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# Increase of magnetization of a paramagnet by an alternating electric field

A. B. Brik, I. V. Matyash, and S. S. Ishchenko

*Institute of Geochemistry and Mineral Physics, Ukrainian Academy of Sciences and Institute of Semiconductors, Ukrainian Academy of Sciences*
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A new effect connected with an increase of the magnetization of a paramagnet by an alternating electric field is observed. The experiment was performed on Al–$O^-$ centers in quartz by the EPR method. The dependences of the effect on the intensity and frequency of the electric field are studied at different temperatures and crystal orientations. An interpretation of the observed phenomenon is offered, with account taken of the fact that not only the electric dipole moment but also the spin moment is connected with the Al–$O^-$ centers. It is shown that the polarization of the electric dipoles by the alternating electric field is transferred to the spin system by relaxation processes. Regardless of the polarity of the electric field, the magnetization of the paramagnet increases in this case. The observed phenomenon is in essense a relaxational magnetoelectric effect.

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Hou and Bloembergen[1] have described an increase of the magnetization of a paramagnet by an electric field. They have shown that the gist of the phenomenon reduces to the magnetoelectric effect predicted earlier[2,3] and observed in antiferromagnets[4] and in ferromagnets.[5] The change of the magnetization in these experiments is attributed to the terms of order **EH** contained in the thermodynamic potential $\Phi$ ($E$ and $H$ are the electric and magnetic field strengths). The effect takes place in both constant and alternating fields. Phenomena of this kind in paramagnets were investigated theoretically by Roitsin[6] by the spin-Hamiltonian method.

In the present study we have observed an appreciable increase (by approximately one order of magnitude) of the magnetization of a paramagnet by an alternating electric field. In its outward attributes, the observed phenomenon is similar to the magnetoelectric effect, but is substantially different with respect to the mechanism of its production and to its behavior. In particular, no increase of the magnetization was observed in a dc electric field in our experiments.

## THE EXPERIMENT

The measurements were performed by the EPR method in the 3-cm band at $T = 4.2$ K and 1.5 K, on single-crystal $\alpha$-quartz with Al–$O^-$ centers. The center density was ~$10^{18}$ $cm^{-3}$. The paramagnetism of these samples is due to a hole (spin $S = \frac{1}{2}$) localized on one of the two oxygen ions (which we shall label 1 and 2) of the $AlO_4)^{4-}$ tetrahedron in which $Si^{4+}$ is replaced by $Al^{3+}$.[7] The energy of hole localization on the two other oxygen ions is 30 meV higher.[8] Owing to the anisotropy of the $g$-factor, the holes in positions 1 and 2 produce two separately registered groups of EPR lines (six lines each, owing to the hyperfine interaction with the $Al^{27}$). For the details of the EPR and structure of the center see Ref. 7.

In our experiments, graphite electrodes were deposited directly on the crystal, and the electric field was fed to them through clamp contacts. The electric field **E** was applied perpendicular to the threefold axis of the crystal, $\mathbf{E} \perp L_3$. In addition, the condition $\mathbf{E} \parallel \mathbf{H}^0$ was satisfied, where $\mathbf{H}_0$ is the external magnetic field. The dc electric field increased the intensity of one group of lines and decreased that of the other, so that the combined net magnetization of the system remained unchanged (for details see Ref. 9).

We have observed that when an alternating electric field of frequency $\nu_E$ was applied to the crystal, the intensities of both groups of EPR lines increased to equal degrees. The effect is a maximum at $\mathbf{E} \perp L_2$ and is absent at $\mathbf{E} \parallel L_2$, where $L_2$ is the twofold axis of the defective tetrahedron in which $Si^{4+}$ is replaced by $Al^{3+}$. In these experiments, the EPR signals were observed on an oscilloscope screen. The frequencies of the electric and magnetic fields were rational multiples, and the signals were obtained at the maximum amplitude of the electric field. The increase of the signal was independent of the polarity of the electric field. When an automatic plotter with a time constant larger than $\nu_E^{-1}$ was used as a recorder, the averaged effect

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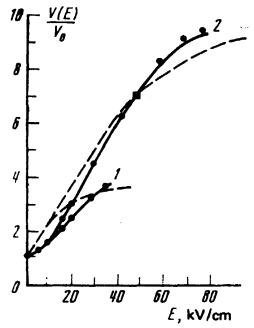


FIG. 1. Increase of the EPR line intensity $V(E)$ vs. the intensity of the alternating electric field at temperatures $T = 1.5$ K (1) and 4.2 K (2). $\nu_E = 200$ Hz, $\theta = 60°$, $V_0$ is the EPR signal intensity at $E = 0$. Dashed lines—theory, solid lines—experiment, ■—fit of experiment to theory.

was observed. When the signals were displayed on an oscilloscope screen, the frequency of the alternating modulating magnetic field was $\nu_H \approx 50$ Hz and its amplitude was $H_m \approx 20$ Oe, and when an automatic plotter was used the corresponding values were $\nu_H = 83$ Hz and $H_m \leq 0.1$ Oe.

Figure 1 shows the dependence of the increase of the EPR signal as a field of the maximum amplitude of the electric field intensity at $T = 4.2$ and 1.5 K. At low intensities of **E** (see Fig. 1) the effect is temperature-independent, while at high intensities the effect decreases with decreasing crystal temperature. The error in the measurement of the relative line intensity did not exceed ±10% in weak fields and ±5% in strong ones.

Figure 2 shows the dependence of the effect on the frequency $\nu_E$. It is seen that when the crystal temperature is lowered from 4.2 to 1.5 K the curves shift towards lower frequencies; the frequency corresponding to the maximum of the effect decreases by approximately a factor of three.

No changes in the shapes and positions of the resonance lines, were observed in either alternating or constant electric fields ($E \leq 80$ kV/cm), thus indicating that the usual[10] electric-field effects are weak. There was no microwave saturation in the described experiments.

## DISCUSSION OF EXPERIMENT

In quartz containing Al–O$^-$ centers, the $Al^{3+}$ and O$^-$ ions (the hole is localized on O$^-$) form an electric dipole oriented along the Al–O bond. Experiments with a constant electric field, as well as measurements of the dielectric losses[9,11] indicate that the holes can hop over from the oxygen ion 1 to oxygen ion 2, meaning that reorientation of the electric dipole is possible.

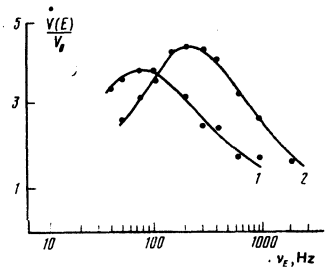


FIG. 2. Dependence of the EPR line gain on the electric-field frequency at temperatures $T = 1.5$ K (1) and 4.2 K (2). $E = 29$ kV/cm, $\theta = 60°$.

If $\mathbf{E} \perp L_3$, then, when account is taken of the data on the quartz structure, the energy difference of the dipoles oriented along the two possible directions can be represented in the form

$$U_E = 1.4 dE \sin\theta, \tag{1}$$

where $d$ is the value of the dipole, $\theta$ is the angle between **E** and the $L_2$ axis of the defective tetrahedron, and **E** is the field acting on the dipole,

$$E = E \sin 2\pi\nu_E t.$$

According to Ref. 12, in the case of a dielectric such as quartz the electric field acting on the dipole is equal to the external field.

The energy level scheme of the considered system, in the presence of a constant magnetic field $\mathbf{H}_0$ and an alternating electric field, is shown in Fig. 3, where $U_H = g\beta H_0$ is the Zeeman energy. The splitting of the Zeeman levels due to the hyperfine interaction is not shown in the figure.

The ratio of the magnetizations of the system in the presence and absence of an electric field will be defined in general form by the expression

$$\frac{M(E)}{M_0} = \frac{\Delta n_1(E) + \Delta n_2(E)}{\Delta n_1^0 + \Delta n_2^0}, \tag{2}$$

where $\Delta n_1^0$ and $\Delta n_2^0$ are the population differences of the Zeeman sublevels for hole localization on the oxygen ions 1 and 2 at $E = 0$.

The following relaxation processes can take place in the considered system: electric-dipole $T_d(U_E)$, spin $T_s(U_H)$, and mixed $T_{ds}(U_E + U_H)$ and $T_{ds}(U_E - U_H)$. The quantities in the parentheses are the energy gaps of those levels between which the corresponding transitions occur. In Fig. 3 these transitions are numbered 1, 2, 3, and 4, respectively.

A rigorous calculation of (2), starting from the level populations of the considered four-level system, is in principle a straightforward but most cumbersome task.

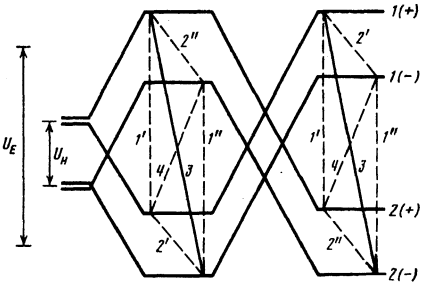


FIG. 3. Picture of energy levels of the considered system in the presence of a constant magnetic field and an alternating electric field. The 1(±) and 2(±) indicate the oxygen ion on which the hole is localized, and the ± signs correspond to the spin projection on the magnetic field.

To avoid unwieldy expressions and to emphasize the gist of the observed phenomenon, we present therefore the results of a simplified calculation that preserves at the same time the main features of the actual processes.

It can be shown in general form that to obtain an increase in the magnetization, $M(E)/M_0>1$, the following condition must be satisfied:

$$T_{ds}(U_E+U_H)<T_{ds}(U_E-U_H). \quad (3)$$

The reorientation of the electric dipoles in $SiO_2$ takes place, according to Ref. 11, by phonon-induced tunneling. In this case the probability of the transitions is proportional to the phonon density at the frequency of the corresponding transition.[11] Since the energy gap between the levels coupled by the transition 3 is always larger than that between levels coupled by transition 4, this may be the factor that ensures satisfaction of condition (3).

The transitions $1'$ and $1''$ act during the first and second half-cycles of the electric field between the same levels. Whereas during the first half-cycle (see Fig. 3) the transition $1'$ decreases the population of the level 2(+) and decreases that of level 1(+), in the second half-cycle, on the contrary, it decreases the population of level (1+) and increases that of level (2+), the action of the transitions $1'$ and $1''$ in an alternating electric field thus averages out to zero. It can be furthermore assumed, on the basis of experiments on EPR-signal saturation,[9] that the transitions $2'$ and $2''$ alter very slowly (compared with the period of the electric field) the differences of the Zeeman sublevel populations. Taking the foregoing into account, assuming that $T_{ds}(U_E+U_H)\ll T_{ds}(U_E-U_H)$, and recognizing that the EPR signal is proportional to the magnetization, we obtain

$$\frac{V(E)}{V_0}=\frac{M(E)}{M_0}=\frac{1-\exp[-(U_E+2U_H)/kT]}{\{1+\exp[-(U_E+U_H)/kT]\}[1-\exp(-U_H/kT)]}. \quad (4)$$

The effect described by Eq. (4) can be qualitative explained as follows. During the first half-cycle (see Fig. 3) the process $T_{ds}(U_E+U_H)$ decreases the population of the level 2(+) and increases that of level 1(−). This increases the population difference of both pairs of the Zeeman sublevels. During the second half-cycle the very same process acting between levels 1(+) and 2(−), decreases the population of the level 1(+) and increases that of level 2(−). Thus, the differences $\Delta n_1(E)$ and $\Delta n_2(E)$ increase during the first as well as the second half-cycles.

The dependences of $V(E)/V_0$ on the maximum aplitude of the alternating electric field, calculated in accord with (4) at $T=4.2$ and 1.5 K, are shown by the dashed lines in Fig. 1. The dipole value determined from the point at which the theory and experiment are reconciled turned out to be ≈5 D, close to the value obtained from experiment with constant electric fields.[9,11] This serves as a confirmation that expression (4) is a correct reflection of the gist of the processes that occur in the investigated system.

Expression (4), with account taken of formula (1), in which it is assumed that the dipole is oriented along the Al–$O^-$ bonds, explains also the dependence of the magnitude of the effect on the crystal orientation, since $U_E\approx 0$ at $\mathbf{E}\parallel L_2$.

It is seen from Fig. 1 that (4) describes satisfactorily the course of the plots, the independence of $V(E)/V_0$ of the temperature in the initial section, as well as the decrease of the effect with decreasing temperature at large $E$. The systematic discrepancy between theory a and experiment at small $E$ can be explained in the following manner. Obviously, $T_{ds}(U_E+U_H)$ and $T_{ds}(U_E-U_H)$ are functions of $E$. At $E\approx 0$

$$T_{ds}(U_E+U_H)\approx T_{ds}(U_E-U_H),$$

and in this case, as indicated $M(E)/M_0=1$.

Starting from the value of $\nu_E^m$ corresponding to the extremum of the experimental curves on Fig. 2, we can estimate the dipole-reorientation time, by setting it, as usual equal to $(2\pi\nu_E m)^{-1}$. At $T=4.2$ K we have $(2\pi\nu_E m)^{-1}\approx 0.9\times 10^{-3}$ sec, which agrees with the time obtained from dielectric-loss experiments.[9,11] The decrease of the effect at $\nu_E<\nu_E^m$ is obviously explained by the fact that in addition to $T_{ds}(U_E+U_H)$, other depolarizing processes have time to come into play, and at $\nu_E>\nu_E^m$ the dipoles are incapable of following the variation of the electric field. From the size of the shift of the curves on Fig. 2 at $T=4.2$ and 1.5 K it follows that the time of dipole reorientation is proportional to $T^{-1}$ in this temperature range.

The observed phenomenon is in essence a relaxational magnetoelectric effect. Its observation requires that the center have an appreciable electric dipole moment (with several possible orientations) and of an associated spin moment, as well as a favorable ratio of the relaxation times. The effect is easiest to observe in systems comprising a paramagnetic center (PC) amd impurity, a PC and a vacancy, etc. Since the increase of the magnetization is appreciable in the considered phenomenon, the effect can be registered in measurements of the static susceptibility. We note that the described experiments serve as a new method of polarizing electron spin, and under appropriate conditions thay can also increase the nuclear polarization. In addition, they uncover a possibility of studying in detail the transition of a charged particle with spin from one potential well to another.

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# Effect of vacancies on NMR is solid $He^3$

I. N. Piradashvili

*Physics Institute, Georgian Academy of Sciences*
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The influence of ferromagnetically polarized regions (FPR) produced by vacancies in solid $He_3$ on the spin-lattice and spin-spin relaxation times and on the spin diffusion coefficient is investigated. The FPR absorption line shape is calculated for nonequilibrium vacancy densities at $T \sim 10^{-2}$ K. The values of the FPR diffusion coefficient of the numbers of the spins contained in the FPR are determined from an analysis of the experimental data.

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## 1. INTRODUCTION

The principal characteristic feature of a quantum crystal is the large amplitude of the zero-point vibrations of its atoms and their delocalization as a result of the overlap of the atomic wave functions. The quantum-mechanical tunneling ability manifests itself in the existence of an excitation system that is absent from non-quantum crystals. The mobility of these quasiparticles and their interaction is easy to investigate in solid $He^3$, since the presence of spin-$\frac{1}{2}$ particles in it permits the use of the sensitive methods of nuclear magnetic resonance (NMR).

The available experimental data on NMR in solid $He^3$ are summarized mainly in the review of Guver *et al.*[1] The system of nuclear spins ($Z$) in a constant external magnetic field $H_0$, described by a Zeeman Hamiltonian, arrives at thermal equilibrium with a definite temperature (spin-spin relaxation) via a dipole-dipole interaction that is modulated by the motion by one of the types of the quasiparticles. The same mechanism is responsible also for the spin-lattice relaxation.

The first part of the paper deals with the temperature region $T \sim 1.5$ K, where the dipole-dipole interaction is modulated by delocalized vacancies—vacancions ($V$).

In Ref. 1, the vacancion is regarded as a "bare" quasiparticle that moves through a homogeneous medium. Yet a vecancion in solid $He^3$ produces around itself a ferromagnetically polarized region (FPR) of nuclear spins,[2] and this region influences substantially the vacancion mobility.[3] The character of the modulation of the dipole-dipole interaction is thereby altered.

We derive below the dependences of the spin-lattice and spin-spin relaxation times, as well as of the diffusion coefficient $D_z$ of the nuclear magnetization, on the size and mobility of the FPR. Reduction of the experimental data[4-6] by means of these formulas makes it possible to determine the number of spins contained in the FPR and the diffusion coefficient $D_V$ of a vacancion "dressed" in this manner, and to estimate its band width.

At $T < 10^{-2}$ K the FPR can be regarded as almost immobile,[3] so that their influence on a nonstationary process such as relaxation is very small even for large (nonequilibrium) FPR densities. On the other hand, the strong exchange interaction between the $He^3$ nuclei leads to a relation between the direction of the FPR magnetic moment and the polarization of the nuclear paramagnet. By the same token, the existence of the FPR influences the absorption line shape in the case of stationary detection of the NMR signal. In the second part of the paper, this dependence is derived for weak and strong constant magnetic fields.

The conclusion deals with the conditions under which the results are valid, and contains some numerical estimates.

## 2. RELAXATION AND DIFFUSION

### Relaxation times

At $T \sim 1.5$ K the $V$ system is closely coupled to the lattice and they have the same constant temperature. The temperature of the $Z$ system, which is "heated" by the RF field, approaches asymptotically the temperature of the $V$ system. The large mobility of the nuclear spins in solid $He^3$ leads to a rapid change of the Hamiltonian of the dipole-dipole interaction, so that it is possible to use for the calculations the short-correlation-times approximation (see Ref. 7, Chap. VIII). We ob-

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