Field-even current in ferroelectrics

A. S. Furman
A. F. Ioffe Physicotechnical Institute, Academy of Sciences of the USSR, Leningrad

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A mechanism is proposed for the field-even current which has recently been observed in a ferroelectric: a scattering of carriers by charged centers which create a local asymmetry of the polarization **P**. The current is expressed in terms of the coefficients of an expansion of the free energy in **P**. These coefficients determine the critical temperature behavior of the current. A negative transverse conductivity is predicted in the paraelectric phase.

1. The symmetry of crystals lacking an inversion center allows the existence of a field-even current $j_i = \sigma_{ikm} E_k E_m$. This current has been studied theoretically[1–3] and experimentally[4] in ferroelectric semiconductors. For the most part, the mechanisms which have been proposed for it involve an asymmetric scattering of charge carriers by impurities which have a given dipole or octupole moment.

A transverse current $j_z = \sigma_{zxx} E_x^2$ was recently observed in a ferroelectric near the point of the phase transition.[5] It was observed in a single-domain sample of SbSI, was directed along the polar (z) axis, changed sign upon a 180° polarization reversal, and disappeared in the transition to the paraelectric phase. It was established experimentally that the current j_z is associated with a scattering by charged centers which form an injected space charge. In the absence of this charge, a current j_z does not arise. These features are difficult to explain on the basis of the mechanisms which have been discussed previously.

In this letter we wish to propose a new mechanism, specific to ferroelectrics, for a field-even current: The asymmetry of the scattering required for the occurrence of this current is determined by a local asymmetric distribution of the polarization **P**, which is caused by the Coulomb field of a charged center. These ideas lead in a natural way to an explanation of the basic experimental results.[5] We will show that in a ferroelectric this mechanism is far more important than the mechanisms which have been discussed previously.

2. According to the theory of Ref. 1–3, a field-even current during the scattering by impurities with a potential V is expressed in terms of the antisymmetric part of the transition probability, $W_{\mathbf{kk}'}^{AS}$, which arises in second-order perturbation theory in the Born approximation:

$$W_{\mathbf{kk}'}^{AS} = \frac{1}{(4\pi)^4 \hbar} \operatorname{Im} \int V_{\mathbf{kk}'} V_{\mathbf{k}'\mathbf{k}''} V_{\mathbf{k}''\mathbf{k}} \delta(\epsilon_k - \epsilon_{k'}) \delta(\epsilon_k - \epsilon_{k''}) d\mathbf{k}''. \tag{1}$$

Here $\varepsilon_k = \hbar^2 k^2/2m$ is the energy of the scattered particle, with wave vector **k** and effective mass m. We wish to calculate $W_{\mathbf{kk}'}^{AS}$ for the model in which we are interested here: that of a charged center in a ferroelectric with a second-order phase transition. A model of a similar center was studied in Ref. 6 in an effort to determine its contribution to the specific heat, but its scattering of carriers was not considered.

The distribution of the potential $V = e\varphi$ is described by the equation of state of the ferroelectric and by the Poisson equation:

$$-\frac{\partial\varphi}{\partial z} = -\alpha P + \beta P^3 - \kappa\nabla^2 P, \tag{2}$$

$$\epsilon_\perp\left(\frac{\partial^2\varphi}{\partial x^2} + \frac{\partial^2\varphi}{\partial y^2}\right) + \frac{\partial^2\varphi}{\partial z^2} = 4\pi\frac{\partial P}{\partial z} + \frac{\varphi}{R^2} - 4\pi e\delta(\mathbf{r}). \tag{3}$$

Here α and β are known Landau-Ginzburg-Devonshire expansion coefficients, $k\nabla^2 P$ is a gradient term, and z is the polar axis. In (3) we have taken Debye screening into account: $R^2 = k_B T/e^2 n$, where n is the carrier density, and T is the crystal temperature. (As we will see, we have $W^{AS}_{\mathbf{kk}'} \to \propto$ as $R \to \infty$.

Linearizing (2), (3) for values of P close to the magnitude of the spontaneous polarization $P_0 = (\alpha/\beta)^{1/2}$, we find the symmetric ($V^S_{\mathbf{k}'\mathbf{k}''}$) and antisymmetric ($V^{AS}_{\mathbf{k}'\mathbf{k}''}$) parts of the matrix element $V_{\mathbf{k}'\mathbf{k}''}$:

$$V^S_{\mathbf{k}'\mathbf{k}''} = \frac{4\pi e^2(\kappa k^2 + 2\alpha)}{\Phi_{\mathbf{k}}}; \qquad V^{AS}_{\mathbf{k}'\mathbf{k}''} = -\frac{3\sqrt{\pi}\, i e^3 \beta P_0 \ln(\alpha) k_z}{2(\epsilon_\perp\kappa)^{1/2}\Phi_{\mathbf{k}}};$$

$$\Phi_{\mathbf{k}} = 4\pi k_z^2 + (\kappa k^2 + 2\alpha)(\epsilon_\perp k_\perp^2 + k_z^2 + R^{-2}); \quad \mathbf{k} = \mathbf{k}'' - \mathbf{k}', \ \beta e^2 \ll \kappa. \tag{4}$$

The quantity $V^{AS}_{\mathbf{k}'\mathbf{k}''}$, which arises in second order in the parameter $(P - P_0)/P_0$, was calculated with logarithmic accuracy under the conditions $\alpha \ll 1$, $k^2 \lesssim \alpha/\varkappa$, and $R^2 \gtrsim \varkappa/\alpha$. Substituting (4) into (1), we find

$$W^{AS}_{\mathbf{kk}'} = \frac{1}{32}\left(\frac{\kappa}{\pi\epsilon_\perp^3}\right)^{1/2}\frac{e^4 m}{\hbar^3 q} V^{AS}_{\mathbf{q}} C\delta(\epsilon_k - \epsilon_{k''}), \ \mathbf{q} = \mathbf{k} - \mathbf{k}', \tag{5}$$

$$C = 1 \text{ for } q^2 \gg \frac{\alpha}{\kappa}, \ \frac{\alpha^2}{\kappa} \ll q_z^2 \ll \kappa \cdot k^4; \ C = \frac{1}{4\pi}\left(\frac{\kappa}{2\alpha}\right)^{1/2} q \ln\left(\frac{\alpha R^2}{\kappa}\right) \text{ for } q^2 \ll \frac{\alpha}{\kappa}, q_z^2 \ll \frac{\alpha^2}{\kappa}.$$

3. Let us estimate the magnitude of the field-even current $j_z = \alpha_{zxx} E_x^2$, assuming that the momentum relaxation time τ is determined primarily by some symmetric scatterers which are present along with the centers that we discussed above. A current j_z arises as a result of momentum transfer along the z axis during an asymmetric scattering of nonequilibrium carriers which are moving predominantly along the x axis under the influence of the field E_x. Equating the rate of momentum transfer $P_x \to P_z$ to the rate of relaxation of the momentum P_z, we find

$$j_z \sim j_x\left(\frac{\mu E_x}{v_T}\right)\tau\omega, \ \omega = N\int \frac{k'_z}{k} W^{AS}_{\mathbf{kk}'} d\mathbf{k}', \ j_x = e\mu n E_x. \tag{6}$$

Here $\mu = e\tau/m$ is the carrier mobility, v_T is the thermal velocity of the carriers, $\mathbf{k}$ is a thermal wave vector ($k = mv_T/\hbar$), which is directed along the applied field (along the x axis), and N is the concentration of asymmetric scatterers. A more rigorous derivation of expression (6) could be carried out by analogy with Ref. 2. Substituting (5) into (6), and integrating, we find

$$\omega = -\frac{Ne^7 m^{3/2}}{\hbar^4} \frac{\beta P_0 \sqrt{\kappa}}{(k_B T)^{1/2}} \ln(\kappa k^2) \ln\alpha; \quad \frac{\alpha}{\kappa k^2} \ln\left(\frac{\alpha R^2}{\kappa}\right) \ll \ln(\kappa k^2). \tag{7}$$

Remarkably, according to (6) and (7) the current j_z is directed opposite the polarization P_0, regardless of the sign of the charge of the scattering centers and the carriers.

Let us examine the behavior of the current j_z near the phase-transition temperature T_0. We assume that τ and N vary only slightly over the temperature (T) range under consideration. Substituting $\alpha = \alpha_0(T_0 - T)$ and (7), we find

$$j_z/j_x \propto (T_0 - T)^{1/2} \ln(\alpha_0(T_0 - T)). \tag{8}$$

To estimate the current j_z for SbSI, we assume $N \sim 10^{18}$ cm^{-3}, $\mu \sim 50$ cm^2/(V·s), $\beta \sim 3 \times 10^{-13}$ esu, $\alpha_0 \sim 3 \times 10^{-5}$ K^{-1} and $\varkappa \sim 10^{-15}$ cm^2 (Ref. 7). We find from (6) and (7) that the ratio $j_z/j_x \sim 1\%$ should be reached in fields $E_x \sim 10^3$ V/cm, in good agreement with experiment.[5] In other ferroelectrics, having a stronger nonlinearity ($\beta \sim 10^{-9}$ esu), this current value could be reached in far weaker fields E_x.

For comparison we estimate ω for the scattering by centers with a given dipole moment d of atomic order, which was studied previously:

$$\omega = \omega_d \sim N(edk)^3/\epsilon^3 (k_B T)^2 \hbar.$$

Comparing with (7), we find $\omega/\omega_d \sim 10^2$–$10^4$. The mechanism proposed here is thus predominant, because the potential V^{AS}, in contrast with a dipole potential, falls off only slightly over distances $\sim k^{-1} \ll (\varkappa/\alpha)^{1/2}$.

4. In the paraelectric phase with $P_0 = 0$ the current is zero: $j_z = 0$. When there is a field E_z, however, a polarization $P_0' = E_z/\alpha$ arises, and the asymmetric scattering which we have been discussing here should lead to a current $j_z \sim -E_z$. According to (6) and (7), this current would be directed opposite the vector $\mathbf{P}_0'$ and thus opposite the field E_z. If j_z exceeds the conduction current $e\mu n E_z$ in magnitude, an absolute negative transverse conductivity should arise in the crystal and lead to an electrical domain instability similar to that described in Refs. 8 and 9. Using (6) and (7), we find the condition for this instability:

$$\mu E_x^2 \omega\tau / v_T \alpha P_0 > 1.$$

In SbSI, this condition could be satisfied at $E_x \sim 10^4$ V/cm for a temperature a few degrees above the point of the phase transition.

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