1972
international book year

KFKI-72-26

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Hungarian Academy of Sciences

CENTRAL RESEARCH INSTITUTE FOR PHYSICS

BUDAPEST

2017

KFKI-72-26

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Submitted to Solid State Communications

ABSTRACT

Raman scattering by four magnons in antiferromagnets is studied. It is shown that the excited-state exchange mechanism resulting in two-magnon scattering at the same time gives four-magnon scattering as well. The intensity and line shape of this scattering is computed numerically for simple cubic systems. The agreement with experimental data is fairly good for $KNiF_3$, if magnon-phonon interaction is taken into account.

KIVONAT

A négymagnonos Raman szórást vizsgáljuk antiferromágneses anyagokban. Megmutatjuk, hogy a kétmagnonos szórásért felelős kölcsönhatás ugyanakkor négymagnonos szórást is eredményez. Ennek a négymagnonos szórásnak az intenzitását és vonalalakját határozzuk meg numerikusan egyszerü köbös rendszerre. $KNiF_3$ esetén a kisérleti adatokkal való egyezés elég jó, ha a magnon-fonon kölcsönhatást is figyelembe vesszük.

РЕЗЮМЕ

Исследуется четырёхмагнонное рассеяние Рамана в антиферромагнетиках. Покажем, что взаимодействие ответственное за двухмагнонное рассеяние, вызывает одновременно и четырёхмагнонное рассеяние. Определяется интенсивность и форма линии для этого рассеяния для простой кубической системы. Принимая во внимание взаимодействие магнонов с фононами, в случае $KNiF_3$ согласие с экспериментальными данными является хорошим.

The excitation of two antiferromagnetic magnons in Raman scattering has been the subject of detailed investigations in the last few years /see e.g.[1-6]/. Starting from the electrical dipolar interaction between light quanta and the magnetic electrons, and invoking the strong exchange interaction in antiferromagnets, a simple effective interaction Hamiltonian, H_1, can be derived[7]. H_1 describes the scattering of photons and the simultaneous flip of two neighbouring spins. Each of these spin flips can generate a spin wave, the subsequent scattering of which gives rise to a typical resonance-like Raman scattering cross section.

In an earlier publication[8] it was shown that using the same interaction mechanism as above, four-magnon /4M/ creation processes are also possible. It was pointed out that in the Dyson-Maleev formulation the product S^+S^- or S^zS^z in H_1 contains terms quartic in the boson operators. Introducing the spin wave operators by a Bogoliubov transformation, these terms describe - among others - the creation of four magnons.

At the same time, working independently, Dietz et al.[9] observed a weak peak in the Raman scattering spectrum of NiO and $KNiF_3$ whose position and temperature dependence indicated that it arises from the excitation of four magnons. These investigators proposed several mechanisms to explain the scattering, including the same possibility as is proposed here, but after estimating the total intensity they concluded that this mechanism can be only partially responsible for the observed peak. They suggested that in the virtual excited state the hole propagates, thus leading to four spin deviations on neighbouring atoms /in some cases to two deviations on the same atom/. From estimates of the energies of these states, in their view the excited-state propagation effect can account for the experimental facts.

The aim of this paper is twofold. First, the line shape and intensity of Raman scattering by four magnons is investigated quantitatively, using the same interaction as that which describes the two-magnon /2M/ scattering. Second, an attempt will be made to evaluate the contribution coming from the excited-state propagation effect.

Using the same mathematical formalism as for Raman scattering by two magnons [5,8], the Green's function

$$G_{\delta\delta'}(\omega) = \frac{1}{2}\int_{-\infty}^{\infty} dt\, e^{i\omega t} \left\langle \hat{T} \left\{ \left(\sum_i S_i(t)\, S_{i+\delta}(t)\right)\left(\sum_j S_j(0)\, S_{j+\delta'}(0)\right)\right\}\right\rangle \qquad /1/$$

will be considered. The calculation will be done at temperature T=0. A few possible processes giving rise to 4M scattering are displayed schematically in Fig. 1, applying the diagrammatic representation technique proposed by Vaks et al.[10] S^+ is represented by a vertex with one outgoing line, the vertex for S^- has one incoming or two incoming and one outgoing lines, while a vertex with one incoming and one outgoing lines stands for S^z.
Considering the contribution of the processes in Fig. 1/a-d/, we get

$$\mathrm{Im}\, G_i(\omega) = \pi \frac{1}{N^2} \sum_{\substack{k_1,k_2\\k_3}} \delta\left(\omega - \varepsilon_{k_1} - \varepsilon_{k_2} - \varepsilon_{k_3} - \varepsilon_{k_1-k_2+k_3}\right) \times$$

$$\times \Big\{ \varphi_i(k_1)\, u_{k_1} u_{k_2} v_{k_3} u_{k_1-k_2+k_3} + \varphi_i(k_1)\, v_{k_1} v_{k_2} u_{k_3} v_{k_1-k_2+k_3}$$

$$-\varphi_i(k_1-k_2)\, v_{k_1} u_{k_2} u_{k_3} v_{k_1-k_2+k_3} - \varphi_i(k_2-k_3)\, v_{k_1} v_{k_2} u_{k_3} u_{k_1-k_2+k_3} \Big\}$$

$$\times \Big\{ \varphi_i(k_1-k_2+k_3)\, v_{k_1} u_{k_2} v_{k_3} v_{k_1-k_2+k_3} + \varphi_i(k_1-k_2+k_3)\, u_{k_1} v_{k_2} u_{k_3} u_{k_1-k_2+k_3}$$

$$-\varphi_i(k_1-k_2)\, v_{k_1} u_{k_2} u_{k_3} v_{k_1-k_2+k_3} - \varphi_i(k_2-k_3)\, u_{k_1} u_{k_2} v_{k_3} v_{k_1-k_2+k_3} \Big\} \qquad /2/$$

with

$$u_k^2 = \frac{\varepsilon_o + \varepsilon_k}{2\varepsilon_k} \quad \text{and} \quad v_k^2 = \frac{\varepsilon_o - \varepsilon_k}{2\varepsilon_k} \qquad /3/$$

where ε_k is the energy of a magnon with wave vector k, ε_o is the maximum of the spin wave band and φ_i is a basis function of an irreducible representation[5], depending on the structure and the polarization of the light. For scattering of Γ_3^+ type by simple cubic systems

$$\varphi_3(k) = \frac{1}{\sqrt{6}}\left(\cos k_x a - \cos k_y a\right). \qquad /4/$$

Eq. /2/ was numerically computed for a uniaxial anisotropic simple cubic system. The relative anisotropy is taken to be 0.0066, which is a realistic value for $KNiF_3$[4]. The resulting curve is plotted in Fig. 2 /solid line/. The intensity of the scattered light shows a peak at about 2.5 times the maximum single magnon energy. For an isotropic system the peak is almost at the same position, the effect of anisotropy being negligible.

The transversal contribution of the scattering /i.e. the contribution of the process represented in Fig. 1/a// is also shown in Fig. 2 /dashed line/. The line shape is similar to that of the total scattering.

The Hamiltonian H_1 yields 4M processes even if each spin flip creates only one magnon, but in an intermediate state one magnon decays into three. This is an allowed process for antiferromagnets. Such processes are shown in Fig. 1/e-h/. They give a small correction to the result obtained above, increasing the total intensity by a few per cent without changing the line shape drastically.

It is known from the calculation of 2M resonances[1] that the magnon-magnon interaction is very important in describing the proper line shape. Calculation with noninteracting magnons yields a singular Raman scattering intensity. The interaction eliminates this singularity, but far from the singularity its effect is small. Until now this interaction has been neglected for the 4M scattering. What is argued here is that this is a reasonable approximation and interaction effects seem to be negligible, as without interaction the cross section is already quite smooth. Considering the 4M scattering as coming from two pairs of magnons with momentum k and -k, the momentum of the pairs should be integrated, while for 2M scattering the momentum of the pair is zero. The singularity appearing at k=0 is eliminated by the integration.

For the ratio of the total intensity of 4M and 2M scattering we have $\sim 1/120$. The uncertainty of at least 10-20% comes from the uncertainty of the numerical calculations. This value is by an order of magnitude bigger than the ratio $\sim 10^{-3}$ estimated by Dietz et al.[9]

Experimentally for both $KNiF_3$ and NiO the observed peak occurs at about 3 times the maximum energy of magnons. The measured intensity ratio for $KNiF_3$ is close to the calculated one /experimentally it is $\sim 1/150$/. As the 4M scattering discussed above should appear in any case if 2M resonance appears, the good agreement of the intensities suggests that the considered effect is the relevant one. The position of the peak, however, needs further clarification.

For $KNiF_3$ the 4M peak lies quite close to another peak, identified as a two-phonon resonance. Actually this resonance is almost at the position where the 4M peak should appear. As Zawadowski and Ruvalds[11] have shown, if there are two optically active modes lying energetically close enough to each other for the spectra to overlap, and due to an interaction they can transform into each other, this interaction changes completely the spectrum. The implication of this idea in the present case is that by coupling the two-phonon and 4M states, the phonon-magnon interaction can change the line shape drastically. The effect is illustrated in Fig. 3 for a very simple model. The two-phonon and 4M states are described by Lorentzians and are coupled by a constant coupling. If the sign of the coupling is appropriately chosen, there is a repulsion between the peaks, suggesting that magnon-magnon interaction can shift the 4M peak from $2.5\varepsilon_0$ to $3\varepsilon_0$, as observed experimentally.

In this view, then, the excited-state exchange effect explains satisfactorily the 4M scattering in $KNiF_3$. Nevertheless, an attempt was made to investigate quantitatively the excited-state propagation effect proposed by Dietz et al.[9] The particular case chosen is that of two spin deviations on two neighbouring lattice sites /Fig. 3/n/ in Ref. 9/. In order to get the Raman scattering cross section, the Green's function

$$G_{\delta\delta'}(\omega) = \frac{1}{2} \int_{-\infty}^{\infty} dt\, e^{i\omega t} \left\langle \hat{T}\left\{\left(\sum_i S_i^+(t)\, S_i^+(t)\, S_{i+\delta}^-(t)\, S_{i+\delta}^-(t)\right) \times \right.\right.$$
$$\left.\left. \times \left(\sum_j S_j^+(0)\, S_j^+(0)\, S_{j+\delta'}^-(0)\, S_{j+\delta'}^-(0)\right)\right\}\right\rangle \qquad /3/$$

was considered. Neglecting the magnon-magnon interaction, the calculated curve is shown in Fig. 2 /dotted line/. If the interaction is taken into account, the singularity at $4\varepsilon_0$ should disappear, but on the other hand it probably cannot produce the sharp peak at $3\varepsilon_0$, since without interaction there is a rather flat minimum there. No numerical calculations were carried out for the other processes, but there is little hope that any of the other four spin deviation states would give good agreement if this particular one fails.

It is therefore possible that for NiO as well the excited-state exchange and not the excited-state propagation effect is important. The difference in the 4M scattering by $KNiF_3$ and NiO may be due to differences of structure. In NiO in the excited state, the exchange between first neighbours may be important. In Eq. /1/ S_1 and $S_{1+\delta}$ may thus belong to the otherwise non-interacting sublattices. In that case the process in Fig. 1/b/ alone gives a contribution, and the two pairs of magnons propagate independently on the two sublattices. For the basis function φ_i , the function corresponding to a fcc structure should be taken. Whether or not this yields the observed scattering for NiO requires further numerical confirmation.

Acknowledgements - I should like to express my gratitude for useful discussions and encouragement to the scientific staff of the Institut Max von Laue-Paul Langevin /Grenoble/, where the main part of this work was completed. Numerical calculations were carried out on the IBM 360-67 computer of the University of Grenoble. Enlightening discussions with Dr. A. Zawadowski were very helpful.

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FIGURE CAPTIONS

Fig. 1. Typical four-magnon processes resulting from H_1.

Fig. 2. Calculated energy dependence of the intensity of Raman scattering by four magnons coming from a/ H_1 /solid line/, b/ the transversal part of H_1 /dashed line/, c/ a four spin deviation state /dotted line/.

Fig. 3. Typical line shape coming from two modes of energy $E_1=2.4$ and $E_2=2.6$ /the unrenormalized widths are $\Gamma_1=0.01$ and $\Gamma_2=0.5$, respectively/ without /solid line/ and with /dashed line/ coupling between the modes. The coupling constant is chosen to be $g=-0.3$.

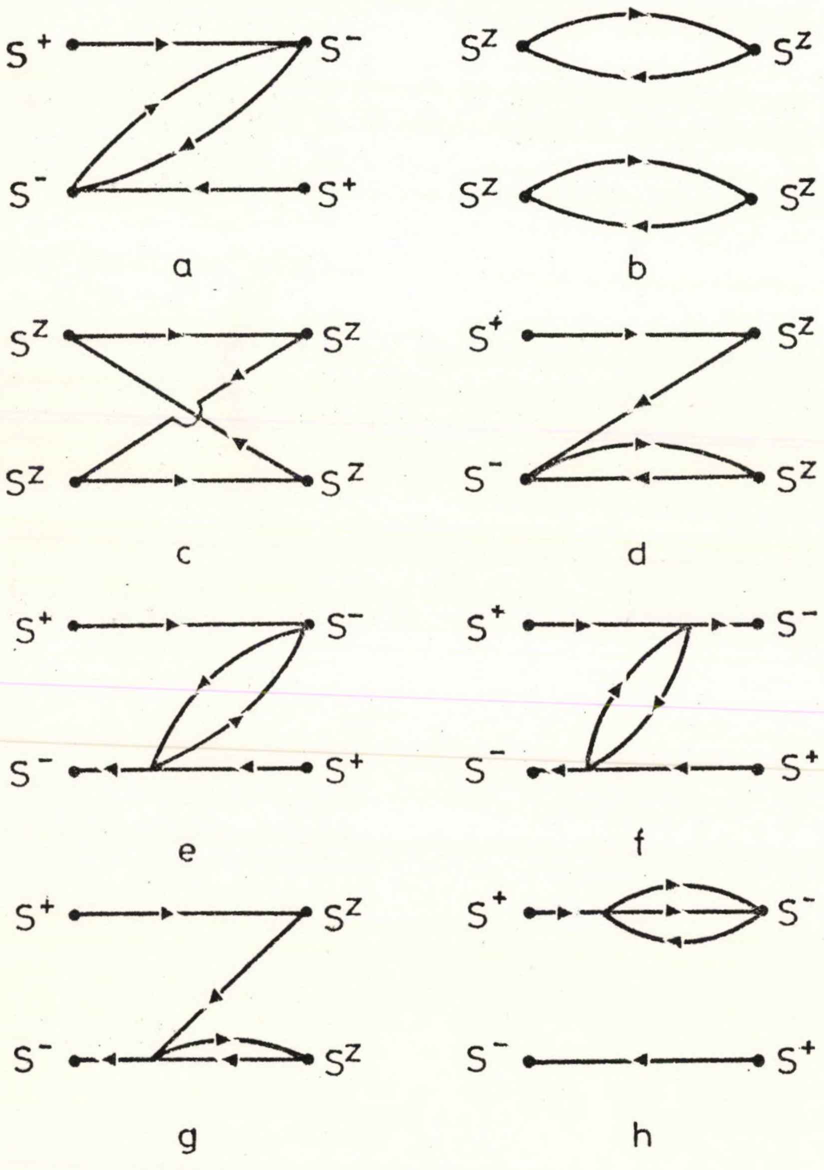
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Fig. 1

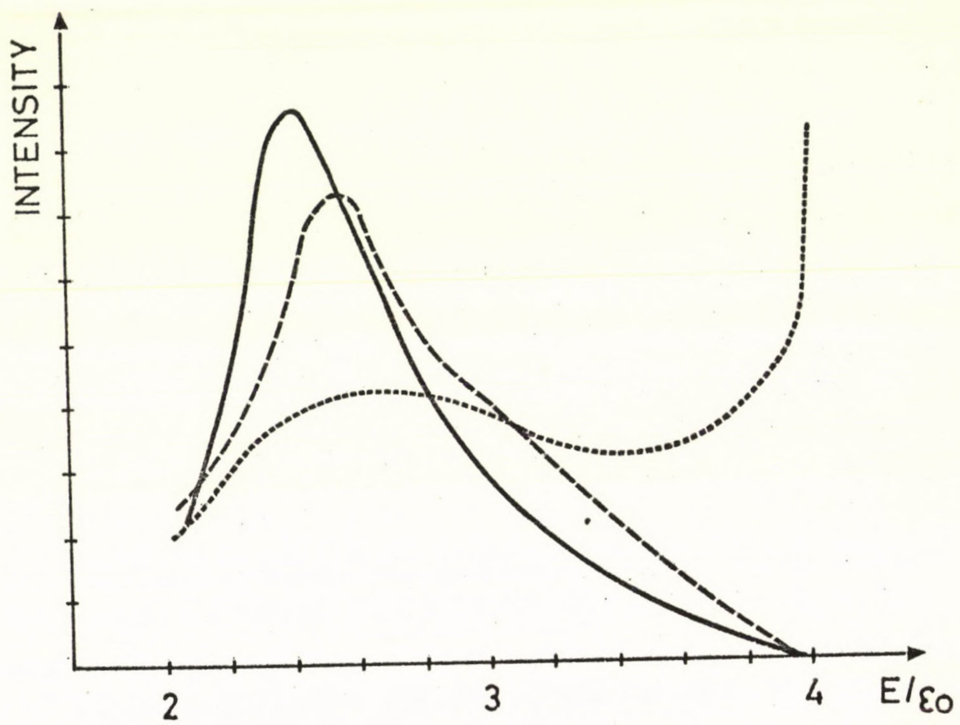

Fig. 2

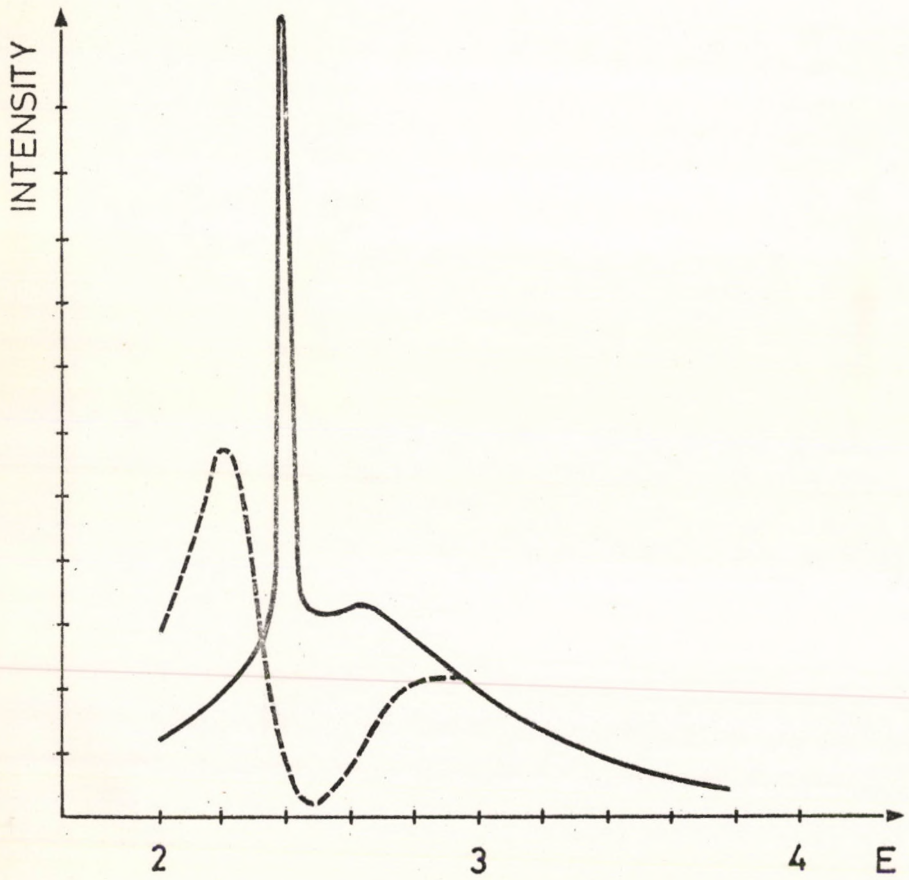

Fig. 3

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Kiadja a Központi Fizikai Kutató Intézet
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Szilárdtestfizikai Tudományos Tanácsának elnöke
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1972.április hó