Light superscattering by the spin system of paramagnetic ions

B. I. Kochelaev, Yu. G. Nazarov, and A. Kh. Khasanov
V. I. Ul'yanov-Lenin Kazan' State University

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An intense Raman scattering by paramagnetic ions has been discovered. This scattering is caused by coherent spin excitations.

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It has been shown previously[1] that a spin Raman effect can be observed for paramagnetic ions. Although the effect has been found to be weak, it can be intensified by several orders of magnitude by imposing a resonant microwave field to arrange coherence in the motion of the spins. In the case of complete coherence the scattering centers become equivalent[2] to N radiators radiating in phase with a resultant radiation

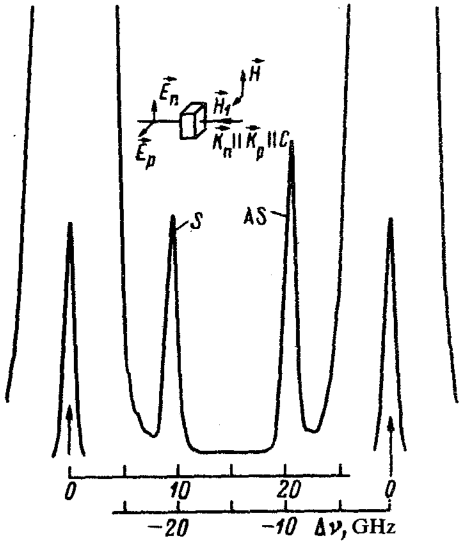

FIG. 1. Observation arrangement and spectrogram of the coherent spin Raman scattering in a CMN crystal.

intensity proportional to N^2 along the wave synchronization direction. In the present experiments we have observed this superscattering of light for Ce^{3+} ions in $Ce_2Mg_3(NO_3)_{12}{\cdot}24H_zO$ (CMN) crystals and $La_2Mg_3(NO_3)_{12}{\cdot}24H_2O{:}Ce^{3+}$ (5 at.%) crystals (LMN:Ce) at 1.7 K under ESR-saturation conditions.

In the experiments we studied the light scattering at the angle $\theta = 0°$, which corresponds to the synchronization condition for light waves which are interacting with localized coherent spin excitations. The crystalline sample was placed in an optical cryostat near a microwave resonator with a loaded quality factor $Q = 65$. Figure 1 shows the directions of the external magnetic fields—the static field **H** and the microwave field $\mathbf{H}_1$—and the directions and polarizations of the incident and scattered light in the sample. The ESR of the Ce^{3+} ions was saturated at the frequency $\nu_0 = 9.42$ GHz by a microwave power of 400 mW. The exciting light, from a He-Cd laser (442 nm, 20 mW), polarized either parallel or perpendicular to **H**, propagated strictly along the C optic axis of the crystal. The scattered light passed through an analyzer (in the position corresponding to total extinction of the light in the absence of the field H), a three-pass Fabry-Perot interferometer with a contrast of 5×10^5, and an interference filter to an FÉU-79 photomultiplier connected through an amplifying electrometer to a chart recorder.

The spectrum of the scattered light in the CMN (Fig. 1) found during continuous saturation of the ESR of the Ce^{3+} ions in the field H, held near the maximum of the absorption line, contains an intense doublet. The components of this doublet are shifted by an amount of exactly ν_0 from the frequency of the laser beam. According to our estimates, the cross section for the coherent effect per Ce^{3+} ion exceeds the corresponding value for the 90° spontaneous spin Raman scattering in the same crystal[1] by a factor $\sim 10^5$. We see from Fig. 1 that there is a difference in the intensities of the

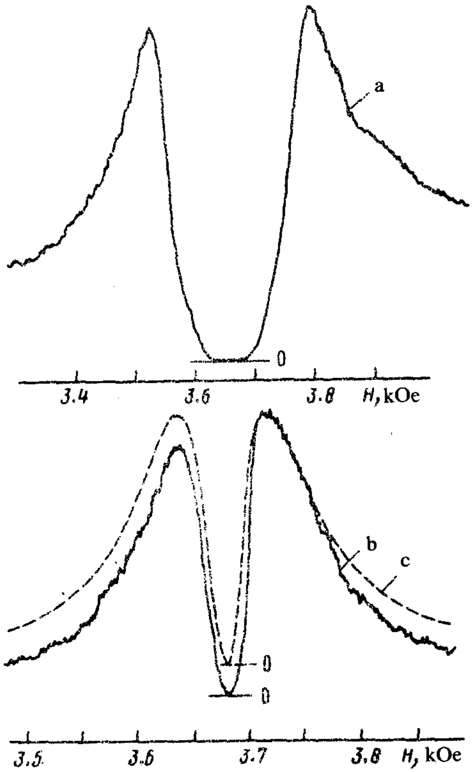

FIG. 2. Intensity of the scattered light vs the magnetic field strength. a—In CMN; b—in LMN:Ce; c—approximating function (for clarity, this function is shifted upward by the distance 0-0 from curve b).

Stokes (S) and anti-Stokes (AS) components of the doublet. This difference stems from a competition between the optical dispersion and the birefringence induced by the field H (the Cotton-Mouton effect); these effects influence the coherence length of the incident and scattered light waves in the sample in different ways.[2] When the electric vectors E_n and E_p are rotated 90°, the intensity ratio of the Stokes and anti-Stokes components is reversed. A similar spectrum was observed in the LMN:Ce crystal.

Figure 2 shows the field dependence of the effect found by recording the integrated intensity of one of the components of the Raman-scattering doublet as the field H was slowly tuned through the resonant value $H_0 = 3.68$ kG, which corresponds to a g-factor $g_\perp = 1.84$ of the Ce^{3+} ions. It is a simple matter to explain the qualitative shape of the field dependence on the basis of phenomenological considerations. The intensity of the coherent spin Raman scattering should be proportional to the square of the transverse magnetization, which is determined, under these strong-saturation conditions, primarily by the component which precesses in phase with the rotating component of the field H_1. These experimental results are thus proportional to the square of the paramagnetic dispersion of the saturated ESR lines of the Ce^{3+} ions in two samples. The asymmetry of the lines in Fig. 2 in the CMN case is due to the Cotton-

Mouton effect (whose contribution to the observed spectrum can be taken into account by detecting the intensity of the spectrally unshifted line as a function of H), while in the case of LMN:Ce it is apparently due to the asymmetry of the unsaturated ESR line of the Ce^{3+} ions, which has an approximately Lorentzian shape.

The spectrum of the optically detected ESR in the LMN:Ce crystal can be described satisfactorily by the relation found from the Provotorov equations (we are assuming a Lorentzian line shape) in the strong-saturation approximation:

$$\chi'^2 \sim \left[\frac{H - H_0}{1 + T_{1Z}^{-1} D^{-2} T_D (H - H_0)^2} \right]^2, \tag{1}$$

where D is the local field, and T_{1_Z} and T_D are the spin-lattice relaxation times of the Zeeman and dipole reservoirs, respectively. By fitting an approximating function of the type in (1) to the coordinates of the maxima of the experimental curve, using the known value $T_{1_Z} = 16$ ms, we find $D^2/T_D = 10^5$ G^2/s. The corresponding function is shown in Fig. 2c.

It can be seen from Figs. 2a and 2b that the shapes of the ESR lines in CMN and LMN:Ce are quite different (particularly in the region between the maxima). The difference is apparently due to the phonon-bottleneck effect, which plays an important role in the spin-lattice relaxation of Ce^{3+} ions in CMN (Ref. 3). The ESR spectrum in this crystal can be analyzed quantitatively with allowance for this factor by working from the coupled kinetic equations[3] for the Zeeman, dipole, and phonon subsystems.

In addition to studying the ESR spectra it would be interesting to use coherent spin Raman scattering to study the transient effects which occur in spin systems under the influence of a pulsed microwave field.

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