Spin Raman scattering of light by Ce^{3+} ions in a cerium-magnesium nitrate crystal

S. A. Al'tshuler, Yu. G. Nazarov, and A. Kh. Khasanov
V. I. Ulyanov-Lenin State University, Kazan

(Submitted 10 April 1981)

Pis'ma Zh. Eksp. Teor. Fiz. **33**, No. 10, 525–528 (20 May 1981)

Spin Raman scattering of light in a paramagnetic ionic crystal was observed for the first time.

PACS numbers: 78.30.Gt

It is known that inelastic scattering of light due to spin-phonon interaction can occur in magnetic materials. Until now, spin Raman scattering of light (RSL) has been studied only in magnetically ordered materials[1] and in semiconductors.[2] We have detected spin RSL by Ce^{3+} ions in a paramagnetic $Ce_2Mg_3(NO_3)_{12} \cdot 24\,H_2O$

 0021-3640/81/10508-04$00.60

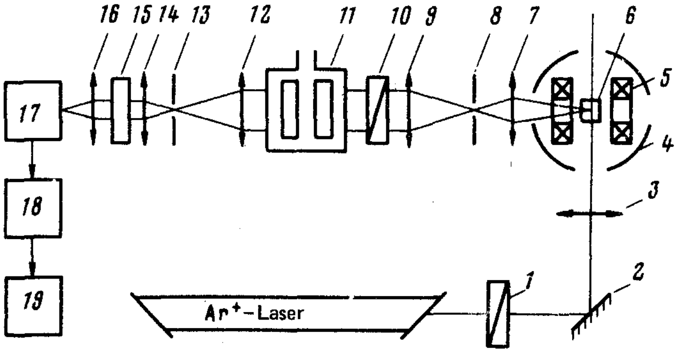

FIG. 1. Block diagram of the experimental setup. 1—Rotator of the polarization plane, 2—mirror, 3,7,9,12,14,16—objective lenses, 4—optical cryostat, 5—superconducting Helmholtz coils, 6—sample, 8 and 13—diaphragms, 10—polarizer, 11—Fabry-Perot interferometer enclosed in a pressure chamber, 15—interference filter, 17—FÉU-79 photo-multiplier, 18—electrometric amplifier, 19—recorder.

(CeMgN).

We have investigated a 90° scattering of light in CeMgN at 1.7 K in a constant magnetic field. The experimental arrangement is illustrated in Fig. 1. A single-frequency laser beam with a 488-nm wavelength and 0.3-W power, scattered by a crystal, was analyzed by a pressure-scanned Fabry-Perot interferometer. An interference filter with a 0.1-nm pass band cut off the spectrum of the interfering vibrational Raman scattering. The time constant of the recorder was 4 sec.

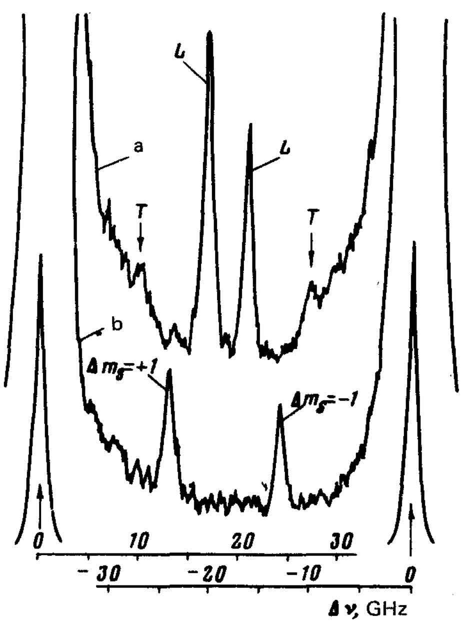

FIG. 2. (a) Spectrograms of Mandel'stam-Brillouin scattering and (b) spin Raman scattering of light in CeMgN (orientation 4, H = 5.13 Oe).

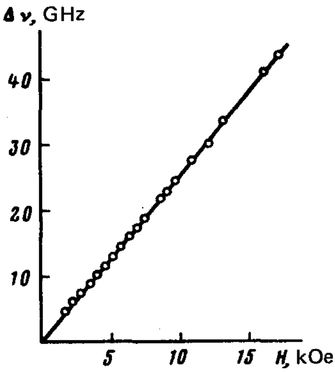

FIG. 3. Dependence of the spectral displacement of spin RSL lines on the magnetic field.

A sample oriented compatibly with the crystal-physics apparatus was used for the measurements.[3] Polarized-scattering spectra were obtained for four orientations of the sample relative to the wave vectors $\mathbf{k}_p$ and $\mathbf{k}_s$ of the incident and scattered light and relative to the direction of the external magnetic field H, which was always perpendicular to the optical axis of the crystal. The spectrograms obtained at $H=0$ contained only the components of Mandelstam-Brillouin scattering (MBS) that was studied previously.[3] The doublet lines, whose shifts $\Delta\nu$ relative to the laser-beam frequency coincided with the transition frequency ν_0 between the Zeeman levels of the effective spin $S=1/2$ of Ce^{3+} ions in the electronic ground state: $\Delta\nu=\nu_0 = g_\perp \beta H/h$, were observed in the presence of a field H. Different kinds of scattering spectra, which were obtained by polarizing the incident light at 90° angle are shown in Fig. 2. The difference in intensities of the Stokes and anti-Stokes components in the MBS is determined by the temperature of the crystal lattice and in the RSL spectrum it is determined by the population of the spin energy levels. It follows from Fig. 2 that the RSL transitions, in which the magnetic quantum number $\Delta m_s = 1$ varies, predominate over the transitions with $\Delta m_s = -1$, in approximate agreement with the equilibrium temperature of the crystal. The shape of the observed lines of the spin doublet was determined mainly by the instrumental function of the Fabry-Perot interferometer. Note that the width of the spin levels, between which an RSL transition occurs, must contribute to it. This contribution was masked in our experiment by the instrumental width of the spectrometer.

Figure 3 shows the field dependence of RSL, investigated in the interval $H \approx 2$

TABLE I.

№ orientation	k_i	k_s	H	e_i	e_s
1	001	010	y	y	z
2	010	001	y	z	y
3	100	010	y	y	z
4	010	100	x	x	z

−17 kOe. The measured values of $\Delta\nu$ are in agreement with the equality $\Delta\nu = \nu_0$ (we assumed that $g_\perp \approx 1.83$ for Ce^{3+} ions) to within an accuracy of 1–2% corresponding to the experimental measurement errors. The intensity of the RSL doublet, as expected, was independent of H in the experiment.

The effect was detected only in certain polarizations of the light beams $\mathbf{e}_i$ and $\mathbf{e}_s$ when one of them (chosen randomly) was polarized longitudinally and the other was polarized at right angles to the direction of H. This result is in agreement with the polarization selection rules that proceed from the theoretical model of the effect under study, which takes into account the electric, dipole, and spin-orbit interactions.

We can conclude from a comparison of the obtained spectra of phonon and spin scattering of light that the latter is related to the weak effects. The following approximate relation for the differential cross section of atomic RSL is valid at a certain distance from the optical-absorption lines[4]:

$$d\sigma/d\Omega \sim \left| \sum_i \frac{(\mathbf{e}_i \cdot \mathbf{D}_{2i})\,(\mathbf{D}_{i1} \cdot \mathbf{e}_s)}{E_i - E_1 - \hbar\omega} \right|^2, \qquad (1)$$

where $\mathbf{D}$, E, and ω are the matrix elements of the electric dipole moment, the energies of the levels, and the light frequency, respectively; the subscripts 1, 2, and i pertain to the initial, final, and intermediate states of the RSL transition. The lowest $5d$-configuration level in the Ce^{3+} ions of CeMgN, which accounts for the largest contribution to the sum (1), is separated from the main level by a distance of 3×10^4 cm^{-1},[5] i.e., it lies $\sim 10^4$ cm^{-1} above the virtual state. Such a large value of the denominator in the relation (1) qualitatively accounts for the low intensity of this effect.

The other triply charged lanthanide ions are apparently not expected to have a large effect, since their $5d$-configuration levels are located even higher. The forecast is more favorable for doubly charged ions of this group and for ions of the iron group, which have intense absorption bands in the visible region of the optical spectrum. The cross section of the spin RSL increases under any circumstances as the laser-beam frequency approaches the optical-absorption line.

In conclusion, we note that the spin RSL method allows for the Zeeman splitting of spin levels. This is particularly important in the investigation of paramagnetic materials with broad spin levels of magnetic ions.

1. A. S. Borovik-Romanov, N. M. Kreĭnes, and V. G. Zhotikov, A book entitled "Problemy magnitnogo rezonansa (Magnetic Resonance), Nauka, Moscow, 1978.
2. J. F. Scott, Rep. Progr. Phys. **43**, 951 (1980).
3. R. M. Valishev and A. Kh. Khasanov, Fiz. Tverd. Tela **12**, 2847 (1970) [Sov. Phys. Solid State **12**, 2299 (1970)].
4. V. B. Berestetskii, E. M. Lifshitz, and L. P. Pitaevskii, Kvantovaya elektrodinamika (Quantum Electrodynamics), Nauka, Moscow, 1980.
5. E. Tataru and J. Boskovitz, Solid State Comm. **23**, 99 (1977).

Translated by S. J. Amoretty
Edited by Robert T. Beyer