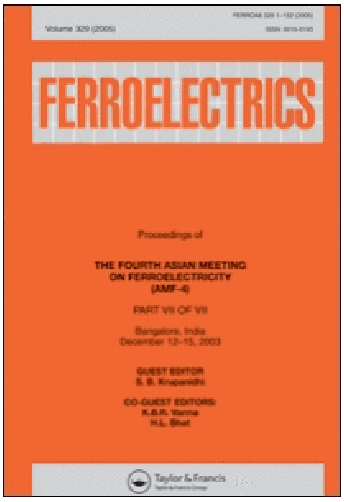

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Analysis of Leakage Current Mechanisms in $BiFeO_3$ Thin Films

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Phonon-assisted tunneling (PhAT) model is applied for explication of temperature-dependent I-V characteristics measured by other investigators for $BiFeO_3$ films. Our proposed model describes well not only current dependence on temperature measured in a wide temperature range but also temperature-dependent I-V data using the same set of parameters characterizing material under investigation. The values of active phonons energy and field strength for tunneling are estimated from the fit of current dependence on temperature and I-V-T data with the two different phonon-assisted tunnelling theories.

Keywords Ferroelectrics; phonon-assisted tunneling; *I-V* characteristics

PACS: 77.55.fp; 73.50.Fq; 73.43.Jn; 63.20.kd

1. Introduction

Recently, lead-free ferroelectric $BiFeO_3$ (BFO) has been the subject of intense studies due to its superior ferroelectric and piezoelectric properties [1, 2]. Despite the fact that electronic conduction properties of those structures have been studied in some works [3–7], the current nonlinear behavior and its temperature variation are not fully understood yet. For instance, Qi et al. [3] asserted that the main conduction mechanism for pure and Ni^{2+} doped BFO was a space charge limited current (SCLC), while Pabst et al. [5] affirmed that the Poole-Frenkel (PF) emission was the predominant mechanism in the high quality epitaxial BFO film formed by pulsed laser deposition (PLD) symmetric $SrRuO_3$/BFO/$SrRuO_3$ capacitors and no clear dominant current mechanism can be attributed for the asymmetric Pt/BFO/$SrRuO_3$ structures. Yang et al. [6] have reported the temperature-dependent leakage mechanisms in Pt/BFO/$SrRuO_3$ thin film capacitors with a 200 nm thick BFO film formed by PLD. It was found that the leakage mechanisms were a strong function of temperature and voltage polarity. At temperatures between 80 and 150 K, in the authors [6] opinion, SCLC was the dominant leakage mechanism for both negative and positive biases, while at temperatures between 200 and 350 K the authors of Ref. [6] envisaged the Poole-Frenkel emission and Fowler-Nordheim tunneling as dominant leakage mechanisms for negative and positive biases, respectively. In a very recent publication, Zhong and Ishiwara [8] affirmed that "The leakage current density in a BFO film was found to be subject to space charge limited conduction instead of Poole-Frenkel emission." Thus, judgments of the different investigators on the leakage mechanism in the BFO films are very controversial.

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In the present work, we argue that the main peculiarities of current dependence on temperature and electric field observed in the BFO films could be explained on the basis of contemporary phonon-assisted tunneling (PhAT) theory, which was previously used for the explanation of the temperature-dependent current-voltage (I-V*)* characteristics in some dielectrics [9, 10].

2. Model and Comparison with Experimental Data

The PhAT model is based on the assumption that the temperature-field-dependent conductivity is caused by a thermally activated free charge carriers generation process, which is an electric field initiated phonon-assisted tunneling of electrons from the electronic states near the interface to the conduction band. Assuming that the released carriers achieve the opposite electrode the current density j_t is expressed as

$$j_t = eN_sW_t, \tag{1}$$

where *e* is the electronic charge unit, N_s is the occupied state density at the interface and W_t is the rate of phonon-assisted tunneling, which is the function of temperature T and field strength E, and is given by [9, 11]:

$$W_1 = \frac{eE}{(8m^*\varepsilon_T)^{1/2}}[(1+\gamma^2)^{1/2}-\gamma]^{1/2}[1+\gamma^2]^{-1/4}\exp\left\{-\frac{4}{3}\frac{(2m^*)^{1/2}}{eE\hbar}\varepsilon_T^{3/2} \times [(1+\gamma^2)^{1/2}-\gamma]^2\left[(1+\gamma^2)^{1/2}+\frac{1}{2}\gamma\right]\right\}, \gamma = \frac{(2m^*)^{1/2}\Gamma^2}{8e\hbar E\varepsilon_T^{1/2}}. \tag{2}$$

Here $\Gamma^2 = \Gamma_0^2(2n+1) = 8\alpha(\hbar\omega)^2(2n+1)$ is the width of the center absorption band, where $\hbar\omega$ is the phonon energy, $n = [\exp(\hbar\omega/k_BT)-1]^{-1}$ is the temperature distribution of phonons density, ε_T is the trap depth and *a* is the electron-phonon coupling constant.

At first, let us compare the temperature-dependent I-V characteristics measured in the temperature range from 200 to 350 K by Yang et al. [6] on Pt/BFO/$SrRuO_3$ thin film capacitors with a 200 nm thick BFO film with the theory.

The tunneling rate W_1(E,T), for the comparison with the experimental data was computed using a value for the electron effective mass of $0.2m_e$, for the ε_T the activation energy estimated in Ref. [6] at the low field of 0.6 eV and the phonon energy of 50 meV was taken. The electron-phonon coupling constant *a* was chosen so as to get the best fit of the experimental data to the calculated dependences on the assumption that field strength is proportional to square root of applied voltage, i.e. the tunneling occurs in the Schottky barrier.

The fit of I-E curves in the temperature range from 200 to 350 K measured for negative biases extracted from Fig. 1 [6] to the theoretical W(E,T) dependences are exposed in Fig. 1. As can be seen, the theoretical W_1(E,T) vs E dependences describe well the experimental data obtained at higher field strength. From the fit of experimental data with the theory in Fig. 1 the centers density in the interface estimated using the equation (1) is found to be equal to 1.3×10^{12} cm^{-2}, which is very reliable value.

In the Fig. 2 temperature dependence of the leakage currents in the high quality epitaxial BFO film formed by pulsed laser deposition symmetric $SrRuO_3$/BFO/$SrRuO_3$ capacitors measured by Pabst et al. [5]. The authors of Ref. [5] affirmed that the PF emission was the predominant mechanism in these capacitors. As can be seen in Fig. 2 the PhAT model describes the lnJ against 1/T curves well.

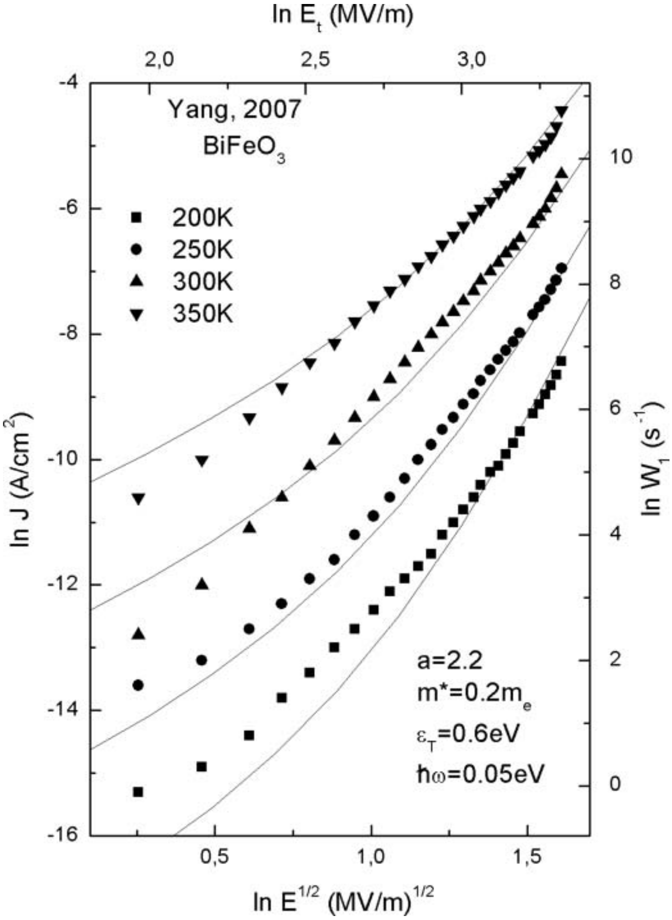

Figure 1. The experimental J-V characteristics of Pt/BiFO$_3$/SrRuO$_3$ ferroelectric thin films from Fig. 1 in [6] (symbols) fitted to the theoretical W$_1$-E$_t$ dependences (solid lines).

As noted above, the presence of a Poole–Frenkel conduction mechanism has been reported for $(1-x)$BiFeO$_3$–xPbTiO$_3$ ($0.5 < x < 0.3$) thin films with $x = 0.4$ and $x = 0.5$ Khan et al. [7]. The fit of that data to the PhAT model is displayed in Fig. 3, from which one can see that the theoretical curves describe the experimental data also well for all field strength.

We want to note that the similar temperature-dependent tunneling rate on field strength also gives phonon-assisted tunneling theory developed by Makram-Ebeid and Lannoo [12]. To confirm this assertion in Figure 4, the comparison of the same experimental data as is shown in Fig. 1 with W$_2$(F,T) dependences computed using the theory from [12]. The phonon-assisted tunneling rate of the electrons from the impurity center of depth ε_T derived by the use of the Condon approximation is given by the following expression (Eq. (18) in Ref. [12]):

$$W_2(E,T) = \sum_{p=-p_o}^{+p_o} RW_e(\varepsilon_T + p\hbar\omega), \tag{3}$$

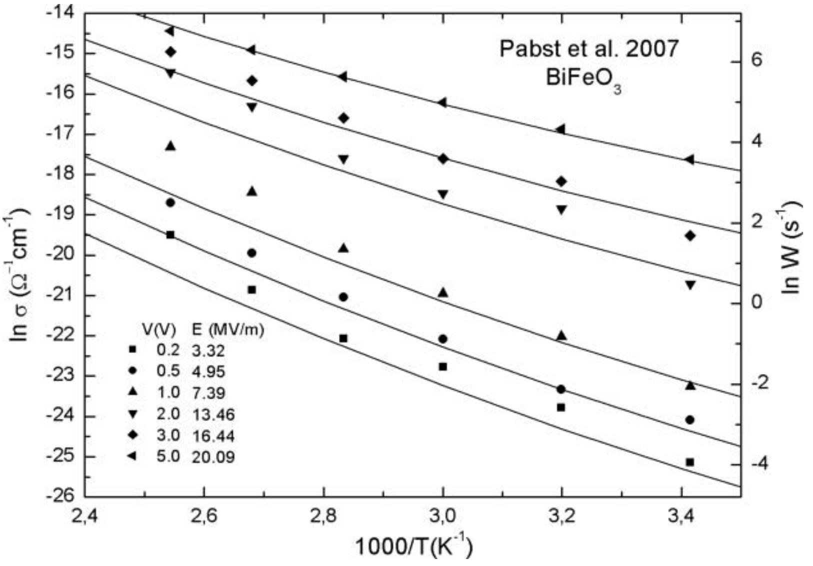

Figure 2. The experimental dependences ln (σ) against 1/T of $SrRuO_3$/(Pt/BFO/$SrRuO_3$ thin films from Fig. 3 in [5] symbols) fitted to the theoretical lnW_1 against 1/T dependences (solid lines), computed for parameters: $a = 1.8$, $m^* = 0.2$ m_e, $\varepsilon_T = 0.60$ eV, $\hbar\omega = 50$ meV.

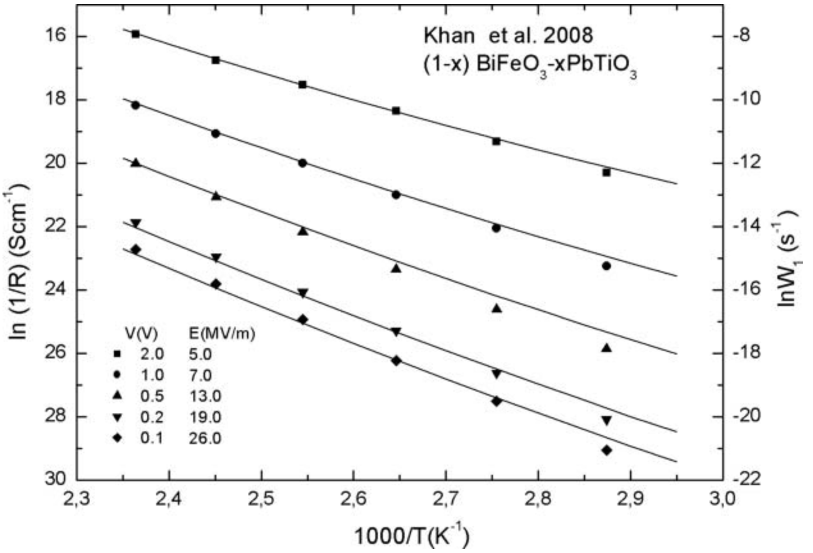

Figure 3. The experimental dependences ln (σ) against 1/T of $(1-x)BiFeO_3$–$xPbTiO_3$ ($0.5 < x < 0.3$) thin films from Fig. 3 in [7] (symbols) fitted to the theoretical ln W_1 against 1/T dependences (solid lines), computed for parameters: $a = 3.2$, $m^* = 0.2$ m_e, $\varepsilon_T = 1.05$ eV, $\hbar\omega = 50$ meV.

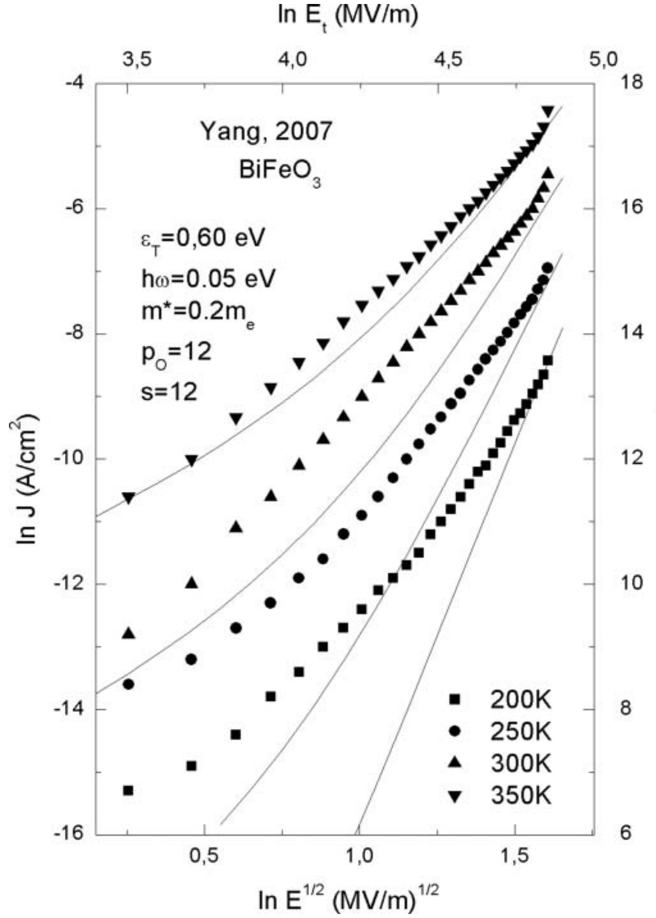

Figure 4. The same experimental J-V characteristics of Pt/BiFO$_3$/SrRuO$_3$ ferroelectric thin films as in figure 1 fitted to the theoretical W$_2$-E$_\mathrm{t}$ dependences (solid lines).

where

$$R = \exp\left(\frac{p\hbar\omega}{2k_BT} - S\mathrm{cth}\frac{\hbar\omega}{2k_BT}\right) I_p\left(\frac{S}{\mathrm{sh}(\hbar\omega/2k_BT)}\right),$$

$$W_e(\varepsilon_T) = \left(\frac{2eE}{(2m*\varepsilon_T)^{1/2}} - \right)\exp\left[-4\frac{(2m*)^{1/2}}{3e\hbar E}\right]{\varepsilon_T}^{3/2} \tag{4}$$

Here $p_o = \varepsilon_T/\hbar\omega$, $\hbar\omega$ is the phonon energy, I$_\mathrm{p}$ is the modified Bessel function and S is the Huang-Rhys coupling constant.

It is seen in Fig. 4 that this theory describes the peculiarities of the I-V characteristics variation with temperature at high fields well enough. The density of the electronic states estimated from this match is found to be equal to 1.8×10^9 cm^{-2}. The field strengths for tunneling in this case are somewhat higher then in Fig. 1.

Finally, in Fig. 5 we expose the fit of the I-V data from the very recent publication of Zhong and Ishiwara [8] for BFO film with both theories. The authors of [8] have affirmed

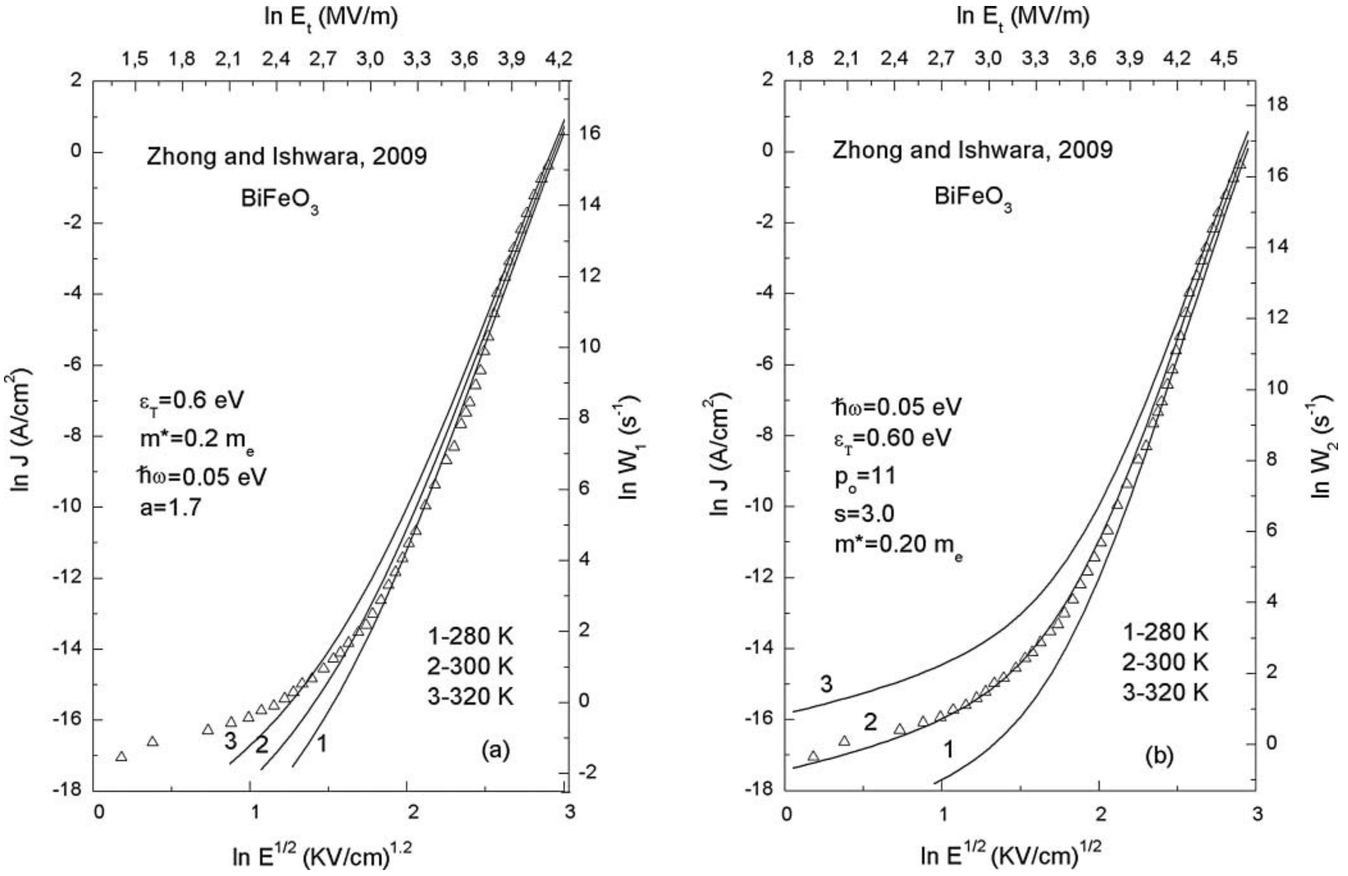

Figure 5. The experimental J-V characteristics of BiFO$_3$ ferroelectric thin films from Fig. 1 in [8] (symbols) fitted to the theoretical: a) W$_1$-E$_t$ dependences computed for three temperatures (solid lines); b) W$_2$-E dependences (solid lines).

that the leakage current density in a pure BFO film was found to be subject to the space-charge-limited conduction, instead of the PF emission. As seen from Fig. 5, both theories describe the experimental data at higher voltage very well. The excellent match theory with experiment in all range of bias voltages gives the Makram-Ebeid equation. The centers density in the interface estimated from the fitting using Eq. (2) and Eq. (3) is found to be equal to 1.2×10^{12} cm^{-2} and 2.6×10^{11} cm^{-2}, respectively.

Conclusions

One can expect a match of the experimental I-V-T data with the theoretical W(E,T) dependences in the wide temperature range and field strengths to be not accidental, consequently, the tunneling process stimulated by the phonons must be taken into account in describing the experimental data in the metal-$BiFeO_3$ structures. The classical PF emission and SCLC models, which are often invoked by the investigators to explain the dependence of the current on temperature, are workable only in a very limited range of temperatures and field strength. From the fit of experimental data with the tunneling theories the values of active phonons energy, density of electronic states in the interface and the field strength at which the tunneling occurs are estimated. Therefore, the phonon-assisted tunneling must be taken into account in describing the electrical conductance in films of inorganic materials.

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