Parametric generation of high frequency magnons with momenta at the Brillouin zone boundary in antiferromagnetic materials

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It is shown that if the light intensity I at the frequency of exciton-magnon absorption in antiferromagnetic materials exceeds a threshold intensity $I_{\rm th}$, a parametric generation of magnons with momenta at the Brillouin zone boundary occurs. In this case magnons are generated at specific points of the Brillouin zone.

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1. It is well known (see, for example, Ref. 1) that spin waves can be excited parametrically in magnetic materials; however, their wave vectors q (as well as frequencies) are sufficiently small compared to threshold frequencies, so that $q \leqslant 10^5 - 10^6\ \mathrm{cm}^{-1} \ll q_{Br}$, where q_{Br} is the quasi momentum at the Brillouin zone boundary, can be attained. Below we examine the mechanism of parametric generation of spin waves, which can generate high-frequency magnons with wave vectors $q = q_{Br}$.

Let us examine a two-particle absorption in magnetic crystals with two sublattices. It is known [2] that exciton-magnon absorption of light occurs in such crystals in the exciton region of the spectrum of a given crystal. Therefore, if the light intensity at the frequency of the exciton-magnon absorption is sufficiently large, then the probability (per unit time) of induced magnon and exciton radiation, which is proportional to the intensity of light, will be larger than the relaxation frequencies; as a result, a parametric generation of quasi particles occurs.

2. In examining this effect, it is convenient to use the generalized kinetic equations[3] obtained by using Keldysh's procedure.[4] For the magnon occupation numbers $N_{\mathcal{M}}(\mathbf{q})$ [and hence for exciton occupation numbers $N_{ex}(\mathbf{q})$] the balance equation holds

$$N_{\mathcal{M}}(\mathrm{q})\,\gamma_{\mathcal{M}} = \lambda[N_{\mathcal{M}}(\mathrm{q}) + N_{ex}(\mathrm{q}) + 1]\,\phi(\omega_{12}(\mathrm{q})), \tag{1}$$

where $\lambda = (2/\pi) I K / \hbar\nu_0 \sigma_{12} \Delta\nu$, K is the absorption coefficient of light at the exciton-magnon absorption frequency ν_0, I is the intensity of light, $\phi(\nu)$ is the spectral shape of the line, which is assumed to be Lorentzian with a half-width $\Delta\nu$, $\sigma_{12} = \sigma_{12}(\nu_0)$ is the two-particle (exciton-magnon) density of states, and $\omega_{12}(\mathbf{q}) \equiv \omega_{\mathcal{M}}(\mathbf{q}) + \omega_{ex}(\mathbf{q})$. Thus, we can assume that the spectral width of the pumping laser $\Delta\nu$ is larger[1] than the relaxation frequencies of the quasi particles ($\gamma_{\mathcal{M}}$ is the magnon frequency and $\gamma_{\rm ex}$ is the exciton frequency). Solving Eq. (1) and the equation for $N_{ex}(q)$ simultaneously, we can see that the solutions have a characteristic denominator that vanishes at a specific pumping power, which corresponds to the threshold of the parametric instability. Note that the threshold is determined by the smallest relaxation frequency; we assume that $\gamma_{\mathcal{M}} < \gamma_{ex}$ holds. Thus, the concentrations of quasi particles are inversely proportional to the relaxation frequencies $N_{\mathcal{M}}/N_{ex} = \gamma_{ex}/\gamma_{\mathcal{M}} > 1$. The threshold pumping intensity is[2]

$$I_{\Pi} \sim \frac{\hbar\nu_0 \gamma_M(q_{q_{Br}})\Delta\nu}{K a_0^3 \omega_M(q_{q_{Br}})}, \tag{2}$$

where a_0 is the lattice constant. Allowance for nonlinear attenuation of quasi particles gives a finite value of the magnon and exciton occupation numbers and also of the line width at the frequency $\Delta\omega$ in the distribution of the generated quasi particles. The total number of quasi particles and $\Delta\omega$ are determined by the α constant in the nonlinear attenuation $\gamma_{NL} = \alpha N$, which is dependent upon "adhesion" processes of magnons. The total number of magnons is proportional to α^{-2} and $\Delta\omega \sim \alpha$.

3. As a result of increasing light intensity at the exciton-magnon absorption frequency, the absorption coefficient increases substantially as it approaches the threshold value. The absorption coefficient approaches infinity at the threshold when the nonlinear attenuation is not taken into account and it is proportional to α^{-2} when the nonlinear attenuation is taken into account. The parametric generation of short-wave magnons can also be determined from the variation of the spectrum of other spin waves and from the sharp increase of single-magnon scattering of neutrons.

We note that the motion of short-wave magnons can be investigated in large samples whose linear dimensions are $l > K^{-1}$ (usually $K \sim 3 - 10\ \text{cm}^{-1}$).

Magnons participate in exciton-magnon absorption at certain symmetrical points at the zone boundary, which means that magnons with a boundary quasi momentum q_{Br}, which has a specific direction in the Brillouin zone, are generated. Thus, for MnF_2 magnons are generated at points Z and A on the zone boundary as a result of $E_\omega - C_4$ polarization of light and for $E_\omega - C_4$ they are generated at the point X, where C_4 is the fourfold axis of symmetry. Thus, the nonlinear attenuation is anomalously small for generation of magnons with a quasi momentum q_A, which gives a correspondingly large total number of excited magnons with q_A. The small α in this case is attributed to the fact that, since these magnons have a maximum frequency, the four-magnon exchange interaction does not lead to relaxation (as a result of interaction of the two magnons with q_A two different magnons with the same frequency are produced). As a result, only substantially weaker (relativistic) interactions can contribute to the nonlinear attenuation. Note also that the threshold intensity $I_{\rm th}$ for the magnons with q_A is smaller than for the other short-wave magnons.

4. Let us turn to estimates. We shall examine an "easy-plane" hexagonal $CsMnF_3$ antiferromagnet which at low temperatures $T\sim 1.5$ K has sufficiently low magnon relaxation frequencies.[5] Thus, at $K\sim 3$ cm^{-1} and $\gamma_{\mathscr{M}}\sim 10^7$ sec^{-1} the threshold intensity of pumping for generation of magnons with q_{Br} is of the order of 5×10^5 W/cm^2. Thus, we assume that pulsed, solid-state lasers, whose spectrum width is $\Delta\nu\sim 10^9$ sec^{-1}, can be used[3)] for pumping.

We note that the required threshold power is much lower than of the order 10^8 W/cm^2 at which non-steady-state, acoustic short-wave phonons were experimentally observed in a diamond crystal.[6]

[1)]In the opposite case (for narrow-band pumping) the equations are more cumbersome.[3] Note that at low temperatures wide-band pumping can be used for magnetic materials when solid-state lasers are used.

[2)]Here we take into account the fact that the group velocity of a magnon is much larger than that of an exciton.

[3)]*The dye lasers can also be used in the synchronizing mode; in this mode $\Delta\nu$ for the solid-state lasers can* also be less than 10^9 sec^{-1}.

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