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Electric-field effect in electron–nuclear double resonance of Cr^{3+} ions in ruby

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External static electric fields up to 800 kV/cm were applied in a study of the linear effect of these fields on the spectra of electron–nuclear double resonance of $(^{53}Cr)^{3+}$ impurity ion nuclei and "distant" ^{27}Al nuclei in ruby. The measured splitting of the spectral lines was used to find the electric-field-induced changes in the hyperfine interaction parameter of the impurity ions and in the electric field gradients at the ^{53}Cr and ^{27}Al nuclei. The results indicated that the mechanisms responsible for the changes in the electric-field gradients were the same for both nuclei. Estimates were obtained of the orbital contribution and the contribution of the polarization of the ion cores to the change in the hyperfine interaction parameter. The change in the electron g factor of the impurity ions was estimated.

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INTRODUCTION

The capabilities of the electron-nuclear double resonance (ENDOR) method in investigations of paramagnetic centers in crystals can be greatly extended by the use of external agencies and, in particular, by the application of static electric fields.[1,2] High fields of 10^5–10^6 V/cm intensity are needed to observe and measure the elec-

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tric-field effect in ENDOR spectra.[3,4] The main difficulty which hinders experimental utilization of the field effect is the creation of such high electric fields in the interior of a sample located inside a spectrometer resonator. Earlier investigations of the influence of electric fields on ENDOR have been limited to alkali halide compounds with F centers. Only recently studies have been made of the ENDOR spectra of a crystal containing impurity paramagnetic ions and subjected to an external electric field: the field effect was determined for the ligand ENDOR of Gd^{3+} ions in CaF_2 (Ref. 4).

Crystals containing paramagnetic centers as impurity ions may exhibit not only the ligand ENDOR due to the nuclei of the ions themselves (if they have a nonzero nuclear spin) but also "distant" ENDOR due to nuclei located some distance away from the paramagnetic centers.[5,6]

Our aim was to investigate experimentally the electric-field effect in the spectra of the two types of ENDOR mentioned above. Our material was ruby (α–Al_2O_3:Cr) exhibiting the spectra of impurity ions and distant nuclei. In both cases the linear field effect was observed. This effect was determined by our own method for creating high electric fields in samples, which was used earlier[4] and described there in greater detail.

The results obtained were used to determine the electric-field-induced changes in the hyperfine interaction parameter of the $(^{53}Cr)^{3+}$ impurity ions and in the electric field gradients at the nuclei of these ions and also at distant ^{27}Al nuclei. Estimates were obtained of the change in the electron g factor of the impurity ions of the contributions of the various mechanisms to the changes in the hyperfine interaction parameters.

SPIN HAMILTONIAN AND SPECTRUM

Our ruby crystals contained paramagnetic impurities in the form of a natural mixture of the chromium isotopes. The odd ^{53}Cr nuclei with a natural abundance of 9.5% have a nonzero nuclear spin $I = 3/2$ so that ENDOR of these nuclei is possible. The ground state of the $3d^3$ ions of Cr^{3+} in ruby (local symmetry C_3) is an orbital singlet with the spin $S = 3/2$. In the absence of external agencies the energy spectrum of the $(^{53}Cr)^{3+}$ ions is described by the spin Hamiltonian[7,8]

$$\mathcal{H}_{Cr}=g_{\parallel}\beta H_z S_z+g_{\perp}(H_xS_x+H_yS_y)+D[S_z^2-{}^1/_3S(S+1)]+AS_zI_z+B(S_xI_x+S_yI_y)-g_n\beta_n(H_xI_x+H_yI_y+H_zI_z)+Q_{Cr}'[I_z^2-{}^1/_3I(I+1)], \quad (1)$$

where $g_{\parallel} = 1.9840$, $g_{\perp} = 1.9867$, $2D(4.2°K) = -11.447$ GHz, $A = B = 48.5$ MHz, $g_n\beta_n = -0.2406$ kHz/Oe, and $Q'_{Cr} = -0.21$ MHz.

When the magnetic field is parallel to the z axis, which coincides with the crystal axis C_3, the energy levels corrected for the second-order hyperfine interactions, are

$$W_{M,m}=g_{\parallel}\beta HM+D[M^2-{}^1/_3S(S+1)]+AMm-g_n\beta_nHm+Q_{Cr}'[m^2-{}^1/_3I(I+1)]+\frac{1}{4}B^2\frac{[S(S+1)-M(M+1)][I(I+1)-m(m-1)]}{-g\beta H-D(2M+1)+A(M-m+1)}+\frac{1}{4}B^2\frac{[S(S+1)-M(M-1)][I(I+1)-m(m+1)]}{g_{\parallel}\beta H+D(2M-1)+A(m-M+1)}, \quad (2)$$

where M and m are the eigenvalues of the operators S_z and I_z, respectively. The ENDOR resonance frequencies, governed by the allowed $(M, m) \leftrightarrow (M, m-1)$ transitions, can be expressed in the form

$$\nu(M,\ m\leftrightarrow m-1)=(W_{M,m}-W_{M,m-1}). \quad (3)$$

Each line in the spectrum is a superposition of components representing nuclei occupying crystallographically inequivalent positions in the Al_2O_3 lattice. In an external electric field the components of the lines are shifted: the shifts of the contributions of the nuclei coupled by the inversion operator are equal and opposite, i.e., the lines experience a pseudo-Stark splitting $\Delta(M, m \leftrightarrow m-1)$, equal to the doubled shift of its components:

$$\Delta(M,\ m\leftrightarrow m-1)=2|\Delta\nu(M,\ m\leftrightarrow m-1)|. \quad (4)$$

The splittings of the lines observed when the C_3 axis is parallel to the magnetic and electric fields ($\mathbf{H}\|\mathbf{E}\|\mathbf{C}_3$) can be explained by the linear (in respect of the field) changes in the parameters D, A, B, and Q'_{Cr} of the Hamiltonian (1), which are altered by the electric field to

$$D\pm{}^1/_2R_{333}E_z,\quad A\pm\alpha,\quad B\pm\beta,\quad Q_{Cr}'\pm\gamma, \quad (5)$$

where

$$\alpha=\frac{\partial A}{\partial E_z}E_z,\quad \beta=\frac{\partial B}{\partial E_z}E_z,\quad \gamma=\frac{\partial Q_{Cr}'}{\partial E_z}E_z; \quad (6)$$

the different signs of the changes in the parameters correspond to the opposite shifts of the line components. The change in D has been determined earlier[9] and it is given by the constant

$$R_{333}=\frac{2}{3}\frac{\partial D}{\partial E_z}=0.180\frac{\text{MHz}}{\text{kV/cm}}. \quad (7)$$

The resonance frequencies of the line components in an electric field are found from Eqs. (2) and (3) with the modified parameters given by Eq. (5). The differences between the resonance frequencies, governing the line splitting, depend on three unknowns: α, β, and γ, which can be found by measuring the splitting of at least three lines in the spectrum.

The ENDOR spectrum of distant ^{27}Al ($I = 5/2$) nuclei coupled very weakly to the paramagnetic centers appears because the distant ENDOR mechanism operates in ruby[5,6]: this mechanism can be used to record the NMR spectrum of aluminum from changes in the intensities of the saturated microwave lines of Cr^{3+}. Therefore, as in the work of Dixon and Bloembergen,[10] who studied the influence of an electric field on the NMR of the Al nuclei in ruby, we shall describe the ENDOR spectrum by the following Hamiltonian which contains only the nuclear–Zeeman and quadrupole–interactions:

$$\mathcal{H}_{Al}=-g_n\beta_n\mathbf{HI}+Q_{Al}'[I_z^2-{}^1/_3I(I+1)], \quad (8)$$

where $Q'_{Al} = 0.180$ MHz. In this case the electric field effect is due to a change in the value of Q'_{Al}. The parameters Q' are connected to the electric-field gradients at the nuclei by

$$Q'=\frac{3eQ}{4I(2I-1)}q, \quad (9)$$

where Q is the quadrupole moment of the nucleus and q is the electric field gradient; therefore, the changes in Q' are governed by the changes in q. The line splitting

in the $\mathbf{H}\|\mathbf{E}\|\mathbf{C}_3$ case is given by

$$\Delta(m \leftrightarrow m-1)=2\frac{\partial Q_{A1}'}{\partial E_z}(2m-1)E_z. \quad (10)$$

which allows us to find the value of $\partial Q_{A1}'/\partial E_z$ from the measured splittings.

EXPERIMENTAL METHOD

The electric-field effect was determined for ruby samples with a chromium concentration of ~0.05% when the electric and magnetic fields were parallel to the C_3 axis and the temperature was $T = 4.2$°K. High electric fields were created in the samples by the following method. Oriented plane–parallel ruby plates of $5 \times 10 \times 0.25$ mm dimensions were coated with conducting films to which flexible Teflon-insulated wire electrodes were welded. These electrodes retained their flexibility without disturbance of the Teflon insulation in a wide range of temperatures, including liquid helium. The edges of the samples, including narrow parts of the conducting films, were covered along the perimeter by a thin layer of an insulating silicone adhesive (Fig. 1b). Such insulation was sufficient for the application of high voltages up to 20 kV to the conducting films. Care was taken to avoid deposition too much of this adhesive because, as found at $T = 4.2$°K, this deformed the sample at low temperatures as a result of the difference between the thermal expansion coefficients of the adhesive and ruby.

Deformations in ruby were revealed by the value of the initial splitting parameter D, which was sensitive to mechanical stresses.[11] Samples with a thin adhesive coating along the edges were not significantly deformed. The parts of the conducting films not covered by the adhesive were protected from damage by an additional coating with a mixture of picein and quartz powder. A sample was then placed in a rotatable polystyrene holder (Fig. 1c) and located in the middle of a microwave resonator inside an rf pump coil. The ability to rotate the holder in liquid helium made it possible to establish the required orientation in the determination of the angular dependences of the spectra and this could be done with the precision needed in the ENDOR experiments.

The spectra were recorded in a high-sensitivity superheterodyne spectrometer ($f_0 = 9.096$ GHz) in which the signal was provided by a rectangular ceramic resonator of the H_{102} type with a silver coating of the inner surface. The thickness of the coating was such (~5 μ) that its heating by rf eddy currents was negligible and no additional noise appeared in the resonator because of boiling of helium. The silver coating was easily penetrated by a modulating magnetic field generated by coils placed on the outer sides of the wide walls of the resonator. Nuclear transitions were excited by a wide-band nuclear pump system, which created an alternating magnetic field of up to 10 Oe amplitude in a matched rf coil.

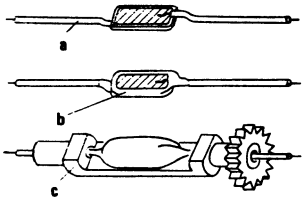

FIG. 1. Sample of ruby used in measurements of the electric field effect in ENDOR: a) electrodes in Teflon insulation; b) insulating coating of silicone adhesive; c) rotatable holder with sample.

In selecting the conditions for the recording of the spectra, an allowance was made for the following two factors which facilitated measurements of the electric-field effect. Firstly, when the magnetic field was oriented along the C_3 axis, the central ESR line of Cr^{3+} corresponding to the $1/2 \longleftrightarrow -1/2$ transition was not split by the electric field.[9] Secondly, without going outside the range of resonance conditions for this line, it was possible to record the ENDOR lines for all values of M between +3/2 and −3/2 (Ref. 7). Therefore, the ENDOR spectra in an external electric field were recorded for the $1/2 \longleftrightarrow -1/2$ electron transition.

The ENDOR spectrum of the Al nuclei was observed under conditions similar to those in Ref. 7. A magnetic field was modulated at a frequency $\nu_m = 3$ kHz and a synchronous detector was tuned to this frequency. The signal intensity was maximal when the resonance magnetic field corresponded to a slope of the ESR line.

The ENDOR spectrum of the ^{53}Cr nuclei was determined without modulation of the magnetic field but in this case the nuclear pump frequency was modulated ($\nu_m = 300$ Hz). Both frequency and amplitude modulation methods were used. The adoption of the pump frequency modulation technique made it possible[8] to resolve the lines of the triplet with $M = -1/2$ that were not resolved by the method used in Ref. 7. The relative intensities of the triplet components depended on the resonance magnetic field; variation of the field within the ESR line profile made it possible to spearate each of the outer lines, suppressing almost completely the other two lines of the triplet. This feature of the ENDOR spectrum enabled us to determine the splitting of the outer lines of the triplet in an electric field. The spectrum could be recorded in just two values of the magnetic field corresponding to the resonance conditions of the outer lines of the hyperfine structure of the $1/2 \longleftrightarrow -1/2$ electron transition in the ESR spectrum of $(^{53}Cr)^{3+}$. The ENDOR lines corresponding to the $(M, 3/2) \longleftrightarrow (M, 1/2)$ transition were recorded for all the values of M in a field $H \approx 3250$ Oe and the other lines in a field $H = 3304$ Oe. The splitting of one of the lines in an electric field is shown in Fig. 2.

EXPERIMENTAL RESULTS AND DISCUSSION

The electric-field effect and the nature of its dependence on the field intensity were determined by recording the ENDOR spectral lines of the ^{53}Cr nuclei in fields ranging up to 600 kV/cm, and for some lines up to 800 kV/cm. The splitting was linear for all the lines. The expressions for the line splitting, obtained from Eqs. (2) and (3) using the parameters (5), and the results of measurements made it possible to derive a system of equations for the determination of α, β, and γ.

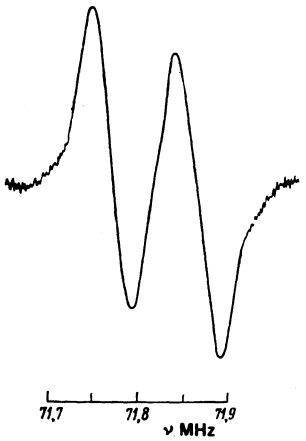

FIG. 2. Splitting of an ENDOR line due to ^{53}Cr nuclei undergoing the $(-3/2, -1/2) \leftrightarrow (-3/2, -3/2)$ transition in an electric field $E = 600$ kV/cm with the $E \| H \| C_3$ orientation.

This system has the following form when it is restricted to terms linear in β and the parameters are expressed in kilohertz:

$$\left.\begin{aligned}
&\Delta(^3/_2, ^1/_2 \leftrightarrow -^1/_2) = 3\alpha + 0.084\beta + 186 = 260 \pm 10 \quad (140),\\
&\Delta(^3/_2, -^1/_2 \leftrightarrow -^3/_2) = 3\alpha - 0.084\beta - 4\gamma - 242 = 203 \pm 13 \quad (138),\\
&\Delta(^1/_2, ^3/_2 \leftrightarrow ^1/_2) = \alpha - 0.127\beta + 4\gamma - 166 = -200 \pm 100 \quad (\sim 500),\\
&\Delta(^1/_2, ^1/_2 \leftrightarrow -^1/_2) = \alpha + 0.059\beta + 212 = 300 \pm 100 \quad (\sim 500),\\
&\Delta(^1/_2, -^1/_2 \leftrightarrow -^3/_2) = \alpha + 0.236\beta - 4\gamma + 630 = 720 \pm 100 \quad (\sim 500),\\
&{}^*\Delta(-^1/_2, ^3/_2 \leftrightarrow ^1/_2) = \alpha + 0.0003\beta - 4\gamma + 8 = 6 \pm 3 \quad (22),\\
&{}^*\Delta(-^1/_2, -^1/_2 \leftrightarrow -^3/_2) = \alpha + 0.057\beta + 4\gamma - 3 = 43 \pm 2 \quad (34),\\
&\Delta(-^3/_2, ^3/_2 \leftrightarrow ^1/_2) = 3\alpha - 0.007\beta - 4\gamma - 2 = 50 \pm 10 \quad (\sim 50),\\
&\Delta(-^3/_2, ^1/_2 \leftrightarrow -^1/_2) = 3\alpha + 0.007\beta + 3 = 80 \pm 10 \quad (\sim 50),\\
&{}^*\Delta(-^3/_2, -^1/_2 \leftrightarrow -^3/_2) = 3\alpha + 0.021\beta + 4\gamma + 8 = 98 \pm 3 \quad (48),
\end{aligned}\right\} \quad (11)$$

The right-hand sides of the system (11) give the experimental values of the splitting $\Delta(M, m \leftrightarrow m-1)$ in an electric field $E = 600$ kV/cm. The numbers in parentheses are the widths (from one peak of the derivative to the next one) of the corresponding ENDOR lines in $E = 0$. The last terms in the equations represent the contribution made to the splitting by the known change in the value of D. It is difficult to measure the line splitting for the $(-1/2, 1/2) \leftrightarrow (-1/2, -1/2)$ and $(3/2, 3/2) \leftrightarrow (3/2, 1/2)$ transitions because of the smallness of the signal/noise ratio and, therefore, the equations for these transitions are not included in the system (11).

A comparison of the splittings and also of the line widths for different values of M shows that they are considerably greater for the positive values of M than for the negative ones. A similar dependence is observed also for the intervals between the lines in the triplets with different values of M. We can explain these observations by the fact that the expression for the energy (2) contains second-order hyperfine terms describing the contribution of the pseudonuclear electric quadrupole interaction. As mentioned in Ref. 8, the magnitude of this contribution for $M = -1/2, -3/2$ is comparable with the magnitude of the true quadrupole interaction and is an order of magnitude greater than in the $M = +1/2, +3/2$ case. It should also be noted that in a magnetic field corresponding to the resonance of the $1/2 \leftrightarrow -1/2$ transition the magnitude of the second-order terms for $M = +1/2, +3/2$ depends strongly on D. This explains the considerable width of the spectral lines for the positive values of M, which is due to scatter of the values of D from one point to another inside a sample.

The value of D depends strongly on the electric field applied along the C_3 axis.[9] Consequently, the splitting of the lines in the $M = +1/2, +3/2$ case is dominated by the electric-field-induced changes in D. For the same reason the signs of the splitting given in Eq. (11) are governed by the sign of the change in D.

We determined the values of α, β, and γ by selecting the three starred equations in which the D-dependent terms are small. The solutions obtained satisfy, within the limits of the experimental error, the other equations of the system (11). From the values of α and γ obtained in this way it follows, subject to the notation of Eq. (6), that

$$\frac{\partial A}{\partial E_z} = 37 \pm 4 \frac{\text{Hz}}{\text{kV/cm}}, \quad \frac{\partial Q_{cr}'}{\partial E_z} = 9.3 \pm 1 \frac{\text{Hz}}{\text{kV/cm}};$$

the signs of these quantities are identical with the signs of $\partial D/\partial E_z$. Determination of the values of $\partial B/\partial E_z$ from the system (11) is difficult because the contribution made by β to the line splitting is small and does not exceed the experimental error. However, estimates indicate that $\partial B/\partial E_z$ is comparable with, or smaller than, $\partial A/\partial E_z$.

The quantity $\partial A/\partial E_z$ represents the total change in the components of the constant A which are governed by the various hyperfine interaction mechanisms. For the $(^{53}Cr)^{3+}$ ions and other $3d^3$ ions in Al_2O_3 (V^{2+}, Mn^{4+}), which are in the singlet orbital ground state, a considerable contribution to the hyperfine interactions is made by two mechanisms: polarization of the ion cores and admixture to the ground state—in which the orbital momentum is frozen—of states with nonzero orbital momentum.[12] Thus, the parameter A splits into two components:

$$A = A^{po} + A^{orb}, \quad (12)$$

where the isotropic quantity A^{po} represents the hyperfine magnetic field H^{po} at the nucleus, due to the polarization of the ion core, and A^{orb} represents the orbital hyperfine field H^{orb}, proportional to the change in the g factor $\Delta g_{\parallel} = g_{\parallel} - 2.0023$. It is known that for our ions the total hyperfine field at the nuclei and, consequently, the value of A are governed almost entirely by the ion core polarization.[12] In accordance with Eq. (12), the change in the parameter A on application of an electric field is given by

$$\frac{\partial A}{\partial E_z} = \frac{\partial A^{po}}{\partial E_z} + \frac{\partial A^{orb}}{\partial E_z}.$$

The contribution of $\partial A^{orb}/\partial E_z$ to the experimental value of $\partial A/\partial E_z$ can be estimated using the known value of $\partial D/\partial E_z$ from Eq. (7) and the expression[12]

$$\delta H^{orb} = 1.25 \cdot 10^5 \frac{4}{3\lambda} \delta D \left\langle \frac{1}{r^3} \right\rangle [\text{G}].$$

($\langle 1/r^3 \rangle$ is in atomic units) relating the change in the initial splitting parameter and the orbital hyperfine field. Assuming $\lambda = 70$ cm^{-1} for Cr^{3+} (Ref. 13) and $\langle 1/r^3 \rangle = 3.959$ a.u., we find that $\partial H^{orb}/\partial E_z = 0.087$ G/(kV/cm) and hence

$$\frac{\partial A^{orb}}{\partial E_z} = -g_n \beta_n \frac{\partial H^{orb}}{\partial E_z} = 21 \frac{\text{Hz}}{\text{kV/cm}}.$$

When the above value is used, we find that $\partial A/\partial E_z$

agrees with the experimental result if we assume that $\partial A^{po}/\partial E_z = 16$ Hz/(kV/cm). Thus, over half the measured value of $\partial A/\partial E_z$ is due to the change in A^{orb}. Since the orbital part of the hyperfine interaction constant is related to the electron g factor by

$$A^{orb} = 2\beta g_n \beta_n \langle 1/r^3 \rangle (g_\parallel - 2),$$

it follows that measurements of the change in A^{orb} give the change in $g_\parallel$. The experimental value of the change in $g_\parallel$ in an electric field $E = 600$ kV/cm is $\delta g_\parallel \approx 1 \cdot 10^{-4}$. It should be noted that the ESR method is incapable of detecting such a small electric-field-induced change in the g factor.[9]

It is worth noting the great disproportion between the values of A^{po} and A^{orb} ($A^{po} \gg A^{orb}$) and their relative changes, which have the following values in a field $E = 600$ kV/cm:

$$\delta A^{po}/A^{po} \approx 2 \cdot 10^{-4}, \quad \delta A^{orb}/A^{orb} \approx 6 \cdot 10^{-2}.$$

This disproportion is not unexpected if we bear in mind that in the case of the $3d^n$ ions the hyperfine magnetic field at the nucleus H^{po} is independent, to within <1%, of the distance to ligands if the chemical environment is the same. The same constancy of the field applies also to the sequence of the $3d^n$ ions in the same chemical environment. However, considerable changes in the constant A occur because of its small orbital part A^{orb} (Ref. 12). This feature of the hyperfine interaction allows us to assume that the experimentally observed relative electric-field-induced change in A^{po} is small compared with the relative change in A^{orb}.

The electric-field effect in ENDOR of the distant Al nuclei was measured in electric fields up to 800 kV/cm. Splitting of the spectral lines corresponding to the $\pm 1/2 \longleftrightarrow \pm 3/2$ and $\pm 3/2 \longleftrightarrow \pm 5/2$ transitions varied nonlinearly with the field. Figure 3 shows the dependence of the splitting of the $-3/2 \longleftrightarrow -5/2$ transition plotted on the basis of our measurements and the data of Dixon and Bloembergen.[10] It is clear from Fig. 3 that our results agree with the electric-field effect in NMR of the Al nuclei in Al_2O_3. The measured splittings give

$$\partial Q_{Al}'/\partial E_z = 7.8 \text{ Hz/(kV/cm)}.$$

Using Eq. (9) and the quadrupole moments $Q_{Al} = 0.149b$ and $Q_{Cr} = -0.058b$ (Ref. 14), we can employ our values of $\partial Q_{Al}'/\partial E_z$ and $\partial Q_{Cr}'/\partial E_z$ to find the changes in the electric field gradients at the ^{27}Al and ^{53}Cr nuclei. These changes are

$$\partial q_{Cr}/\partial E_z = -2.6 \cdot 10^9 \text{ cm}^{-1}, \quad \partial q_{Al}/\partial E_z = -2.9 \cdot 10^9 \text{ cm}^{-1}.$$

The relative changes in the electric-field gradients at both nuclei are the same and, in particular, in a field of $E = 600$ kV/cm they amount to

$$\delta q_{Cr}/q_{Cr} \approx \delta q_{Al}/q_{Al} \approx 0.026.$$

The equality of the relative changes in the electric-field gradients may indicate that the mechanisms responsible for the electric-field-induced changes in these gradients at the ^{27}Al and ^{53}Cr nuclei are the same. Flyagin *et al.*[15] showed that in the case of ^{27}Al in Al_2O_3 the changes in the gradient induced by an electric field should be determined allowing for the overlap of the ion shifts. In the case of chromium the changes in the gradient are governed by the lattice contribution and by the contribution of electrons from the partly filled $3d$ shell. However, in the case of Cr^{3+} in Al_2O_3, the latter contribution is small. This follows from the fact that the charge of an electron shell of an ion has a highly symmetric distribution because, in the first approximation, the orbital momentum of Cr^{3+} in the ground state is zero and the admixture of excited states is small, as indicated by the similarity of the g factors of the ion and free electron. Consequently, we may assume that the ion shift mechanism plays also a decisive role in the changes of the gradients at the chromium nuclei.

It follows that our method for generating high electric fields used in Ref. 4 and in the present study makes it possible to investigate all three types of nuclei responsible for ENDOR in crystals: nuclei of paramagnetic ions, and ligand nuclei, and nuclei located relatively far from paramagnetic centers.

An investigation of the electric-field effect in ENDOR of impurity ions, together with the additional information on the hyperfine interaction makes it possible to realize the E-ENDOR method whose essence is that—in contrast to the conventional ENDOR techniques—quantum transitions in the nuclear spin system are induced by an alternating electric field.[1]

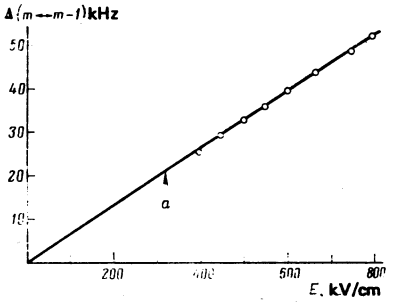

FIG. 3. Dependence of the splitting of a distant-ENDOR line on the electric field ($m \longleftrightarrow m-1 = -3/2 \longleftrightarrow -5/2$). The part of the dependence below the point a corresponds to the results of Dixon and Bloembergen.[10]

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Emission of γ rays by channeled particles

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A theory is developed for the emission of γ rays by electrons and positrons in planar channeling in the case when the recoil on radiation and the interaction of the particle spin with the radiation field become important. Analytic expressions are obtained for the spectral and angular densities of the probability of radiation for two models of the planar potential. It is shown that recoil on radiation is the cause of buildup of transverse oscillations of particles in the channel.

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INTRODUCTION

A relativistic particle channeled in a crystal moves on the average in a straight line along planes or strings of atoms of the crystal (for example, see the review by Gemmell[1]). The motion of a particle in the transverse direction is finite and consequently the particle energy associated with the transverse motion takes on discrete values. Thus, a channeled particle is a model of a one-dimensional or two-dimensional (for channeling along a string) atom uniformly moving in a crystal with a relativistic velocity. From this point of view, electromagnetic radiation occurs in spontaneous transition of the particle from the initial state of the transverse motion to the final state. This phenomenon has been discussed by Vorobiev *et al.*[2] However, the energy of the observed photon, generally speaking, does not coincide with the difference in the energy levels of the transverse motion. As a result of the Doppler effect, the photon energy depends on the angle θ between the direction of the particle's longitudinal velocity and the direction of observation. For this reason, as has been shown by Kumakhov,[3,4] the maximum of the spectral density of radiation by relativistic particles is shifted toward the x-ray frequency region and increases with increasing particle energy—in contradiction to the conclusions of Ref. 2. A detailed analysis of the errors contained in Ref. 2 has been given Kumakhov.[5]

The radiation arising in spontaneous transitions between the levels of the transverse motion can be called the "characteristic" radiation of channeled particles. As noted by Kumakhov,[4] and also as studied in more detail by Bazylev and the present author,[6] the spectrum of this radiation is determined to a significant degree by the form of the interplanar potential.

Another type of radiation (analogous to radiation in the radiative recombination of ions) arises in capture of a particle from the energy continuum to a level of the transverse motion.[7] A similar question has been discussed also by Fedorov and Smirnov[8] in a discussion of radiation by an electron diffracted in a single crystal.

After Kumakhov's work,[3] a number of articles by other authors appeared in which the theory of electromagnetic radiation in channeling was discussed. Baryshevskiĭ and Dubovskaya[9] discuss the general formulation of the problem of radiation by channeled electrons in a single crystal of finite thickness and the possibility of complex and anomalous Doppler effects. A. A. Vorob'ev and his colleagues[10] and also Terhune and Pantell[11] estimate the spectral distribution of the probability of radiation of relatively soft photons by electrons in axial channeling on the basis of the well known results of the theory of synchrotron radiation (for example, see Ref. 12). Akhiezer *et al.*[13] generalized to the case of an arbitrary potential the results of the classical calculation by Kumakhov[3] of the intensity of dipole radiation in planar channeling in a parabolic potential.

Bazylev and Zhevago[6] carried out a quantum-mechanical calculation of the spectral and angular distributions of the probability of radiation of relatively soft photons ($\hbar\omega \ll E$) in planar channeling for an arbitrary form of interplanar potential with inclusions of the effect of frequency and spatial dispersion of the electromagnetic

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