Optical detection of double electron-nuclear resonance in isolated nuclei of $Al_2O_3:Cr^{3+}$ (0.02%)

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Double electron-nuclear resonance (DENR) in isolated Al^{27} nuclei is studied by using an optical detection method in the B_1 and B_2 absorption lines of Cr^{3+} ions of Al_2O_3. This was achieved by pumping different transitions between the Zeeman sublevels of the ground state of an ensemble of Cr^{3+} ions that are bound by a weak exchange interaction $J \sim 10^{-3}$ cm^{-1}.

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In studying the mechanisms of the dynamic polarization of nuclei (DPN) in a crystal produced as a result of pumping a system of impurity electron spins in a magnetic field under the influence of a high-frequency shf field,[1] it is necessary to control the spin state of the system, for example, by measuring the absorption coefficient of the shf field. However, a direct measurement of the absorption of pumping at a power that greatly exceeds the saturation threshold, when DPN is especially effective, is difficult by traditional EPR techniques. In this work we study the properties of the optical detection (OD) method of absorption of powerful shf pumping during simultaneous action of a radio-frequency (rf) field that saturates the nuclear Zeeman subsystem of the lattice: OD of the DENR effect (double electron-nuclear resonance) in isolated nuclei of $Al_2O_3:Cr^{3+}$.[2]

We investigated the dependence of magnetic circular dichroism (MCD) on the constant magnetic field $\mathbf{H}_0$ (0 –7 kG) as a result of simultaneous action of the shf field $\mathbf{H}_1$ and the rf field $\mathbf{H}_2 \sim 0.2$ G in the range 0.2 –5 MHz at $T = 1.8$ K in the absorption lines B_1 ($\lambda = 4762$ Å) and B_2 ($\lambda = 4745$ Å) of Cr^{3+} ions in Al_2O_3. The sample of $Al_2O_3:Cr^{3+}$ ($\sim 0.02\%$) with dimensions $4 \times 4 \times 4$ mm^3 was placed inside the shf cavity of type TE_{011}, $Q \sim 2000$, for an X-band ($\nu_e = 8.90$ GHz) with openings for the passage of a sensing light beam ($\Delta\lambda \sim 1$ Å) along the axis of the crystal $\mathbf{c} \| \mathbf{H}_0$. The orientation error of the axis $\mathbf{c}$ was $< \mathbf{c}$, $\mathbf{H}_0 < 2°$. The fields $\mathbf{H}_0$, $\mathbf{H}_1$, and $\mathbf{H}_2$ were mutually orthogonal. In the absence of an shf field the value of MCD $\Delta\kappa(H_0)$ is determined by Boltzmann population of the Zeeman sublevels of the ground state $^4A_2$ ($S = 3/2$) of the Cr^{3+} ion and by circular polarization of the optical transitions from the ground-state sublevels to the excited states, which are different for the B_1 and B_2 lines.[3]

The shf power $P_e \leqslant 100$ mW distorts the monotonic dependence $\Delta\kappa(H_0) \sim H_0$ (Figs. 1b and 1c), in which the observed extrema correspond to the resonance values of

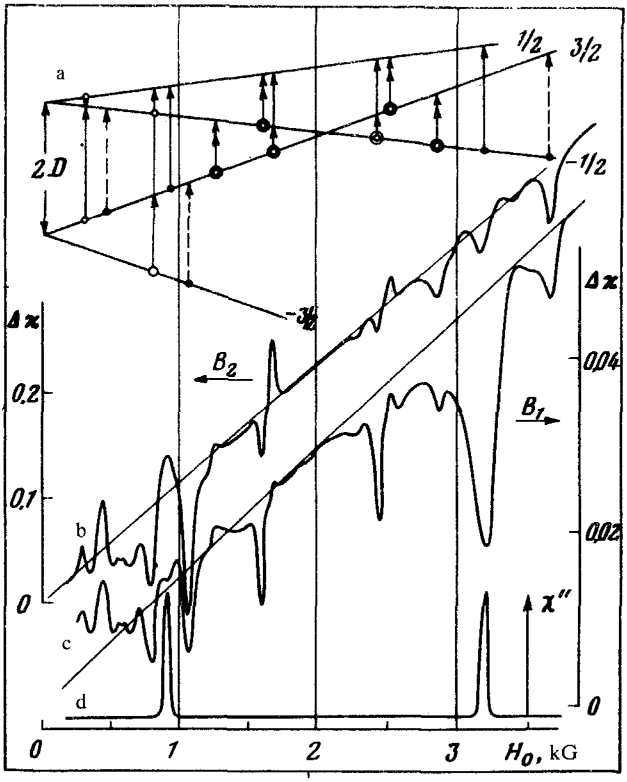

FIG. 1.

the H_0 field at which transitions between the Zeeman sublevels of the ground-state of the Cr^{3+} ion (OD EPR spectrum) occur. For comparison, Fig. 1d shows the EPR absorption spectrum χ'' for $P_e \sim 1\ \mu$W. Figure 1a identifies the OD EPR spectrum. In addition to the lines for the allowed transitions in the EPR spectra, the OD EPR spectrum also has forbidden transitions $\Delta M = \pm 2$ and 3. A distinguishing feature of the OD EPR spectrum is the presence of "combination" transitions with an accuracy ~ 0.001 cm^{-1} within the limits of experimental error, which satisfy the relation $h\nu_e = |h\nu_i \pm h\nu_j|$, where $h\nu_i$ and $h\nu_j$ are the energy separations between any two sublevels of the isolated Cr^{3+} ion a magnetic field, in particular $h\nu_e = 2h\nu_i$.[1)] It should be noted that the intensity of the OD EPR spectrum is determined by the absolute number of shf quanta absorbed during resonance. Therefore, the use of shf power $P_e \approx 100$ mW, which exceeds by ~ 50 dB, the saturation threshold for the allowed transitions $-\frac{1}{2} \to \frac{1}{2}$ made it possible to clearly detect the forbidden transitions (exceeding the saturation threshold by 20–30 dB) and combination transitions (exceeding the threshold by ~ 10 dB) in the OD EPR spectrum. The intensity of the forbidden transitions $\Delta M = \pm 2$ can be explained by the error in orientation of the **c** axis of the crystal in the magnetic field.

Analysis of the experimental data on the position of the lines and on the probability of the combination transitions (10 dB/50 dB), which is $\sim 10^{-4}$ of the probability

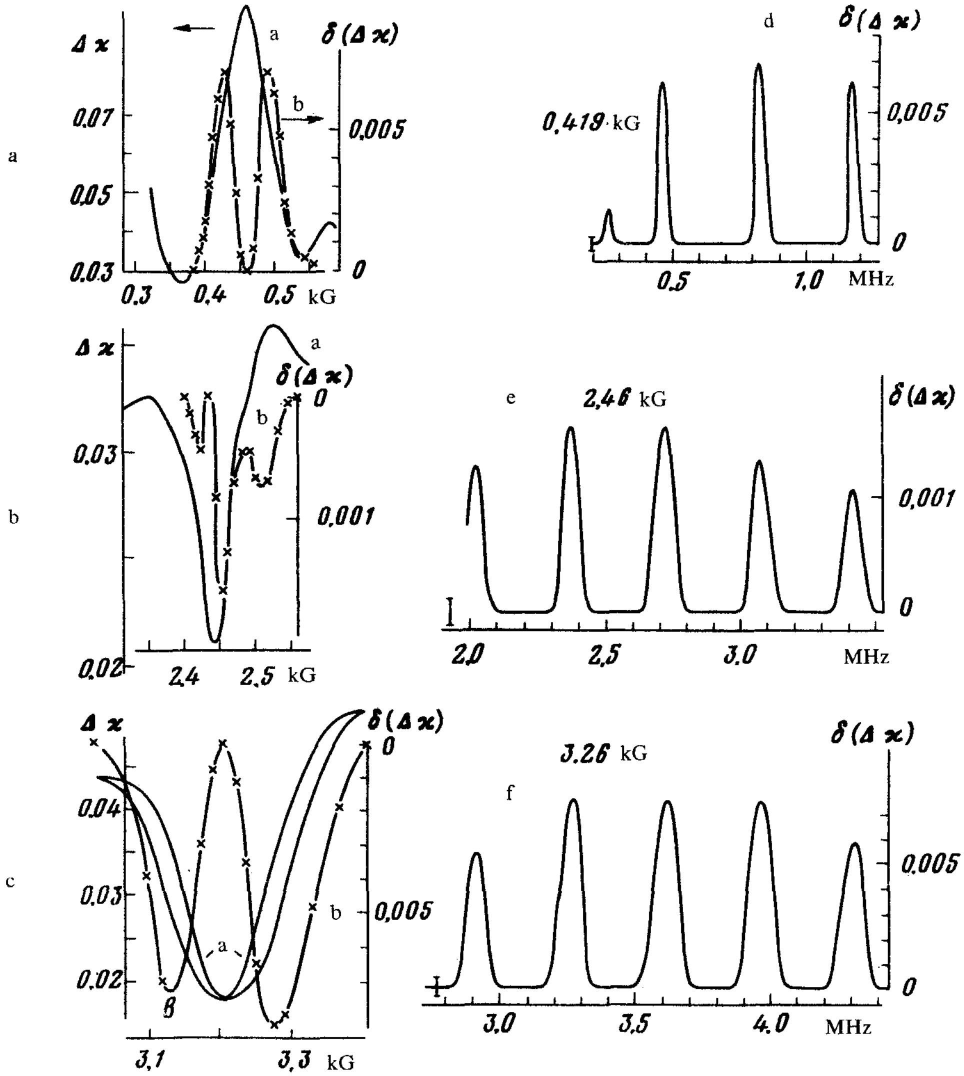

FIG. 2.

of the allowed transitions leads us to conclude that the exchange interaction[4] $J \sim 10^{-3}$ cm^{-1} between the Cr^{3+} ions is responsible for the combination spectrum. In addition, there are apparently no Cr^{3+} (n_c) ions in the Al_2O_3:Cr^{3+} (0.02%) crystal with larger values of J, for which an effective transition of shf excitation to the "isolated" Cr^{3+} ions (in the concentration n_0) is possible: $n_c(\tau_2/\tau_1)n_0 \sim 10^{-4}\, n_0$, where τ_1 and τ_2 are spin-lattice (SL) and spin-spin (SS) relaxation times of the Cr^{3+} (n_0) ions. Since the estimated value is $J \sim \Delta\nu_0$, where $\Delta\nu_0$ is the width of the nonuniformly broadened EPR line of the Cr^{3+} ion in Al_2O_3,[2] it can be assumed that pair excitations of the entire ensemble of Cr^{3+} ions[1)] are observed in the OD EPR spectrum (in contrast to the EPR spectra for isolated Cr^{3+} — Cr^{3+} pairs with $J > 0.3$ cm^{-1} studied by Statz *et al.*[5]).

It is interesting to note that the shape of the line corresponding to the transition $-\frac{1}{2}\rightarrow\frac{1}{2}$ in the OD EPR spectrum does not change (with an accuracy of a few percent) as P_e varies from 0.1 to 100 mW. This result indicates the establishment of quasi-equilibrium in the SS-interactions of the Cr^{3+} ions at a single temperature T_{ss}, which is different from the lattice temperature T_0, and of the Zeeman subsystem of the ions Cr^{3+}: T_z under pump conditions of the $-\frac{1}{2}\rightarrow\frac{1}{2}$ transition. The estimated values[1] of the SL-relaxation time (τ_1') of the SS reservoir are: $\tau_1' \sim (12/130)^{2)} \times (\tau_1 \sim 0.4$ sec)$\sim 0.34 \times 10^{-2}$ sec.

As a result of the action of the saturating rf field at frequencies of the resonance transitions between the Zeeman sublevels of Al^{27} nuclei of the Al_2O_3 lattice, we observed a variation of the shape of the lines of the OD EPR spectrum. This variation in the shape $\delta(\Delta\kappa)$ (Fig. 2b), which usually is $\sim 10\%$ of the intensity of the corresponding OD EPR lines at $P_e \approx 100$ mW (Fig. 2a), has a characteristic H_0 dependence which indicates that the DPN mechanism is valid under conditions of nonresonance pumping.[1,2] Figures 2d–2f, show spectrum for variation of MCD in the region of different OD EPR spectral lines ($P_e \simeq 100$ mW) as a function of the frequency of the rf field (OD EPR spectrum).

It is interesting to note that an increase in the power of the nonresonance shf pumping $P_e = 0.1 \rightarrow 100$ mW in the wing of the line $-\frac{1}{2}\rightarrow\frac{1}{2}$ as a result of simultaneous action of the rf field, which saturates the nuclear Zeeman subsystem of Al^{27}, causes a monotonic broadening of the line in the OD EPR spectrum (in Fig. 2c, $P_e \approx 100$ mW) and hence an increase of the OD DENR effect from $\sim 10\%$ to $\sim 27\%$ of the intensity of the $-\frac{1}{2}\rightarrow\frac{1}{2}$ line in the OD EPR spectrum.

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1) The transition $h\nu_e = 2\,h\nu_{1/2\rightarrow3/2}$ is also observed in the OD EPR spectrum at $H_0 = 5.73$ kG (not shown in Fig. 1).

2) The width of the spin packet ~ 0.4 mHz of the nonuniform EPR line[2] $\Delta\nu_0 \sim 40$ MHz is apparently attributable to fluctuations of the exchange interaction.

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