Magnetic control of nonlinear dielectric properties of polar crystals

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A previously predicted large variation of the dielectric properties and of the phase transition temperature T_c due to the influence of a magnetic field (~ 1 deg/kOe) was observed in a large number of ferroelectric and antiferroelectric materials. It was determined that the shifts of T_c in a field have opposite signs for ferroelectric ($\Delta T_c > 0$) and antiferroelectric ($\Delta T_c < 0$) ordering. Possible mechanisms of the effect are discussed.

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It has been shown[1-3] within the framework of the vibron theory of ferroelectric materials[4] that the dielectric properties of nonmagnetic ferroelectric crystals are altered by the influence of a magnetic field. The effect is due to a rearrangement of the electron subsystem in the magnetic field and the associated change in the vibron contribution to the frequency of the soft mode and other characteristics of the transition. These papers stimulated experimental studies of the change in the properties of polar crystals in magnetic fields. The influence of a magnetic field on the anomalies of electrical conductivity associated with the ferroelectric phase transition in $Pb_{1-x}Ge_xTe$ solid solutions was observed in Refs. 5 and 6.

This paper presents the results of the first systematic study of the direct influence of a magnetic field on the dielectric properties of a large number of polar crystals that possess different order parameters and structures (23 ferroelectric and 13 antiferroelectric crystals) in the vicinity of the phase transition. This study confirmed that the electron subsystem plays an important role in the generation of ferroelectricity and explained the characteristic features of the effect. The $\epsilon(T)$ measurements were performed in a constant, uniform magnetic field with an intensity up to 15 kOe. The $\sim$0.5-mm-thick and 5-mm-diam samples were heated by a microheater which was used to hold the sample. The sample temperature was measured by a thermocouple with ± 1°C accuracy. The capacitance was measured by a type R-385 digital microfaradmeter at a frequency of 1 kHz.

It follows from the obtained data that: 1) the influence of the magnetic field on the transition temperature T_c is very large (~ 1 deg/kOe) and does not vary significantly for different crystals; 2) the sign of $\Delta T_c(H)$ is determined by the dipole ordering: $\Delta T_c > 0$ for ferroelectric crystals and $\Delta T_c < 0$ for antiferroelectric crystals. Some typical examples are shown in Fig. 1. Figure 2 graphically illustrates the second rule by using as an example the two phase transitions–ferroelectric (FE) and antiferroelectric

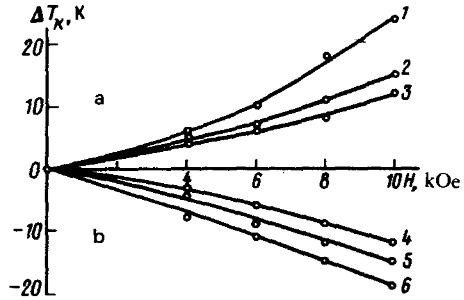

FIG. 1. Dependence of the shift $\Delta T_c = T_c(H) - T_c(0)$ on the magnetic field intensity; (a) ferroelectric materials: 1, $Bi_{0.4}Pb_{0.6}(Fe_{0.7}Nb_{0.3})O_3$; 2, $SrTi_2Ta_2O_9$; 3, $BaTiO_3$; (b) antiferroelectric materials: 1, $Pb_3V_2O_8$; 2, $PbZrO_3$; 3, $(Bi_{0.7}La_{0.3})FeO_3$.

(AFE)–that were observed in the same KNO_3 sample. We can see that the $\epsilon(T)$ peaks corresponding to ferroelectric (with cooling) and antiferroelectric (with warming) transitions change places in fields $H \gtrsim 8$ kOe.

As follows from the vibron theory, the variation of the dielectric properties in a magnetic field is determined by two factors: variation of the energy spectrum of electrons because of Landau quantization and of the vibron constants $B(\mathbf{k}, \mathbf{q})$ that characterize the relation between the electron sub-system and the polar modes of the crystal ($\mathbf{k}$ and $\mathbf{q}$ are the electron and phonon wave vectors). The first of these effects is deter-

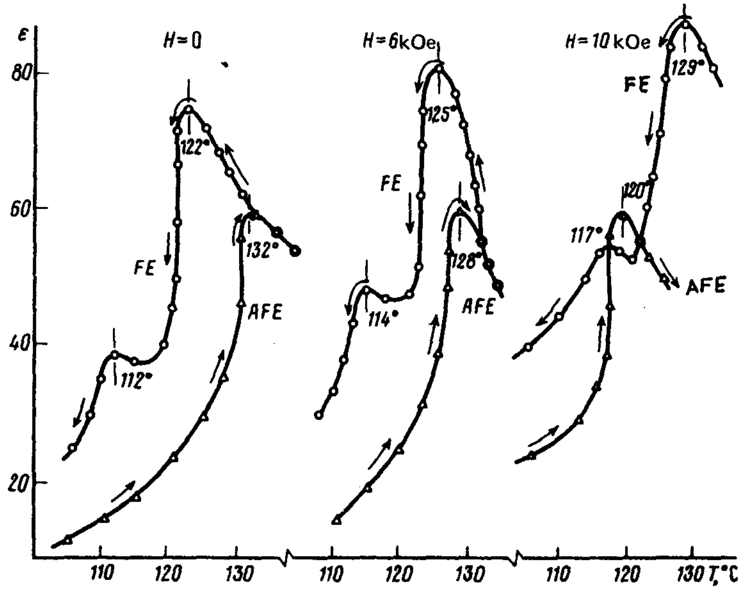

FIG. 2. $\epsilon(T)$ dependence for KNO_3 during warming and cooling in magnetic fields: $H = 0$, $H = 6$ kOe, and $H = 10$ kOe.

mined by the ratio of the cyclotron quantum to the widths of the bands. The second effect, which is due to the dispersion of $B(\mathbf{k},\mathbf{q})$, is determined by the derivative of $B(\mathbf{k},\mathbf{q})$ with respect to the electron wave vector **k**. Its contribution to T_c is proportional to the parameter a^4/R^4, which is almost independent of the crystal (a is the lattice constant and $R = \sqrt{(\hbar c/eH)}$ is the magnetic length). Estimates show that the corrections for T_c due to this mechanism can amount to 1–5% in fields of 10^4 Oe. The phase relation between $B(\mathbf{k},0)$ and $B(\mathbf{k},\pi/a)$ in this case may correct the opposite sign for ferroelectric and antiferroelectric crystals. These consequences, which are attributable to the redefinition of $B(\mathbf{k},\mathbf{q})$ in the field, are in qualitative agreement with the observed behavior; however, additional studies are necessary to obtain a better agreement between theory and experiment. Other possible mechanisms of the effect should be investigated, in particular, the role of impurities and of the domain structure.

The observed strong influence of a magnetic field on the dielectric properties of polar crystals broadens the existing ideas about the interaction of nonmagnetic materials with a magnetic field. It can also be used as a new method of investigating the structural phase transitions and as an additional tool for controlling the nonlinear, dielectric properties of polar crystals.

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