On magnetoelectric resonances in piezoelectrics

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A close relationship is shown to exist between magnetoelectric resonances (MER) — identified earlier in $BaCoF_4$ and $BaMnF_4$ — and the piezoelectric effect. MER properties are studied in detail for quartz. A high sensitivity of MER values to very small crystal deformations is shown.

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Discovery and subsequent study of so-called magnetoelectric resonances (MER) in the ferroelectrics $BaMnF_4$ and $BaCoF_4$ was reported by Al'shin and coworkers.[1] The effect consists of the occurrence, in the presence of a permanent magnetic field H, of a magnetic emission in a crystal when an alternating electric field $E^{\sim}$ at certain

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frequencies is applied to the crystal. This effect was observed in both paramagnetic and antiferromagnetic phases of the crystal. It is concluded that the nature of MERs is magnetic and they are not associated with the piezoelectric properties of the test sample.

The present report reports results of subsequent studies of this effect. The methodology is basically the same as the one described in detail in Ref. 1. The electric field $E^{\sim}$ was applied to samples by means of burned-in silver electrodes. Magnetic output was recorded by a pick-up coil connected at the input of the U2–6 amplifier.

We studied a broad class of dielectrics with very different magnetic properties (dia-, para-, antiferromagnetism) and having amorphous, poly- or single-crystal states. We showed that MERs are observed only in single-crystal samples, both, in ferromagnetics (fluorometallates, boracites), and in diamagnetics (quartz, Mg-Cl-boracite, potassium tartrate, TGS).

We found that the necessary condition for the existence of MER is the alignment of $E^{\sim}$ with the direction of the piezoelectric effect in a crystal. Thus, MERs in TGS disappeared in the course of transition from the ferroelectric phase to axisymmetric. In a quartz, MERs were observed when $E^{\sim}$ was superimposed along the x-and y-axis, and were not observed for $E^{\sim}$ along the z-axis (non-piezoelectric direction). In fused quartz glass, the effect was nonexistent.

A sharp change in the magnitude of MER was observed in Ni-Cl and Ni-I boracites, $BaMnF_4$ and $BaCoF_4$ at the magnetic transition temperatures.

We studied MER in detail in crystalline quartz which did not have magnetic and electric dipole ordering. Investigations were carried out in the temperature interval 4.2–300 K for $E^{\sim}$ ($\leqslant$3 kV/cm) applied along the x-axis in the frequency interval 0.5–5 kHz. At room temperature, MERs were observed for any relative position of the measuring coil, sample, and field **H**. Temperature measurements were carried out for the case of a magnetic field in the yz crystal plane, and the coil and x-axis coincident. MERs were observed at both $E^{\sim}$-field and doubled frequencies. The MER values were cosine functions of the magnetic field direction, and attained maxima at different directions of the field **H** for resonances at different frequencies. The field directions did not correspond to any specific directions in the test crystals. At 300 K, the width of MER (at the 0.7 level) was several tens of Hz. In the interval 100–300 K, the strongest MERs were observed at doubled frequencies of the field $E^{\sim}$. The MER width decreased with temperature drop and at 4.2 K amounted to several Hz. An increase in the resonant frequencies was observed with decreasing temperature, reading 2 Hz/K. We found that individual MERs exist only in the temperature interval of several tens of degrees, although some MERs were always in evidence at any temperature.

We observed no shift in the MER frequency at changing values of $E^{\sim}$ and H (up to 10 kOe).

The nature of the dependence of the resonant signal amplitude on field strength for the same temperature was dissimilar at different frequencies (Fig. 1), and it remained constant with increasing values of $E^{\sim}$. The dependence of MER values—observed at the fundamental and doubled frequencies—on $E^{\sim}$ values was, linear and quadratic respectively.

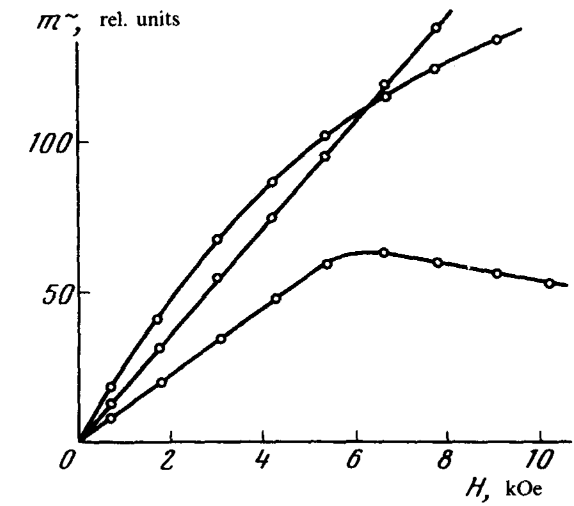

FIG. 1. Dependence of resonance on magnetic field.

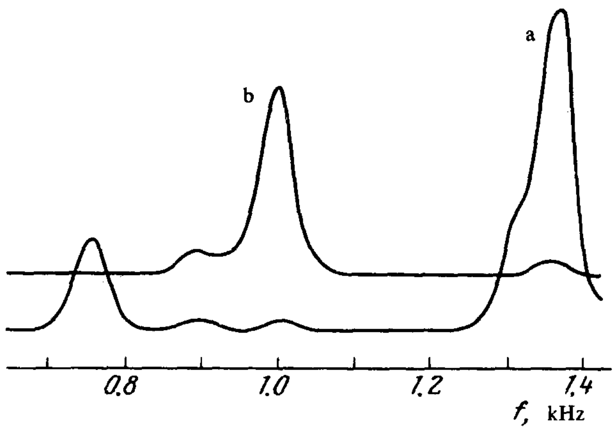

FIG. 2. Part of MER spectrum at pressure $\sigma = 300$ g/cm^2 (a) and $\sigma = 0$ (b).

It should be noted that the form of the MER spectrum at a given temperature was frequently irreproducible after thermal cycling, and from one sample to another, thus hampering the study of the physical nature of the effect. We found that one cause of irreproducibility is the extraordinary sensitivity of MER values to small crystal deformations. Thus, pressure of the order of 300 g/cm^2 in a certain direction in the *yz* plane has led to a several order of magnitude increase in the amplitude of certain MERs and virtual disappearance of others (Fig. 2). Similar effects were also observed in other piezoelectrics.

Identification of MER in a quartz indicates that the resonances are associated with the piezoelectric effect but are unassociated with magnetic or electric ordering of any type.

Low-frequency oscillations of the charged crystal lattice fail to produce a magnetic field near the crystal. Therefore, in view of the existence of MERs in samples with the most diverse physical properties, it may be assumed that MERs are associated with secondary magnetic centers whose presence is a common property of normal crystal lattices. However, questions concerning the nature of interaction of these centers with the lattice certainly remain open.

[1]B.I. Al'shin, D.N. Astrov, and R.V. Zorin, Zh. Eksp. Teor. Fiz. **63**, 2198 (1972) [Sov. Phys. JETP **36**, 1161 (1973)]; R.V. Zorin, B.I. Al'shin, and D.N. Astrov, ibid. **62**, No. 3 (1972) [Sov. Phys. JETP, **35**, No. 3 (1972)].
[2]L.D. Landau and E.M. Lifshits, Field theory, Nauka, M., 1973.