Observation of a new light-scattering mechanism in an antiferromagnet

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A study of the Brillouin scattering of light in the antiferromagnet EuTe has revealed a previously unobserved light-scattering mechanism that involves an *s-f* exchange interaction. This interaction causes the refractive index to depend on the magnetization of the crystal. As a result, in a highly skewed antiferromagnet, with a large static magnetization and longitudinal oscillations of the moment, an inelastic scattering of light not accompanied by a rotation of the polarization plane is observed.

How the various magnetooptic effects contribute to the intensity of Brillouin scattering (or "Mandel'shtam-Brillouin scattering") of light by spin waves in magnetically ordered materials has been discussed repeatedly in the literature.[1,2] Experiments have revealed contributions from magnetooptic effects which arise from the large spin-orbit interaction in a magnetic material and which are therefore relativistic in nature. Among these effects are the Faraday effect, magnetic birefringence, and linear and circular dichroism. In this letter we report the observation of a previously unobserved mechanism of one-magnon scattering, which is of an exchange nature. This mechanism leads to a contribution to the Brillouin scattering intensity from an isotropic magnetic refraction which describes the dependence of the refractive index on the magnetization of the crystal.[3]

The test sample is made of the cubic europium chalcogenide EuTe. Below $T_N = 9.6$ K, this crystal becomes an antiferromagnet with an easy-plane anisotropy.[4] At $T \simeq 2$ K, the penetration depth in EuTe for light at $\lambda = 632.8$ nm is $\simeq 80$ μm. According to Refs. 5 and 6 and our measurements, the Faraday effect in this material is $\sim 2 \times 10^3$ rad/(cm · kOe) ($\lambda = 632.8$ nm). The other anisotropic magnetooptic effects are weaker by at least an order of magnitude.

We studied the scattering of light by spin waves with $q = 0$ (q is the wave vector of the spin wave) excited by a microwave field (antiferromagnetic resonance). The general arrangement of the experimental apparatus is described in detail in Refs. 1 and 7. The scattered-light spectrum is studied with a Burleigh (USA) five-pass Fabry-Perot interferometer. The antiferromagnetic resonance is excited in the frequency range 35–45 GHz. The measurements were taken at $T \simeq 2$ K over the magnetic-field interval 0–65 kOe. In addition to the apparatus described in Refs. 1 and 7, we used a system that continuously records the magnetic-field dependence of the scattering intensity. Experiments were carried out in two configurations (Fig. 1).

Under resonance conditions in each configuration the scattering spectrum contains two additional satellites, which are displaced from the principal line by the frequency of the antiferromagnetic resonance, demonstrating the occurrence of an inelastic light scattering by spin waves[1] $q = 0$. We studied the dependence of the satellite

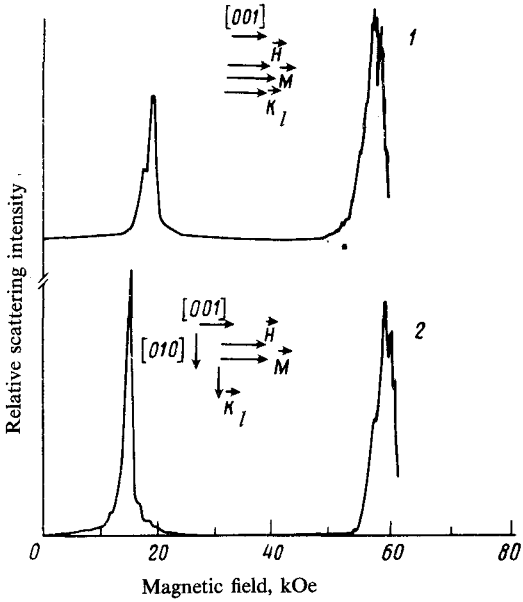

FIG. 1. Intensity of the scattered light versus the magnetic field at an antiferromagnetic resonance in EuTe for two experimental configurations: 1—$\mathbf{k}_l \| \mathbf{H} \| [100]$; 2—$\mathbf{k}_l \perp \mathbf{H}$, $\mathbf{K}_l \| [100]$, $\mathbf{H} \| [010]$. ($\Omega_{\mathrm{MW}} = 35.2$ GHz, $P_{\mathrm{MW}} = 200$ mW, $T = 1.8$ K, $\lambda_l = 632.8$ nm).

intensity on the magnetic field; figure 1 shows some representative results. The resonance is observed in both weak fields and strong fields, near the spin-flip transition.[8] Two other important experimental results follow from Fig. 1. First, the Brillouin scattering intensities are comparable at the two resonances. Second, the Brillouin scattering intensity depends only slightly on the experimental configuration.

A study of the polarization of inelastically scattered light in configuration 1 revealed that in a strong field ($H \gtrsim 40$ kOe) the polarization of the scattered light is the same as that of the light transmitted through the crystal; i.e., the scattering occurs without a rotation of the polarization plane. In a weak field ($H \approx 20$ kOe), the scattered light becomes elliptically polarized, with a 3:1 principal-axis ratio of the ellipse. The major axis makes an angle $\sim 10°$ with the polarization direction of the transmitted light.

Let us analyze the experimental data. The intensity of light scattered by spin waves in a magnetically ordered material is related to the magnetic part of the dielectric tensor $\delta\epsilon_{ik}(\mathbf{M},\mathbf{L})$ (**M** and **L** are the ferromagnetic and antiferromagnetic vectors, respectively) by[1] $I_{\mathrm{sc}} \propto E_{\mathrm{sc}}^2 \propto [\delta\epsilon_{ij}(\mathbf{M},\mathbf{L}) E_{\mathrm{inc}}^j]^2$. The tensor $\delta\epsilon_{ij}(\mathbf{M},\mathbf{L})$ can be written as an expansion in components of the vectors **M** and **L**:

$$\delta\epsilon_{ij}(\mathbf{M},\mathbf{L}) = i f_{ijk} M_k + g_{ijnk} M_n M_k + g'_{ijnk} L_n L_k + a\delta_{ij} M^2. \tag{1}$$

The first term in (1) describes the Faraday effect and the circular dichroism. The second and third terms are responsible for the linear birefringence in **M** and **L**, respectively, and also for the linear dichroism. The last term in (1) describes the isotropic increment in the refractive index. Each of these magnetooptic effects can contribute to the light scattering. Since the predominant anisotropic magnetooptic effect in EuTe is the Faraday effect, we will consider only the first and last terms in (1). Expanding (1) in small deviations of **M**, we find the electric field of the inelastically scattered wave to be

$$E^i_{sc} \propto \{ife_{ijk} m_k(t) + 2a\delta_{ij} M_0 m_z(t)\} E^j_{inc} \quad (2)$$

where e_{ijk} is the Levi-Civitas density, and f and a are the magnetooptic constants of the Faraday effect and the isotropic magnetic refraction. If we consider only the Faraday effect, we cannot describe all the experimental data available on EuTe. It can be seen from (2) that the scattering of light due to the Faraday effect should involve a 90° rotation of the polarization plane, while the scattering due to the isotropic magnetic refraction should not involve a rotation. The results of polarization experiments thus show that it is necessary to consider the isotropic magnetic refraction in the light scattering. In strong magnetic fields, this is the governing contribution, because of the quadratic dependence of this effect on the magnetization. In weak fields, the contribution from the Faraday effect is also significant; it leads to a slight eccentricity in the polarization of the scattered light. The magnitude of this eccentricity can be used to evaluate the magnetooptic constant, $a = 3\times 10^{-7}$ Oe, and thus the change in the refractive index when the crystal is magnetized to saturation, $\Delta n = 0.05$. The rotation of the polarization plane of the scattered light is apparently due to the slight dichroism.

We have calculated the intensity of Brillouin scattering at an antiferromagnetic resonance in EuTe, taking into account either the Faraday effect or the isotropic magnetic refraction, for the two experimental configurations. The results of these calculations are shown in Fig. 2. At the high field resonance, the intensity of the scattering due to the Faraday effect should be highly dependent on the experimental configuration. In contrast, the light scattering due to the isotropic magnetic refraction is isotropic and should not depend on the light-propagation direction; this is what we, in fact, observe experimentally.

It can be concluded from this analysis that the light scattering in the antiferromagnetic EuTe is due primarily to a magnetooptic effect which has not previously been taken into account: isotropic magnetic refraction. It can be seen from (2) that this contribution is effective when there are longitudinal oscillations of the magnetic moment $m_z(t)$ in the spin system and when there is a substantial magnetization M_0. Such a situation is possible only in a highly skewed antiferromagnet (in a magnetic field on the order of the exchange field). As we have already mentioned, the elementary nature of the isotropic magnetic refraction is different from that of the other magnetooptic

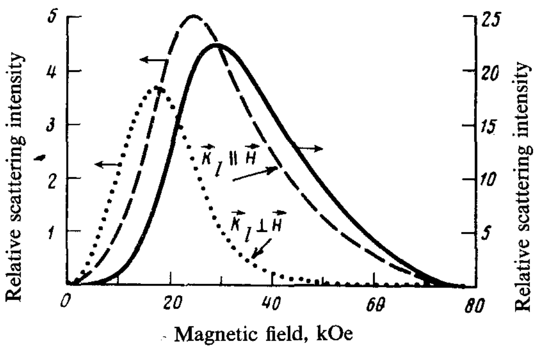

FIG. 2. Theoretical functional dependence of the scattering intensity on the magnetic field at the antiferromagnetic resonance for two magnetooptic effects: Dashed curve—Faraday effect, $\mathbf{k}_l \parallel \mathbf{H}$; dotted curve—Faraday effect, $\mathbf{k}_l \perp \mathbf{H}$; solid curve—isotropic magnetic refraction.

effects. This refraction, discovered by Wachter[3] in the europium chalcogenides EuO, EuS,and EuSe, was attributed to a pronounced shift of the optical-absorption edge upon magnetization of the crystal. This shift is due to the exchange interaction of an electron in a $5d$ excited level with electrons localized at the $4f$ level.[9,10] It is unrelated to the presence of an orbital angular momentum of the ion. In EuTe, a shift of electronically excited levels has been observed directly,[11] but only in ~70-kOe fields, where the magnetization reaches saturation. Since this scattering mechanism is of an exchange nature, its contribution to the scattering intensity should be substantial. The detection of this mechanism extends the list of objects in which magnetic scattering can be observed.

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