The use of thermal ballistic pulses to excite EPR signals in the upper Stark states of the impurity Er^{3+} ion in an $LiYF_4$ crystal

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Using the impurity Er^{3+} ions in an $LiYF_4$ crystal, it is possible to detect an EPR effect in the upper Stark levels which are populated with the help of thermal ballistic pulses.

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Electron paramagnetic resonance (EPR) produced due to strong spin-phonon interactions can be observed in crystals containing rare-earth ions only at helium temperatures.[1] Because of this, the resonance lines belonging solely to the ground state of a rare-earth ion can usually be observed. The EPR spectra of first excited states can sometimes be observed if the corresponding Stark levels are separated from the main level by an interval $\leqslant 15$ cm^{-1}. The situation can be changed radically if the population of the upper levels of a crystal is increased by subjecting it to the influence of thermal ballistic pulses, rather than to steady-state heating. Using this method, we were easily able to measure the EPR spectra associated with the first two excited states of Er^{3+} in an $LiYF_4$ crystal, which are separated 17 cm^{-1} and 28 cm^{-1} from the ground state. The diagram of Stark and Zeeman splittings for the lower levels of Er^{3+} in $LiYF_4$ is shown in Fig. 1.[1,2]

The experiments were performed by using a superheterodyne EPR spectrometer at a frequency $\approx$36.2 GHz and temperature $T = 4.2$ K. One end of the crystal, which had the shape of a cylinder 2.5 mm in diameter and 5 mm in height, was placed in the rf magnetic field of an H_{01}-wave cavity. The tetragonal axis of the crystal was directed along the length of the cylinder at right angles to the constant magnetic field H_0. A thin-film constantan heater with a resistance $R_H = 25$ ohms was vacuum sputtered onto the end of the crystal situated outside the cavity. The amplitude of the thermal pulses was 18 V and their duration was 200 nsec. The obtained results are shown in Figs. 2 and 3. Note that the EPR spectrum (see Fig. 3) was recorded by means of narrow-band amplification and simultaneous recording of thermal pulses at a 250-Hz pulse repetition frequency. The signals of the opposite phase on the EPR line of the second excited Er^{3+} doublet (Fig. 3) correspond to the hyperfine-structure lines of Er^{167} of the main doublet.

The time variation of the intensity of the EPR line produced by the ions at the E_1 level is shown in Fig. 2a. It evidently takes some time for a thermal pulse to affect an appreciable part of the crystal. In all probability, therefore, the EPR signal

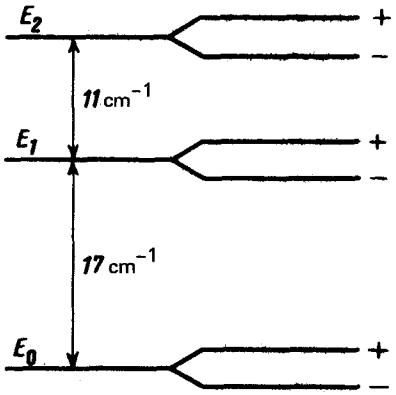

FIG. 1. The splitting scheme of the main electron energy level $4_{I_{15/2}}$ of an Er^{3+} ion in the tetragonal-symmetry field of an $LiYF_4$ crystal and in the external magnetic field (three lower Stark levels are given).

appears $t_1 \leqslant 5$ μsec after the pulse enters the crystal. The crystal face opposite the heater requires approximately the same time. The effect subsequently increases rapidly in a time $t_2 - t_1$; this is attributable to a resonance absorption of phonons whose frequency ν_1 corresponds to the $E_{0+} \rightarrow E_{1+}$ and $E_{0-} \rightarrow E_{1-}$ transitions which

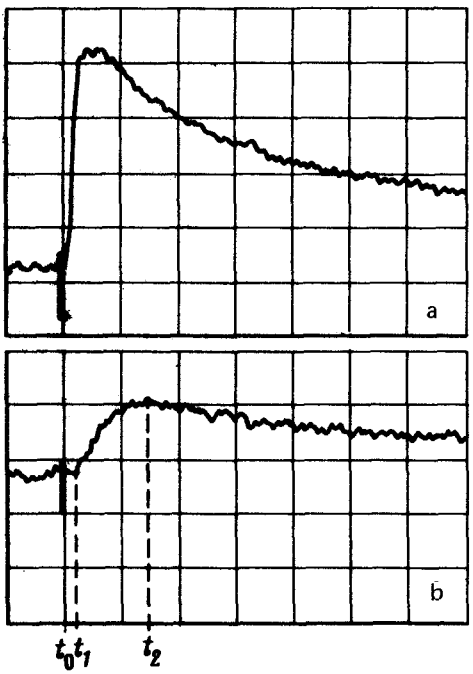

FIG. 2. Oscillograms of the time dependences of the EPR signals on (a) the first and (b) the second excited Er^{3+} doublets in $LiYF_4$ (t_0 is the time of completion of the thermal pulse, 100 μsec/division is the scale of the horizontal axis).

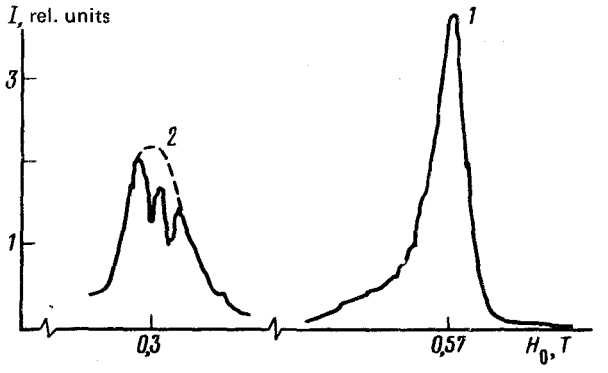

FIG. 3. Trace of the EPR spectrum of the first (1) and the second (2) excited Er^{3+} doublets in $LiYF_4$, obtained by using the thermal-pulse method.

occur without a change in orientation of the magnetic moment. Because of this, the relative population of the components of the $E_{1\pm}$ doublet is the same as that of the components of the main $E_{0\pm}$ doublet at 4.2 K. Finally, at $t > t_2$ the intensity of the effect decreases because of different relaxational processes, among which the processes of inelastic scattering of resonance phonons of frequency ν_1 presumably play the main role. The time dependence of the EPR effect induced by ions at the E_2 level is of the same nature (see Fig. 2b), but t_1 is larger by several factors, which is attributable primarily to a smaller number of phonons of frequency $\nu_2 = (E_2 - E_0)/h$ in the thermal pulse.

It seems to us that the method described here can be used successfully to detect EPR from the excited states of: 1) Van Vleck paramagnetic materials, 2) impurity rare-earth ions in metals, and 3) exchange-coupled pairs with short spin-lattice relaxation times. In magnetically ordered media containing rare-earth ions, this method can be used for studying the effects like those described in Ref. 3. Note also that an in-depth analysis of the kinetics of EPR signals induced by a thermal pulse makes it possible to determine the mechanisms responsible for the propagation of nonequilibrium, terahertz-frequency phonons in paramagnetic crystals.

[1)]The Gd^{3+} and Eu^{2+} ions in the S state are the exception.

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