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EXCITON SCATTERING ON SPIN WAVES IN QUASI-ONE-DIMENSIONAL ANTIFERROMAGNET

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Abstract. — Exciton absorption bandwidths have been measured in a quasi-one-dimensional antiferromagnet CsMnCl₃.2H₂O and a three-dimensional MnF₂ in magnetic fields oriented along crystal "easy" axes. In one-dimensional antiferromagnets exciton-magnon interaction is shown to result in a considerable broadening of exciton bands in the vicinity of the critical field of the spin-flop transition.

In one-dimensional (Id) antiferromagnetic (AFM) dielectrics the spin wave dispersion has strong anisotropy: at the wave vector $k$ oriented along the strong exchange interaction direction, the magnon energy changes from $\varepsilon_0$ ("a slit" in the spectrum, i.e. the spin wave energy in the Brillouin zone center) to $\varepsilon_{\text{max}}$ (the maximum energy at the zone boundary); for transverse directions the energy dispersion is too small. It leads to additional peculiarities in the energy distribution of density of magnon states, $\rho(\varepsilon)$: besides the peak near the band "top", $\rho(\varepsilon_{\text{max}})$, there appears the peak near its "bottom", $\rho(\varepsilon)$.

In the present work the exciton absorption bandwidth has been studied in Id AFM CsMnCl₃.2H₂O. It is shown that its behavior near the orientational phase transition induced by an external magnetic field (the spin-flop transition, $H_c=1.8$ T at $T=1.96$ K) is due to exciton scattering on spin waves. In this case the high density of magnon states near the band "bottom" in Id AFM plays a predominant role. For comparison similar studies of the exciton bandwidth were performed in a three-dimensional (3d) AFM MnF₂.

In order to study experimentally the broadening mechanism of light absorption bands, the temperature dependences of their half-widths are generally investigated [1]. However, for such an experiment, it is difficult to distinguish contributions to the bandwidth from both the interaction of excitons with magnons and their interaction with acoustic phonons. In the present work we fixed temperature (as low as possible, in order to diminish the phonon effect of the bandwidth) and varied the number of thermally excited magnons, thereby decreasing "a slit" in the spin-wave spectrum by an external field $H$ oriented along the crystal "easy" axes.

Objects of investigation were the exciton band of the $^6A_{1g}(^6S) \rightarrow ^4T_{2g}(^4P)$ transition in a CsMnCl₃.2H₂O crystal in a field $H \parallel b$ at 1.96 K, and the exciton band of the $^6A_{1g}(^6S) \rightarrow ^4T_{1g}(^4P)$ transition in MnF₂ in a field $H \parallel C_4$ at $T = 14$ K. Figure 1 gives the dependences of band half-widths in CsMnCl₃.2H₂O ($\circ$) and MnF₂ ($\bullet$) on the magnetic field strength oriented along the "easy" axes of these AFM crystals. In the case of manganese fluoride the exciton band half-width increases by about 10% in a field $H \approx H_c$ as compared with the initial value. For CsMnCl₃.2H₂O the band broadens approximately by a factor of 3.5 near the $H_c$ field. In the spin-flop phase ($H > H_c$) the exciton band half-width in CsMnCl₃.2H₂O takes again its initial value in the absence of an applied field.
The exciton-magnon interaction operator responsible for the exciton scattering on magnons can be represented in the form

\[ \mathcal{H}_{\text{exc-mag}} = \frac{JS_z}{N} \sum_{k_1, \ldots, k_3} \times \left[ \Phi_{11}(k_2, k_3) B_{\mu}^+(k_1 - k_2 + k_3) B_{\mu}(k_1) b^+(k_2) b(k_3) + \Phi_{22}(k_2, k_3) B_{\mu}^+(k_1 + k_2 - k_3) B_{\mu}(k_1) b(k_2) b^+(k_3) \right] \]

Here \( J \) is the exchange integral along the \( a \)-direction, \( z = 2 \) the number of nearest neighbours along the \( a \)-chain; \( B_{\mu}^+(k) \) and \( B_{\mu}(k) \) exciton creation and annihilation operators, respectively, in the \( \mu \) band, and \( b^+(k) \) and \( b(k) \) those of magnons of the low frequency branch; \( N \) the number of crystal lattice sites, \( S \) the spin of the ground state.

The interaction amplitudes \( \Phi_{\mu} \) are of the form

\[ \Phi_{11}(k_2, k_3) = -\varphi V_{k_2} U_{k_3} \gamma_{k_3} + e U_{k_2} U_{k_3}; \]
\[ \Phi_{12}(k_2, k_3) = -\varphi V_{k_2} U_{k_3} \gamma_{k_3} + \rho U_{k_2} U_{k_3} \gamma_{k_2 - k_3}; \]
\[ \Phi_{21}(k_2, k_3) = -\varphi V_{k_2} U_{k_3} \gamma_{k_3} + \rho V_{k_2} V_{k_3} \gamma_{k_2 - k_3}; \]
\[ \Phi_{22}(k_2, k_3) = -\varphi V_{k_2} U_{k_3} \gamma_{k_3} + e V_{k_2} V_{k_3}; \]

where

\[ \varphi = \frac{J'}{J} \left( \frac{S'}{S} \right)^{1/2} - 1; \quad \rho = \frac{J' S'}{JS} - 1; \quad e = \frac{J'}{J} - 1; \]

\( J' \) and \( S' \) are the exchange integral and the spin in the excited state, respectively; \( U_{k} \) and \( V_{k} \) coefficients of the \( u-v \) transformation; \( \gamma_{k} = \frac{1}{z} \sum_{\alpha} \exp(ik\Delta_{\alpha}) \) with the summation over the nearest neighbours of \( a \).

The exciton absorption band shape is described by the one-particle delay exciton Green function. Taking into account the exciton-magnon interaction (1), the approximate calculation of this function near the \( H_c \) field showed that for a 1d AFM a change of the exciton band half-width is \( \delta H \sim (1 - H/H_{c1})^{-1} \), for a 2d AFM it is \( \delta H \sim -\ln (1 - H/H_{c2}) \) and for a 3d AFM \( \delta H \) has a finite limit near a field \( H_{c3} \).

For comparison between the calculation and the experimental results, figure 1 presents two curves (solid lines) corresponding to theoretical relations: a) \( \delta H \sim (1 - H/H_{c1})^{-1} \) refers to a 1d AFM, b) \( \delta H \sim -\ln (1 - H/H_{c2}) \) to a 2d AFM. Experimental points (o) relating to CsMnCl\(_3\).2 H\(_2\)O are concentrated between the curves and meet mainly the a) dependence corresponding to 1d AFM. For a 3d AFM the exciton band broadening is negligible in a field \( H \leq H_{c3} \) that is in agreement with the absence of peaks near the band "bottom" in the magnon state density distribution.

In the spin-flop phase the energy of the spin-wave band "bottom" increases by steps leading to a sharp decrease of magnon occupation numbers. As a result, the exciton band half-width in CsMnCl\(_3\).2 H\(_2\)O takes its initial value for \( H = 0 \).

High magnon state density near "the bottom" of the spin-wave band manifests itself in a 1d AFM by the shapes not only of pure exciton bands, but also of their magnon sidebands [2]. Figure 2 indicates spectral distribution of the light absorption coefficient \( K(e) \) in CsMnCl\(_3\).2 H\(_2\)O in the vicinity of the exciton band studied and its magnon sideband. Unlike 3d AFM in CsMnCl\(_3\).2 H\(_2\)O, the exciton-magnon absorption is of a band character with the pronounced maximum not only near the frequency corresponding to the magnon band "top" (\( \varepsilon_{\text{max}} \sim 25 \text{ cm}^{-1} \)), but also to its "bottom" (\( \varepsilon_{\text{bottom}} \sim 2 \text{ cm}^{-1} \)). The analysis of exciton-magnon band shapes showed that besides high magnon state density \( \rho(e) \) near \( \varepsilon_{\text{bottom}} \), the significant factor which defines the band character of the exciton-magnon absorption is the absence of the inversion center in the location of an absorbing magnetic ion (this condition is met in CsMnCl\(_3\).2 H\(_2\)O).

Fig. 2. – Densitogram of a part of the CsMnCl\(_3\).2 H\(_2\)O absorption spectrum near the exciton-magnon band; the sample thickness is \( d = 7.1 \text{ mm}, T = 1.65 \text{ K} \). Light propagates along the \( b \)-axis, the light wave electric vector is \( \mathbf{E}_\omega \parallel c \). Count is from the exciton line frequency.