) The three layers and the average transverse polarizations as functions of the temperature for different values of the surface exchange interaction ($J_s/J_a = 0.3$ (solid lines), $J_s/J_a = 3.0$ (dashed lines)). The parameters adopted for the calculations are: $\Omega/J_a = \Omega_s/J_a = 1.0$, $J_e/J_a = 1.0$, and $L = 10$.

(c) The three layers and the average dielectric susceptibilities as functions of the temperature for different values of the surface exchange interaction ($J_s/J_a = 0.3$ (solid lines), $J_s/J_a = 3.0$ (dashed lines)). The parameters adopted for the calculations are: $\Omega/J_a = \Omega_s/J_a = 1.0$, $J_e/J_a = 1.0$, and $L = 10$.

(d) The three layers and the average pyroelectric coefficients as functions of the temperature for different values of the surface exchange interaction ($J_s/J_a = 0.3$ (solid lines), $J_s/J_a = 3.0$ (dashed lines)). The parameters adopted for the calculations are: $\Omega/J_a = \Omega_s/J_a = 1.0$, $J_e/J_a = 1.0$, and $L = 10$.

(e) The hysteresis loops for different values of the surface exchange interaction ($J_s/J_a = 0.3$ (solid lines), $J_s/J_a = 3.0$ (dashed lines)). The parameters adopted for the calculations are: $\Omega/J_a = \Omega_s/J_a = 1.0$, $T/J_a = 0.2$, $J_e/J_a = 1.0$, and $L = 10$.

FIG. 2:
In Fig. 3e, we plot the inter-layer exchange interaction effect on the ferroelectric hysteresis loops of the system. For the fixed temperature $T/J_a = 0.2$, the layers and the average longitudinal polarizations curves are symmetric for both positive and negative values of the electric field and the loops become larger with an increase of the inter-layer exchange interaction $J_e/J_a$.

Figs. 4a–4d show the temperature dependence of the layer and the average longitudinal and transverse polarizations, the dielectric susceptibility, and the pyroelectric coefficient for two values of the surface transverse field ($\Omega_s/J_a = 1.5$ (solid lines) and $\Omega_s/J_a = 3.0$ (dashed lines)). The selected parameters in these figures are: $J_s/J_a = J_e/J_a = 1.0$, $\Omega/J_a = 1.0$, and $L = 10$. Figs. 4a and 4b present the effect of the surface transverse field $\Omega_s/J_a$ on the layer and the average longitudinal and transverse polarizations. We note that when the surface transverse field increases, the average and the layer longitudinal polarizations decrease (Fig. 4a). However, the average and the layer transverse polarizations increase (Fig. 4b), which leads to a decrease of the critical temperature with $\Omega_s/J_a$. It is known that the pseudo-spins system becomes more disordered as the transverse field increases [39, 50, 51].

In Figs. 4c and 4d, we show the temperature dependence of the pyroelectric coefficient and the susceptibility for the same values of the surface transverse field $\Omega_s/J_a = 1.5$ and $3.0$. We can see that one peak appears in the pyroelectric coefficient and in the dielectric susceptibility curves. However, when the values of $\Omega_s/J_a$ increase, the peak position moves to the low temperatures, which indicates that the phase transition temperature of the system decreases with an increase of the surface transverse field.

Finally, the dependence of the hysteresis loops on the surface transverse field $\Omega_s/J_a$ is shown in Fig. 4e, for the same parameters adopted in Figs. 4. From this figure, we can see that the polarization curves are symmetric for both the positive and negative longitudinal electric field. The loops of the 1st and 2nd surface layers become narrower with an increase of the surface transverse field below the critical temperature. However, the loop of the 3rd layer becomes large.

**IV. CONCLUSION**

We have studied the dielectric and pyroelectric properties and the hysteresis loops of an MSF thin film in the presence of an applied external electric field using the transverse Ising Model with the effective field theory based on the probability distribution technique. We have discussed in detail the influence of the surface transverse field and the exchange interactions on the hysteresis loops, susceptibilities, and pyroelectric coefficient. We have found that the critical temperature increases with the exchange interactions and the surface transverse field make the hysteresis loops narrow and the system becomes disordered. It should be mentioned that Wesselinowa and Dimitrov [36] have studied the influence of substrates on the dielectric properties of ferroelectric thin films. The obtained results indicate that the influence of the substrates is similar to that in STL systems. The substrates
(a) The three layers and the average longitudinal polarizations as functions of the temperature for different values of the inter-layer exchange interaction ($J_e/J_a = 0.3$ (solid lines), $J_e/J_a = 3.0$ (dashed lines)). The parameters adopted for the calculations are: $\Omega/J_a = \Omega_s/J_a = 1.0$, $J_s/J_a = 1.0$, and $L = 10$.

(b) The three layers and the average transverse polarizations as functions of the temperature for different values of the inter-layer exchange interaction ($J_e/J_a = 0.3$ (solid lines), $J_e/J_a = 3.0$ (dashed lines)). The parameters adopted for the calculations are: $\Omega/J_a = \Omega_s/J_a = 1.0$, $J_s/J_a = 1.0$, and $L = 10$.

(c) The three layers and the average dielectric susceptibilities as functions of the temperature for different values of the inter-layer exchange interaction ($J_e/J_a = 0.3$ (solid lines), $J_e/J_a = 3.0$ (dashed lines)). The parameters adopted for the calculations are: $\Omega/J_a = \Omega_s/J_a = 1.0$, $J_s/J_a = 1.0$, and $L = 10$.

(d) The three layers and the average pyroelectric coefficients as functions of the temperature for different values of the inter-layer exchange interaction ($J_e/J_a = 0.3$ (solid lines), $J_e/J_a = 3.0$ (dashed lines)). The parameters adopted for the calculations are: $\Omega/J_a = \Omega_s/J_a = 1.0$, $J_s/J_a = 1.0$, and $L = 10$.

(e) The hysteresis loops for different values of the inter-layer exchange interaction ($J_e/J_a = 0.3$ (solid lines), $J_e/J_a = 3.0$ (dashed lines)). The parameters adopted for the calculations are: $\Omega/J_a = \Omega_s/J_a = 1.0$, $J_s/J_a = 1.0$, $T/J_a = 0.2$, $J_s/J_a = 1.0$, and $L = 10$.

FIG. 3:
(a) The three layers and the average longitudinal polarizations as functions of the temperature for different values of the surface transverse field \( \Omega_s/J_a = 1.5 \) (solid lines), \( \Omega_s/J_a = 3.0 \) (dashed lines). The parameters adopted for the calculations are: \( \Omega/J_a = 1.0, \ j_s/J_a = j_e/J_a = 1.0, \) and \( L = 10. \)

(b) The three layers and the average transverse polarizations as functions of the temperature for different values of the surface transverse field \( \Omega_s/J_a = 1.5 \) (solid lines), \( \Omega_s/J_a = 3.0 \) (dashed lines). The parameters adopted for the calculations are: \( \Omega/J_a = 1.0, \ j_s/J_a = j_e/J_a = 1.0, \) and \( L = 10. \)

(c) The three layers and the average dielectric susceptibilities as functions of the temperature for different values of the surface transverse field \( \Omega_s/J_a = 1.5 \) (solid lines), \( \Omega_s/J_a = 3.0 \) (dashed lines). The parameters adopted for the calculations are: \( \Omega/J_a = 1.0, \ j_s/J_a = j_e/J_a = 1.0, \) and \( L = 10. \)

(d) The three layers and the average pyroelectric coefficients as functions of the temperature for different values of the surface transverse field \( \Omega_s/J_a = 1.5 \) (solid lines), \( \Omega_s/J_a = 3.0 \) (dashed lines). The parameters adopted for the calculations are: \( \Omega/J_a = 1.0, \ j_s/J_a = j_e/J_a = 1.0, \) and \( L = 10. \)

(e) The hysteresis loops for different values of the surface transverse field \( \Omega_s/J_a = 1.5 \) (solid lines), \( \Omega_s/J_a = 3.0 \) (dashed lines). The parameters adopted for the calculations are: \( \Omega/J_a = 1.0, \ T/J_a = 0.2, \ j_s/J_a = j_e/J_a = 1.0, \) and \( L = 10. \)

FIG. 4:
can also make the susceptibility peak position and Curie temperature to occur at either lower or higher temperatures and the susceptibility peak decreases.

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