Light modulation by a magnetic resonance in a paramagnet

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Light modulation by a paramagnetic resonance ($\nu_{\mathrm{ESR}} = 36$ GHz) has been detected in the cubic paramagnetic crystal $Nd_3Ga_5O_{12}$ (NdGG).

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Light modulation by magnetic resonances in magnetically ordered crystals has now been observed in several materials: the ferromagnets $CrBr_3$ (Ref. 1) and K_2CuF_4 (Ref. 2), the antiferromagnet[3] $CoCO_3$, and the ferrimagnets YIG (Ref. 4) and $RbNiF_3$ (Ref, 5). The reasons why the effect can be observed in this variety of materials are (first) a large spontaneous moment (ferro- or antiferromagnetic) and (second) a large magnetooptic effect (a Faraday effect and/or anisotropic magnetic birefringence; see Ref. 6 for details). A similar effect has been observed in the *n*-type semiconductor CdS during excitation of a magnetic resonance of an impurity.[7]

We were interested in detecting the modulation of light by an electron spin resonance in an ordinary paramagnet.[2)] The corresponding theoretical problem was analyzed in Ref. 8.

We selected for study the cubic neodymium-gallium garnet $Nd_3Ga_5O_{12}$ (NdGG), which remains paramagnetic down to $T_N = 0.516$ K and assumes an antiferromagnetic order below this temperature.[9] Previous measurements[10] of the magnetization of this crystal at $T = 4.2$ K have shown that in fields ~ 50 kOe the magnetic moment reaches a rather large value, $4\pi M \simeq 960$ G.

The samples were grown by the Czochralski method in the Problems Laboratory of Magnetism of the Physics Department at Moscow State University.

Preliminary studies revealed a large Faraday rotation in NdGG at $T \leqslant 2$ K. We determined the Verdet constant B at this temperature: $B = 278$ deg/(cm·kOe). We also observed ESR at the frequency $\nu = 36$ GHz ($T \leqslant 2$ K). The observed resonant absorption line consists of several poorly resolved satellites and has a large width. The level of the absorption was 10–15% of the incident power in a crystal with a volume of 2×10^{-2} cm^3.

To observe the light modulation in NdGG we used an apparatus developed for studying Brillouin scattering, described in detail in Refs. 3 and 6. We studied the spectral composition of the light transmitted through the crystal during the excitation of the ESR in it (scattering at an angle $\theta \approx 0°$), As the spectral instrument we used a high-constrast three-pass scanning Fabry-Perot interferometer manufactured by the US company Burleigh. The light source was an LG–38 helium-neon laser ($\lambda = 0.628$ nm, ~20 mW). The test sample was held in an optical helium cryostat in a short-circuited waveguide at an antinode of the magnetic component of the microwave field h. The resonance was excited at the frequency $\nu = 36$ GHz. The incident light propagated along the [111] axis of the crystal, in the direction perpendicular to the static magnetic field **H**. The polarization of the incident light, **E**, was parallel to **H**, and we also arranged **H**⊥**h** (Fig. 1). The measurements were taken at a temperature $T \lesssim 2$ K.

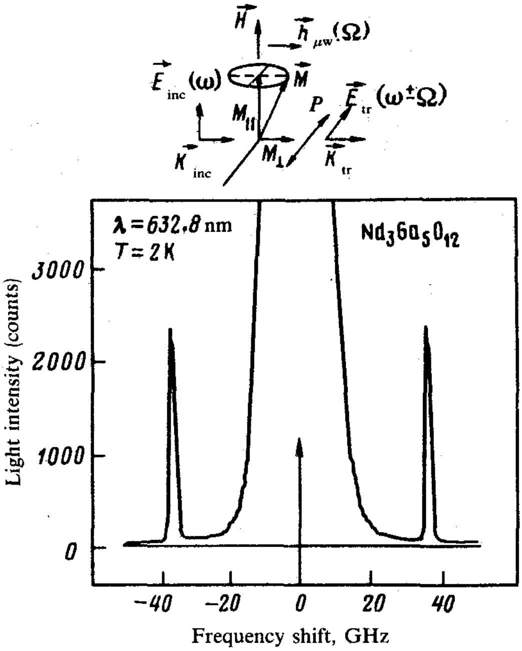

FIG. 1. Spectrum of the light transmitted through the $Nd_3Ga_5O_{12}$ sample during excitation of a paramagnetic resonance in the sample at the frequency $\nu_{\mu w} = 36$ GHz. $\lambda_{light} = 632.8$ nm, $T = 2$ K. The experimental geometry is shown at the top of the figure.

The experimental procedure can be summarized as follows: The electron spin resonance was excited in the sample, and the magnetic field was held at a certain value within the absorption line. These conditions corresponded to the maximum possible cancellation of the light passing through the optical system. The spectrum of the transmitted light was then built up and measured. Figure 1 shows an illustrative spectrum. We see that the measured spectrum has, in addition to the unshifted line, two satellites of much lower intensity, shifted by the resonance frequency ($\pm \nu_{\mathrm{ESR}}$). Their intensity amounts to $10^{-5}\%$ of that of the light incident on the crystal. The appearance of satellites during the excitation of the ESR is evidence for a modulation of the light by the paramagnetic resonance. Correspondingly, the modulation depth is $\sim 10^{-7}$. We observed modulation of the light throughout the microwave absorption line. The satellite intensity was proportional to the microwave absorption at a fixed input power. It can be seen from Fig. 1 that the intensities of the Stokes and anti-Stokes satellites in the scattering spectrum are identical.

The observations can be explained on the basis of a semiclassical picture of the resonance (Fig. 1). The total magnetization of the crystal, M, precesses in phase with the microwave field. The transverse component of the magnetization, $M_\perp$ ($M_\perp \perp H$), which is oscillating along the light propagation direction, acts through the Faraday effect to alter the intensity of the light at the frequency $2\nu_{\mathrm{ESR}}$ passing through the system. Analysis of the transmitted light with the Fabry-Perot interferometer reveals two satellites in the spectrum at frequencies $\pm \nu_{\mathrm{ESR}}$. The effect can be described analytically by means of the formula derived by Dillon for ferromagnetic $CrBr_3$ [Eq. (3) in Ref. 1]. It follows from this equation that the intensity of the satellites is proportional to the square of the Faraday rotation and thus proportional to $M_\perp^2$. More generally, the satellite intensity may depend on other magnetooptic effects.[6] In our case, however, the equality of the intensities of the Stokes and anti-Stokes components means that the other magnetooptic effects are weak (at $\lambda = 632.8$ nm). This conclusion was confirmed by independent measurements. The modulation index (or the satellite intensity) can be used to determine the angle through which the spins are deflected from their equilibrium position at the resonance. Knowing the incident microwave power we can then determine the transverse relaxation time of the system. Our estimates yield a value of 10^{-11} s for this time. The optical experiments thus yield independent data on the relaxation of the system.

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[2)] In the course of this study we learned by a private communication that similar experiments are being carried out at Kazan' State University by B. I. Kochelaev, Yu. G. Nazarov, and A. Kh. Khasanov, working with paramagnetic cerium-magnesium nitrate.

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