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RELAXATION PROCESSES AND DISCRETE SATURATION EFFECTS IN THE EPR SPECTRUM OF Nd^{3+} IN $Ca_5(PO_4)_3F$

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We have investigated the EPR spectra and the relaxation processes of Nd^{3+} ions in single crystals of fluorapatite (FAP) - $Ca_5(PO_4)_3F$[1]. We observed effects of discrete saturation (DS) and investigated the dynamics of this effect, which so far has really not been investigated, and which is important for the understanding of both the nature of the effect and its connection with the character of the line broadening and the relaxation processes.

The measurements were performed on single crystals with Nd^{3+} concentration from 0.02 to 0.68 at.% in the temperature region 1.7 - 12° K at a frequency 9.34 GHz.

It was observed that there are three magnetically-nonequivalent Nd^{3+} complexes in the crystal. The spectrum of each complex, in the field region up to 8 kG, is well described by an axial spin Hamiltonian with effective spin $S' = 1/2$. We determined the parameters of the Hamiltonian, namely $g_z = 6.02 \pm 0.02$, $g_x = g_y = 0.18 \pm 0.02$, and found that the angles between the axes of the complexes equal 120° in the plane perpendicular to the c axis of the crystal. The line width, as well as the spectrum, is strongly anisotropic: it is minimal in z-orientation (∿ 60 G at a concentration 0.68% Nd) and increases appreciably when the perpendicular orientation is approached.

Investigation of the relaxations by the pulsed saturation method revealed that the restoration of the populations n after the saturation of the line is not singly-exponential, and is described in most cases satisfactorily by a sum of two exponentials $\Delta n - \Delta n_{eq} = A \exp(-t/\tau_1) + B \exp(-t/\tau_2)$. When the saturating pulse is shortened ($\tau \leq 10$ msec) the weight of the slow exponential with the characteristic time τ_2 decreases strongly, thus indicating a noticeable role of the cross relaxation processes [3]. We assume that this exponential corresponds to spin-lattice relaxation, whereas the faster one is connected with the cross relaxation processes inside the inhomogeneously broadened line.

Measurements of the temperature dependences of the relaxation rates in a field H = 7 kG, for samples with different Nd^{3+} concentrations, have shown (see Fig. 1) that in the region 1.7 - 4° K we have $\tau_2^{-1} \sim T^2$, and that at higher temperatures (up to 12° K) $\tau_2^{-1} \sim T^9$. The effectiveness of the Raman processes ($\tau_2^{-1} \sim T^9$) in the absence of relaxation via intermediate levels [4] indicates that the excited levels of Nd^{3+} in the FAP are located far away. Indeed, from the optical data [1] it follows that the first excited Nd^{3+} level is separated from the ground level by 400 cm^{-1}. What is still unclear in the nature of the $\tau_2^{-1} \sim T^2$ dependence observed in the region 1.7 - 4° K. Such a dependence is usually connected with the phonon

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[1] Optical spectra and laser generation were recently reported [1, 2] for this new crystal.

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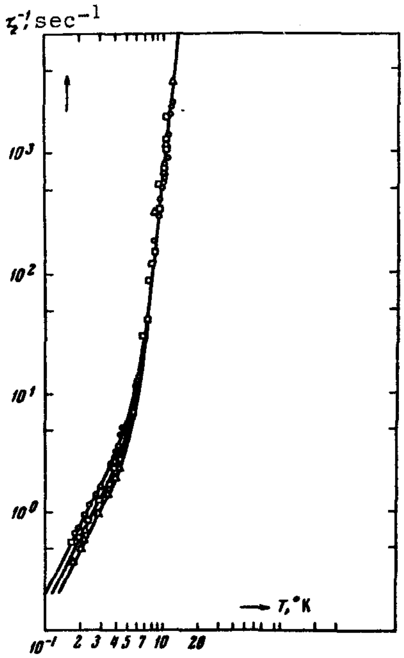

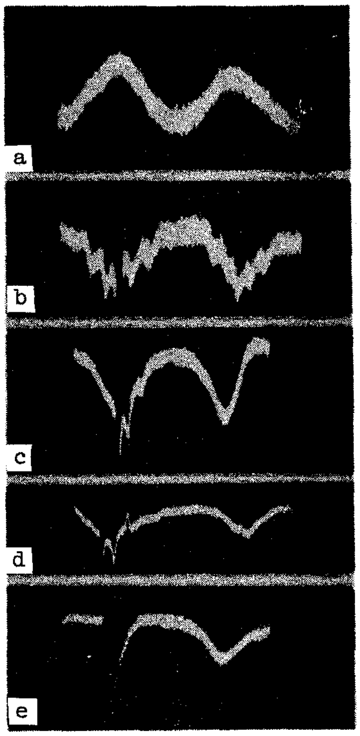

Fig. 1. Temperature dependences of the rates of spin-lattice relaxation of Nd^{3+} in FAP: o - 0.68% $\tau_2^{-1} = 0{,}2\,T^2 + 5{,}2 \cdot 10^{-7} T^9$; □ - 0,4%, $\tau_2^{-1} = 0{,}155\,T^2 + 5{,}2 \cdot 10^{-7} T^9$; Δ - 0,14%, $\tau_2^{-1} = 0{,}125\,T^2 + 5{,}2 \cdot 10^{-7} T^9$; $\nu = 9{,}34$ GHz, $H = 7$ kOe

Fig. 2. Oscillograms of DS in samples with different Nd^{3+} concentration (C in at. %). Oscilloscope sweep is linear, duration 20 msec. Sinusoidal magnetic-field modulation. a) Part of peak of Nd^{3+} EPR line in the absence of a saturating pulse, b - e) the same part of the line under pulsed saturation τ = 50 - 100 sec). b) C = 0.02%, c) C = 0.14%, d) C = 0.4%, e) C = 0.68%, no DS is observed.

heating effect, but the relatively low Nd^{3+} concentration in the investigated samples and the nature of the concentration dependence make such an assumption highly improbable.

To explain the nature of the cross relaxation, we have investigated the character of the line broadening with the aid of the "hole burning" procedure. Part of the EPR line was saturated by a microwave pulse with simultaneous magnetic sweeping at a frequency 50 Hz. The "hole" observed in this case is evidence of inhomogeneous line broadening, which is obviously connected with the scatter of the components or of the axes of the local g-tensors, due to the inhomogeneity of the crystal.

A more detailed investigation of the "hole burning" effect has shown that under certain conditions the latter has a characteristic structure. At saturation-pulse durations ranging from several tens to hundreds of microseconds in samples with low Nd^{3+} concentration, five symmetrically located "holes" appear on the contour of the burned "hole." The distance between the extreme "holes" is ∿ 45 G, that between the middle ones is ∿ 20 G (in a field H ≃ 7 kG) (see Fig. 2b). In weak fields of about 2 kG, the structure of the "hole" disappears. Similar effects of burning of several "holes" in an inhomogeneously broadened line were previously observed by a number of authors [5 - 8] and are connected with the resolution of the super-hyperfine structure of the lines following pulse saturation of "forbidden" transitions. Following [8], we shall call them discrete saturation (DS) effects.

We have observed important features of the DS effect. It depends strongly on the concentration of the paramagnetic ions: the effect was observed only in samples with small Nd^{3+} concentration (≤ 0.4%). This is explained by the swelling of the DS picture as the result of the cross relaxation inside the line, which becomes more effective with increasing concentration. Figures 2b - 2d illustrate the smearing of the DS picture with increasing Nd^{3+} concentration.

Another feature is that when the saturating pulse is shifted along the line contour, it is followed by the entire system of "holes" as a unit. This indicates a twofold nature of the EPR line broadening in FAP: as the result of the local inhomogeneity of the crystal field and as a result of the unresolved super-hyperfine structure due to the interaction of the Nd^{3+} with the nearest nuclei (F^{19} or P^{31}).

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ABSORPTION OF GAMMA QUANTA BY MANGANESE NUCLEI IN THE GIANT-RESONANCE REGION

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We measured, by the absorption method, the total nuclear cross section for the absorption of gamma quanta by the nuclei of manganese Mn^{55} in the energy interval 10 - 30 MeV. The measurements were performed with the bremsstrahlung gamma radiation spectrum ($E_{\gamma max}$ = 260 MeV) using the synchrotron of our institute. The photons were detected with a nine-channel magnetic paired γ spectrometer with resolution $\lesssim$ 1%. The absorber was made of an alloy of manganese with copper (97% Mn - 3% Cu).

The nuclear photoabsorption cross section was obtained by subtracting the normalized calculated atomic-absorption curve of [1] from the experimental atom-plus-nucleus cross section curve. The normalization was by shifting the theoretical cross section curve of [1] upward by 0.8% (18 mb at E = 9 MeV), making the average cross section $\sigma(\gamma, \text{Tot})$ below the threshold of the (γ, n) reaction (E_{thr} = 10.15 MeV) equal to zero.

The cross section of the nuclear photoabsorption $\sigma(\gamma, \text{Tot})$ for Mn^{55} is shown in the figure. The mean-square errors are indicated on the curve. The main part of the cross section (∿ 85%) lies in the interval 14 - 23 MeV. In the region of higher energies are located the well-resolved resonance at E_γ = 24.2 MeV ($\Gamma \approx$ 1 MeV, σ_0 = 90 mb-MeV) and E_γ = 27.4 MeV ($\Gamma \approx$ 1 MeV, σ_0 = 30 mb-MeV). The main broad giant-resonance peak is split into two maxima with an average energy 16.0 ± 0.3 MeV and 20.2 ± 0.3 MeV. For comparison, the figure shows also the cross sections of the reaction (γ, n) from [2, 3] and the reaction (γ, p) from [4].