

For silver we have  $\omega_p \approx 1.39 \times 10^{16} \text{ sec}^{-1}$  [3]; the frequencies  $\omega_L$  and  $\omega_S$  lie in the region of the long-wave edge of quantum absorption, where the function  $\epsilon_0(\omega)$  is large. According to the data of [4],  $\epsilon_0(\omega_L) \approx \epsilon_0(\omega_S) \approx 6$ ,  $\omega_L = \omega_p/\sqrt{\epsilon_0} \approx 5.7 \times 10^{15} \text{ sec}^{-1}$ , and  $(\omega_L - \omega_S)/\omega_L \approx 7 \times 10^{-2}$ . The experimentally observed values are sufficiently close to these calculated ones.

The obtained value  $\hbar\omega_L \approx 3.96 \text{ eV}$  agrees well with the results of the measurements of the characteristic electron energy losses in silver. These data lie in the interval from 3.9 to 4.6 eV [5 - 7]. The experimental data on the transformation of the plasma oscillations into electromagnetic radiation [8] also yield the close value  $\hbar\omega_L = 3.75 \text{ eV}$ .

As shown by the experimental results, the incident wave interacts with the surface oscillations more effectively than with the Langmuir oscillations.

The observed effect of incoherent scattering of light can be used for the investigation of the spectra of elementary excitations of electrons in metals.

- [1] I.A. Akhiezer, Zh. Tekh. Fiz. 33, 935 (1963) [Sov. Phys.-Tech. Phys. 8, 699 (1963)].
- [2] N.Ya. Koparenko and A.M. Fedorchenko, *ibid.* 39, 42 (1969) [14, 27 (1969)].
- [3] V.G. Padalka and I.N. Shklyarevskii, Opt. Spektrosk. 11, 527 (1961).
- [4] V.K. Miloslavskii and R.G. Yarovaya, *ibid.* 21, 708 (1966).
- [5] J.C. Turnbull and H.E. Farnsworth, Phys. Rev. 54, 509 (1938).
- [6] E. Rudberg, Proc. Roy. Soc. A127, 111 (1930); Phys. Rev. 50, 138 (1936).
- [7] W. Klein, Optik 11, 226 (1954).
- [8] W. Steinmann, Phys. Rev. Lett. 5, 470 (1960).

## TWO-MAGNON SCATTERING OF LIGHT IN ANTIFERROMAGNETIC $\text{KMnF}_3$

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An experimental study of Raman scatterings of light in magnetically ordered substances, first considered theoretically by Bass and Kaganov in 1959 [1], has been made possible only following the development of quantum electronics and the appearance of powerful sources of monochromatic radiation, namely lasers. In spite of the fact that only four years have elapsed since the performance of the first experiment [2], the light-scattering method has found extensive use in the study of the energy spectra of magnetic crystals. It suffices to mention the observation of light scattering with excitation of two magnons of limiting energy [2, 3], the investigation of localized magnetic states in impurity antiferromagnets [4, 5], and the proof of existence of magnon-magnon interactions in magnetodielectrics [6]. Just as in the study of the phonon spectrum of a substance, the method of Raman scattering by spin waves supplements successfully the infrared and submillimeter measurements, and in many cases it is the only one possible. Thus, for example, owing to the high symmetry in antiferromagnetic crystals with perovskite structure, the two-magnon absorption is greatly attenuated and is not observed [7], and the intensity of the analogous process in Raman scattering is the same as in crystals of lower symmetry [6].

We report in this paper the results of an investigation of the scattering of light in the antiferromagnetic crystal  $\text{KMnF}_3$  ( $T_N = 88^\circ\text{K}$ ). The measurements

were made with the 4880 Å line of an argon laser (type LG-106) of 1 W power. The light scattered at right angle passed through a polarizer and was focused on the input slit of the DFS-12 spectrophotometer of dispersion 5 Å/mm. The radiation receiver was a photomultiplier cooled in a special Dewar. The registration was with a photon-counting system. The details of the experimental setup were published earlier [6].

The  $\text{KMnF}_3$  single crystals were grown by the Bridgman method [9] and oriented by x-ray diffraction; they were then cut in the form of rectangular parallelepipeds. The measurements were performed on two samples (average linear dimension approximately 4 mm) with the edges directed along [001], [010], [100], and [001], [110], [110], respectively. The temperature ranged from 20 to 300°K and was monitored with a copper-constantan thermocouple.

Two-magnon scattering (and absorption) of light in antiferromagnets, with allowance for magnon-magnon interactions, was recently considered theoretically by Elliott and Thorpe [7]. The Hamiltonian for crystals with perovskite structure ( $\text{RbMnF}_3$ ,  $\text{KMnF}_3$ ) and neglecting spin-orbit interaction is given by

$$\mathcal{H} = \sum_{\mathbf{R}, \mathbf{r}} [B_1 (\mathbf{E} \cdot \mathbf{r})(\mathbf{E}' \cdot \mathbf{r}) + (B_1 - \frac{1}{3} B_3) (\mathbf{E} \cdot \mathbf{r})(\mathbf{E}' \cdot \mathbf{r})] S_{\mathbf{R}} S_{\mathbf{R} + \mathbf{r}}.$$

Here  $\vec{E}$  and  $\vec{E}'$  are the electric vector of the incident and scattered light,  $\mathbf{r}$  a unit vector in the direction between the nearest neighbors from oppositely located sublattices, and  $B_1$  and  $B_3$  coefficients that transform in accordance with the representations  $\Gamma_1^+$  and  $\Gamma_2^+$  respectively. The concrete form of these coefficients depends on the nature of the interaction of the spin system with the light. The expected line shape of two-magnon scattering was calculated in [7] for the ion  $\text{Mn}^{2+}$  ( $S = 5/2$ ) by the Green's function method. The characteristic difference between the  $\Gamma_1^+$  mode and the  $\Gamma_3^+$  mode is that the former does not have a resonant character.

The experimentally investigated spectrum of two-magnon scattering of light in antiferromagnetic  $\text{KMnF}_3$  is shown in Fig. 1, and the temperature dependence of the position of the band maximum is shown in Fig. 2. It should be noted that the shape, spectral position, and intensity of the band are the same for the two investigated crystals of different orientation, with  $\vec{E} \parallel \vec{E}' \parallel [001]$  or  $[110]$ , and  $\vec{E} \parallel [110]$ ,  $\vec{E}' \parallel [110]$ . The intensity  $\alpha_{zx}$  of the spectrum is much weaker than  $\alpha_{zz}$ , although we did not obtain total polarization of the band, unlike the case of  $\text{RbMnF}_3$ , in which the two-magnon scattering of light was investigated for comparison. These results indicate that the scattered light contains only the  $\Gamma_3^+$  mode. The absolute intensity of the two-magnon scattering of light in  $\text{KMnF}_3$  is the same as in  $\text{RbMnF}_3$ , i.e., it amounts to approximately  $10^{12} \text{ cm}^{-1} \text{ rad}^{-1}$ .

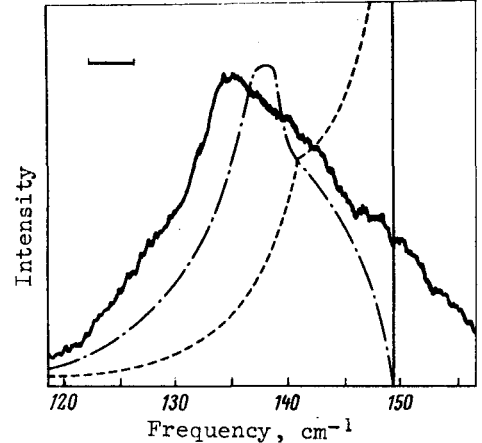


Fig. 1. Experimental (at 20°K) and theoretical two-magnon scattering in  $\text{KMnF}_3$ . Spectral width of slit 4  $\text{cm}^{-1}$ .  $\vec{E}$  and  $\vec{E}'$  parallel to [001].

Figure 1 shows the calculated plots of the expected line shape of two-magnon scattering without allowance for the interaction between the magnons (dashed curve) and with allowance for this interaction (dash-dot curve). The intensity at the maximum of the theoretical curve has been normalized to the experimental one. The good agreement between calculation and experiment is evidence in favor of the theory developed in [7] with allowance for the magnon-

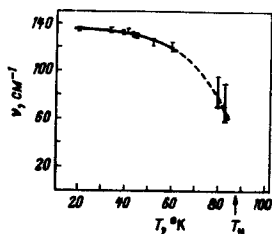


Fig. 2. Temperature dependence of the spectral position of the maximum of two-magnon scattering.

magnon interactions. The value obtained by us for the maximum magnon energy in  $\text{KMnF}_3$ ,  $75 \text{ cm}^{-1}$ , is in excellent agreement with the results of the study of the spin-wave spectrum by inelastic scattering of neutrons [10].

The earlier measurements of two-magnon scattering in antiferromagnetic crystals  $\text{RbMnF}_3$  [6] and  $\text{KNiF}_3$  [11] have led to an almost total agreement between the experimental data and calculation. It should be noted here that the cubic structure of the crystals  $\text{RbMnF}_3$  and  $\text{KNiF}_3$  does not change down to the very lowest temperatures, but  $\text{KMnF}_3$  goes through a number of phase transitions when cooled [12], namely into the orthorhombic phase at  $184^\circ\text{K}$ , and the crystal symmetry decreases to monoclinic at the region of the magnetic-ordering point ( $T_N = 88^\circ\text{K}$ ). These distortions,

which apparently have little influence on the spin-wave spectrum of  $\text{KMnF}_3$ , lead to resolution of the Raman scattering of first order by phonons, and to the appearance of a number of lines in the scattered spectrum, including the immediate vicinity of the two-magnon scattering band, leading to broadening of the latter. For the same reason, it is impossible to study the temperature dependence of the half-width of the two-magnon peak. The scattering of light by optical phonons will be considered in detail elsewhere. Here we wish to mention only the frequencies of the additional lines appearing when the crystal is cooled: 27, 49, 108, 117, 158, 170, 230, and  $252 \text{ cm}^{-1}$  at  $20^\circ\text{K}$ .

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- [1] F.G. Bass and M.I. Kaganov, Zh. Eksp. Teor. Fiz. 37, 1390 (1959) [Sov. Phys.-JEPT 10, 986 (1960)].
- [2] P.A. Fleury, S.P.S. Porto, L.E. Cheesman, and H.J. Guggenheim, Phys. Rev. Lett. 17, 84 (1966).
- [3] P.A. Fleury, S.P.S. Porto, and R. Loudon, Phys. Rev. Lett. 18, 658 (1967).
- [4] P. Moch, G. Parisot, R.E. Deitz, and H.J. Guggenheim, Phys. Rev. Lett. 21, 1596 (1968).
- [5] A. Oseroff and P.S. Pershan, Phys. Rev. Lett. 21, 1593 (1968).
- [6] P.A. Fleury, Phys. Rev. Lett. 21, 151 (1968).
- [7] R.J. Elliott and M.F. Thorpe, J. Phys. C (Solid State Physics) 2, 1630 (1969).
- [8] V.I. Kut'ko, V.I. Fomin, N.M. Nesterenko, A.I. Zvyagin, and Yu.A. Popkov, Trudy FTINT AN UkrSSR, No. 4, p. 203 (1969).
- [9] B.V. Beznosikov and N.V. Beznosikova, Kristallografiya 13, 188 (1968) [Sov. Phys.-Crystallogr. 13, 158 (1968)].
- [10] S.J. Pickart, M.F. Collins, and C.G. Windsor, J. Appl. Phys. 37, 1054 (1966).
- [11] S.R. Chinn, H.J. Zeiger, and J.R. O'Connor, Papers at 15th National Conference on Magnetism (USA, November 1969).
- [12] O. Beckman and K. Knox, Phys. Rev. 121, 376 (1961).