Comment on “Influence of image forces on the electron transport in ferroelectric tunnel junctions”

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Udalov et al. in the recent papers [Phys. Rev. B 95, 134106 (2017); Phys. Rev. B 96, 125425 (2017)] report the strong influence of image forces on the conductance of ferroelectric tunnel junctions. In particular, the authors state that there is enhancement of the electroresistance effect due to polarization hysteresis in symmetric tunnel junctions at nonzero bias. This conjecture seems to be a breakthrough — the common knowledge is that the effect takes place only in non-symmetric junctions. We show that the enhancement of image forces on the conductance of ferroelectric tunnel junctions is highly overestimated due to neglecting the difference between characteristic ferroelectric relaxation and electron tunneling times. We argue that notable enhancement of the electroresistance effect due to polarization hysteresis in symmetric tunnel junctions at nonzero bias might be observed only at anomalously slow electron tunneling through the barrier. The same applies to magnetic tunnel junctions with a ferroelectric barrier also considered by Udalov et al: there is no significant increase in the magnetoelectric effect due to image forces for typical electron tunneling times.

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In a recent papers \cite{1,2} Udalov and Beloborodov (UB) address ferroelectric tunnel junctions where there is ferroelectric layer between metallic electrodes. They investigate non-magnetic \cite{1} and magnetic \cite{2} metallic electrodes. In \cite{1} UB focused on the special case of symmetric junctions with non-magnetic equivalent electrodes. Contrary to common knowledge UB find the strong enhancement of electroresistance effect taking into account the image force contribution \cite{3} to the tunnel probability. In magnetic tunnel junctions UB show that image forces significantly increase the magnetoelectric effect \cite{2}. The predicted effects regarding the symmetric junctions seem very promising not only from academic point of view but also for microelectronic applications. So correct understanding is very important.

Long ago it has been understood that the potential barrier at the metal-vacuum or metal-insulator interface can not change abruptly as in Gamov model of barrier at the metal-vacuum or metal-insulator interface. It is very important.

In magnetic tunnel junctions UB show that image forces significantly increase the magnetoelectric effect \cite{2}. The predicted effects regarding the symmetric junctions seem very promising not only from academic point of view but also for microelectronic applications. So correct understanding is very important.

Below we show that the last statement should be treated very accurately and effects reported in \cite{1,2} should be revisited and in the presented form they can hardly be observed.

The derivation of Eq. \textsuperscript{(3)} implies “adiabatic” approximation when all the contributions to polarization (related to $\epsilon$) are fast enough \cite{5-9} to follow electron moving through the tunnel barrier, see Fig. 1. In fact polarisation consists of several contributions with different

$$V_0 = \int_{-\infty}^{x} F(x)dx = -\frac{\epsilon^2}{4\epsilon x},$$

where dielectric occupies the right half-space.

If we have the standard tunnel barrier – two bulk parallel metallic contacts with dielectric media between them, then infinite series of images appears. The resulting sum for moderate distance $d$ between the metallic contacts is usually approximated by the simple analytical expression \cite{3}:

$$V \approx -\frac{0.795\epsilon^2 d}{4\epsilon x(d-x)}.$$
characteristic times \[ \tau_{\text{FE}} \]:

\[
P = P_{\text{el}} + P_{\text{ion}} + P_{\text{dipols}} + \ldots
\] (4)

Here the first contribution is polarization of the outer electron shells, the second one is related to ion shifts, the third is related to dipole moments of molecules etc... It is important that all the contributions except the first one are slow: their relaxation times are larger or of the order of inverse phonon frequencies. While \( P_{\text{el}} \) relaxation time is electronic (optical frequencies) and thus it is much shorter. Note, also, that the “slow” terms in (4) produce the leading contribution to \( P_{\text{FE}} \).

Relaxation dynamics of the ferroelectric order parameter can be estimated from

\[
\gamma P_{\text{FE}} = -\frac{\delta F_{\text{LD}}[P_{\text{FE}}]}{\delta P_{\text{FE}}} + \mathcal{E}_{\text{ext}}(t),
\] (5)

where \( \gamma_{\text{FE}} = 1/\tau_{\text{FE}} \) is the inverse relaxation time of ferroelectric polarization (order parameter), \( F_{\text{LD}}[P_{\text{FE}}] \) is the Landau-Devonshire free energy [10] that describes ferroelectric, and \( \mathcal{E}_{\text{ext}}(t) \) is time-dependent external electric field.

If we take \( \mathcal{E}_{\text{ext}}(t) \sim \mathcal{E}_{\text{ext}}(0)e^{-i\omega t} \) with \( \omega \) much larger than any characteristic frequency of a ferroelectric, then \( F_{\text{LD}} \)-term becomes irrelevant in Eq. (5), in the Fourier space \( P_{\omega} \sim \mathcal{E}_{\text{ext}}(0)/(-i\omega\gamma_{\text{FE}}) \), and, thus, \( \epsilon(\omega) \sim 1/(-i\omega\gamma_{\text{FE}}) \). This is very rough estimate that only illustrates the well known behaviour of ferroelectric dielectric constant with frequency: ferroelectricity does not respond on large enough frequencies. This is sketched in Fig. 2.

We can conclude about \( \epsilon \) in Eq. (3) – the key equation of Refs. [1, 2]:

\[
\epsilon = \epsilon_{\text{el}}.
\] (6)

This \( \epsilon \) is not known to be notably depending on the voltage bias \( V \) on the tunnel junction (unless the voltage produces the fields of the order of intrinsic atomic fields) and as the consequence, the effects predicted in Refs. [1, 2] are under question and require revision.

There are several ways of such a revision: one way corresponds to significant increase of the frequency response range of a ferroelectric and the other — significant slowdown of electron tunneling. Both opportunities are challenging for an experiment. However, as concerns the correction of theory, it is known that after tunneling, some time is required for the diffusion of extra electric charge over the electrode [12]. Maybe this effect can be the stone under the revised theory.

We should also note that there is a general fundamental question related to the described style of calculation, how ferroelectric polarization — macroscopic quantity can enter microscopic calculation like tunneling probability or magnetic exchange interaction. According to modern theory of polarization [13] at microscales \( \epsilon \) of a ferroelectric material has pronounced frequency and space dispersion \( \epsilon(\omega, k) \) [14] (this is not a problem for \( \epsilon_{\text{el}} \)) that is neglected in Refs. [1, 2].

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