Light-matter interaction in antiferromagnets: the exchange-induced magnetic dipole mechanism

M. V. Eremin,1 M. A. Fayzullin,1 Ch. Kant,2 A. Loidl,3 and J. Deisenhofer4

1Institute for Physics, Kazan (Volga region) Federal University, 420008 Kazan, Russia
2Institut für Festkörperphysik, Technische Universität Wien, 1040 Wien, Austria
3Experimentalphysik V, Center for Electronic Correlations and Magnetism, Institute for Physics, Augsburg University, D-86135 Augsburg, Germany

(Dated: September 6, 2011)

We propose a novel mechanism for exchange-induced exciton-magnon absorptions via hopping between two antiferromagnetically coupled sites and a simultaneous magnetic dipole transition to an excited orbital state. The obtained selection rules correspond to ones for magnetic dipole transitions and are in agreement with the exciton-magnon transitions observed in the quasi-one-dimensional Heisenberg antiferromagnet KCuF3. The calculated magnon density of states in combination with a structure dependent factor identifies the observed optical magnon sideband to originate from transverse magnon modes.

PACS numbers: 78.20.Bh, 71.70.Ej, 75.30.Et, 75.30.Ds, 78.20.Ls

The interaction of light and matter is one of the most fascinating research fields in physics. In materials with coupled electronic, lattice, and spin degrees of freedom such as, for example, antiferromagnetic transition metal compounds, electric-dipole active magneto-optical excitations including two-magnon, exciton-magnon, and exciton-exciton absorptions have been observed [1–3]. In pioneering works Tanabe and coworkers proposed mechanisms to explain these absorption processes in terms of cooperative excitations of antiferromagnetically coupled ions [4–8]. The observation of electric-dipole active three-magnon in α-Fe2O3 [9,11] and one-magnon processes (electromagnons) in multiferroic manganites, respectively, pushed the search for such excitations and their microscopic mechanisms [12,13].

Recently, the observation of magnon sidebands at the onset of the phonon assisted orbital transitions was reported in the quasi-one dimensional Heisenberg antiferromagnet KCuF3 [14]. The structure of this compound is sketched in Fig. 1(a) together with the antiferroorbital arrangement of the d_{x^2−y^2} and d_{z^2−r^2} orbital states of the Cu^{2+} ions with a 3d^9 electronic configuration. In Fig. 1(b) we show the splitting of the d-levels at the copper site in D_{2h} symmetry (considering one hole occupying a d_{x^2−z^2} state) and indicate the excitations A_1−A_4 observed in [14].

It is important to note that transitions to the excited d_{xz}, d_{xy}, and d_{yz} orbital states are magnetic dipole allowed via the orbital momentum operators l_α (α = y, z, x), respectively. In Fig. 1(c) the absorption spectra at 8 K for polarization of the incoming light E || c and E ⊥ c are shown, focussing on the onset of A_2 and A_3. The sharp peaks at 8508 cm\(^{-1}\) and 9775 cm\(^{-1}\) correspond to the respective purely orbital transitions with energy, while the sidebands at a distance of about 88 cm\(^{-1}\) and 114 cm\(^{-1}\) were identified as exciton-magnon transitions [14]. The inset of Fig. 1(c) shows the polarization dependence of the intensity of the peak at 8508 cm\(^{-1}\) following a sin\(^2\)φ dependence, with φ being the angle between the polarization and the crystal axes. This polarization dependence of the zero-magnon line and the whole set of sidebands is in agreement with the selection rules expected for magnetic dipole transitions, but at odds with the expected behavior from the canonical exchange-induced electric dipole (EIED) mechanism.
Here we propose a novel mechanism to explain the exciton-magnon transitions in KCuF$_3$. The resulting exchange-induced magnetic dipole (EIMD) processes allow to describe the observed polarization dependence and are compared to the EIED mechanism.

We consider two exchange-coupled magnetic ions at sites $a$ and $b$. Using the canonical transformation with S-matrix formalism, the effective Hamiltonian

$$H_{\text{eff}} = F + [F, S] + \frac{1}{2}[[F, S], S] + ...$$

is obtained and accounts for all possible processes up to third order of perturbation theory. The operator $F$ describes transitions between different orbital states $\varphi$ and $\varphi'$ of the ion at site $a$

$$F = \mu_B \sum_{\varphi, \varphi'} a_{\varphi}' < \varphi | H_{\alpha}^a a_{\alpha}' | \varphi' > a_{\varphi'}.$$

Here, $\mu_B$ denotes the Bohr magneton, $H_{\alpha}^a$ the magnetic component of the electromagnetic wave along $\alpha = x, y, z$ and $a_{\alpha}'$ is the orbital momentum operator at site $a$. $a_{\varphi}'$ and $a_{\varphi''}$ are the electron creation and annihilation operators. The operator $S$ is given by

$$S = \sum_{\xi, \eta} \frac{t_{\xi \eta}}{\Delta_{\xi \eta}} b_{\xi}^\dagger a_{\eta} - \sum_{\eta, \xi} \frac{t_{\eta \xi}}{\Delta_{\eta \xi}} a_{\eta}^\dagger b_{\xi},$$

where $t_{\xi \eta}$, $t_{\eta \xi}$ are effective hopping integrals and $\Delta_{\xi \eta}$, $\Delta_{\eta \xi}$ are charge-transfer energies. The commutator $[F, S]$ describes processes of the electron hopping between the two sites with simultaneous excitation by light absorption as well as emission from excited states of the $a$-site ion. Note that none of these processes involves a spin flip.

One of the virtual processes we are considering here is sketched in Fig. 2(c). The process involves a magnetic dipole transition to an excited orbital state at site $a$ (dashed arrow) and hopping processes of holes between the $a$ and $b$ sites in the ground states (solid arrows). The effective Hamiltonian corresponding to the relevant processes of the magnetic dipole transition can be derived from the double commutator in Eq. (1). Here we are interested in the effective operator for the absorption process which can be written in terms of spin operators for the exchange-coupled ions at the $a$ and $b$-sites

$$H_{\text{eff}}^{\varphi \rightarrow \varphi'} = \frac{\mu_B}{2\Delta} < \varphi | H_{\alpha}^a a_{\alpha}' | \varphi' > J_{ab}^c [\sigma_a S_b - 1/4],$$

where $J_{ab}^c$ is the superexchange coupling constant between the ground states of ions $a$ and $b$. This contribution has to be summed over all nearest neighbors at sites $b$. Strictly speaking the introduced operator $\sigma_a$ is not a true spin operator, because it has matrix elements between the different orbital states. Evidently, the suggested EIMD process is only effective for a pair of antiferromagnetically coupled ions. This effective Hamiltonian can be interpreted as follows: the absorption (or creation of an exciton) on considering an optically active center on site $a$ gives rise to magnons in the surrounding magnetic sublattice.

Let us now discuss this mechanism for the case of KCuF$_3$ and compare it to the EIED mechanism suggested by Tanabe, Moriya and Sugano [4]. In Fig. 2(a) we schematically show spatial distributions of ground and excited orbital states of copper ions along the antiferromagnetic $c$-axis. The scheme on the left hand side refers to the $A_2$ transition and the one on the right to the $A_3$ transition. The respective ground state orbital states at sites $a$ and $b$ are $\eta = |x_a^2-z_a^2\rangle$ and $\xi = |y_b^2-z_b^2\rangle$, the respective excited orbital states at site $a$ corresponding to $A_2$ and $A_3$ are $\varphi' = |xz\rangle$ and $|xy\rangle$.

The case of the EIED mechanism suggested by Tanabe, Moriya and Sugano [4] is illustrated in Fig. 2(b). The up-spin at the $a$ site ($\eta$) hops to the $b$ site ($\xi$) while the down-spin at site $b$ is transferred to an excited orbital state at site $a$ due to interaction of the electric-dipole moment with the electric-field component of the incoming light. This mechanism will only lead to a non-zero ma-
element for the $A_2$ transition if $E \perp c$. In case of the $A_3$ transition the EIED transition there is no non-zero matrix element. These selection rules, however, are at odds with the experimentally observed polarization dependence.

The processes involved in the EIMD mechanism are schematically sketched in Fig. 2(c). The down-spin at site $b$ (orbital $\xi$) hops to the ground state of ion a, and then is excited to the orbital state $\varphi'$ due to the interaction of the orbital momentum with the magnetic field component of the incoming light and simultaneously the up-spin at site $a$ hops to site $b$. The selection rules, therefore, correspond to the ones for magnetic dipole transitions in agreement with the experimental observation [13].

Furthermore, we calculate the magnon density of states for KCuF$_3$ using the spin-wave Hamiltonian in the framework of a two-sublattice model [15], but neglecting $XY$-like anisotropy and contributions from the Dzyaloshinsky-Moriya interaction [16]. Diagonalization via Holstein-Primakoff and Bogoliubov transformations yields the magnon dispersion

$$E_k = \sqrt{[8J^a(1-\theta_a) + 2J^c] - 4J^c \gamma_c^2}, \quad (5)$$

where $J^c$, $J^a$ are the antiferro- and ferromagnetic exchange coupling constants, respectively and $\theta_a = (\cos k_x a + \cos k_y a)/2$, $\gamma_c = \cos k_z c$. This dispersion was used to fit the spin-wave spectrum observed in inelastic neutron scattering experiments [15] with $J^c = 320$ K and $J^a = 1.6$ K. The same expression for the transverse magnon mode was derived in [17]. The magnon density of states (DOS) $\rho(E)$ was calculated by using this dispersion and summing of the magnetic Brillouin zone (BZ), which is doubled along the $c$ direction. The calculated result is shown in Fig. 3 together with the optical absorption coefficient for light polarization $E \parallel c$. The magnon DOS exhibits four singularities, which corresponds to the points $X[\pi/a, 0, 0]$, $M[\pi/a, \pi/a, 0]$, $Z[0, 0, \pi/2c]$ and $R[\pi/a, 0, \pi/2c]$ in the magnetic BZ. The corresponding energies are 11.1, 15.7, 55.2, and 56.3 meV, respectively. The first two magnon energies correspond very nicely to the optically observed sideband peaks (see Fig. 3). Although the DOS at the $Z$ and $R$ points is very large with respect to $X$ and $M$ points, there are no prominent features at these energies in the absorption spectra. This can be explained by considering structure dependent factors which we discuss in the following.

In general, the absorption coefficient caused by the magnetic field component of light with frequency $\omega$ and polarization $\nu$ is proportional to the imaginary part of the susceptibility and can be evaluated in terms of Green’s function [18]

$$A^\nu(E) \propto -E \nu \frac{1}{V} \lim_{\delta \to 0} \Im[G(M_\nu, M_\nu)_{E + i\delta}], \quad (6)$$

where $V$ is the sample volume, $E = h\nu$, $M_\nu$ is the component of the transition magnetic dipole moment. Based on Eq. 4 in the low temperature approximation we define

$$M_\nu = \sum_{m,l} \pi_{ml}^\nu c_m^+ S_l^z, \quad (7)$$

where $c_m^+$ is a exciton creation operator of the spin-up sublattice, $S_l^z$ is the spin operator of the spin-down sublattice and $\pi_{ml}^\nu = \mu_B < \varphi |l^\nu |\varphi > J_{ml}/4\Delta$. Following the Green’s function formalism [19] we obtain

$$G(M_\nu^*, M_\nu) = \sum_k \frac{\left| \pi_{k}^{\nu} \right|^2 u_k^2}{E - E_{ex} - E_k}, \quad (8)$$

Here, $E_k$ is the magnon energy, $E_{ex}$ is the exciton energy of the zero-magnon (phonon) line, $\pi_{k}^{\nu} = \mu_B J^c |\varphi||l^\nu|\varphi > \cos k_z c/2\Delta$, and $u_k^2 = (1 + \sqrt{1 + Z_k^2})/2$, $Z_k = 2J^c \gamma_c/E_k$. Then substituting the Green’s function (Eq. 8) to (Eq. 6) we finally get

$$A^\nu(E) \propto E \nu \frac{\pi}{V} J^c \mu_B \left| \varphi ||l^\nu|\varphi > \right|^2 \sum_{k}^{BZ} \left| u_k^2 \cos^2 k_z c \delta(E - E_{ex} - E_k) \right|. \quad (9)$$
The obtained absorption coefficient contains the structure-dependent factor $\cos^2 k_z c$, which filters out the singularities corresponding to Z and R points in DOS during the summation over the BZ, but the singularities corresponding to X and M points remain. This explains the absence of prominent magnon sidebands at 55.2 and 56.3 meV. We would like to point out that in the case of the EIED mechanism the factor $\cos^2 k_z c$ is replaced by $\sin^2 k_z c$ and, therefore, low-energy singularities corresponding to X and M points in the magnon DOS are filtered out. The high-energy singularities would be expected to appear in the optical spectrum for $E \perp c$ as a result of the EIED mechanism and the large DOS at the Z and R points, but no experimental evidence of such sidebands is visible in the spectra. The reason of this suppression of the allowed EIED is not clear at present. Thus, the new EIMD mechanism allows to describe the observed low-energy magnon sidebands at 11.1 and 15.7 meV in KCuF$_3$, but the further sideband peaks visible in Fig. 1(c) have no correspondence in the calculated one-magnon DOS and their origin can not easily be settled. Possibly, multi-magnon processes or the presence of spinons will have to be considered and need further theoretical investigations.

In summary, we have presented a new mechanism of a cooperative exciton-magnon absorption in antiferromagnets – the exchange-induced magnetic-dipole mechanism. The selection rule corresponds to the one for magnetic dipole transitions and describes the experimental results in antiferromagnetic KCuF$_3$. Moreover, we showed that for this novel mechanism the magnon density of states and the structure factor in KCuF$_3$ are responsible for the observability of low-energy magnon sidebands, while the high-energy magnon sidebands are suppressed.

It is a pleasure to thank Sergei Nikitin for fruitful discussions. We acknowledge support by DFG via TRR 80 (Augsburg-Munich). MVE is partially supported by Ministry Education of the Russian Federation via Grant No. 1.83.11.

[1] J. W. Halley and I. F. Silvera, Phys. Rev. Lett. 15, 654 (1965).
[2] R. L. Greene, D. D. Sell, W. M. Yen, A. L. Schawlow, and R. M. White, Phys. Rev. Lett. 15, 656 (1965).
[3] J. Ferguson, Australian J. Chem. 21, 307 (1968).
[4] Y. Tanabe, T. Moriya, and S. Sugano, Phys. Rev. Lett. 15, 1023 (1965).
[5] K.-I. Gondaira and Y. Tanabe, J. Phys. Soc. Jap. 21, 1527 (1966).
[6] Y. Tanabe and K.-I. Gondaira, J. Phys. Soc. Jpn. 22, 573 (1967).
[7] K. Shinagawa and Y. Tanabe, J. Phys. Soc. Jap. 30, 1280 (1971).
[8] T. Fujiwara, W. Gebhardt, K. Petanides, and Y. Tanabe, J. Phys. Soc. Jap. 33, 39 (1972).
[9] S. Azuma, M. Sato, Y. Fujimaki, S. Uchida, Y. Tanabe, and E. Hanamura, Phys. Rev. B 71, 014429 (2005).
[10] Y. Tanabe and E. Hanamura, J. Phys. Soc. Jap. 74, 670 (2005).
[11] Y. Tanabe, Y. Fujimaki, K. Kojima, S. Uchida, S. Onari, T. Matsuo, S. Azuma, and E. Hanamura, Low Temp. Phys. 31, 780 (2005).
[12] A. Pimenov, A. A. Mukhin, V. Yu. Ivanov, V. D. Travkin, A. Balbashov, and A. Loidl, Nature Phys. 2, 97 (2006).
[13] M. Mostovoy, Phys. Rev. Lett., 96, 067601 (2006).
[14] J. Deisenhofer, I. Leonov, M. V. Eremin, Ch. Kant, P. Ghigna, F. Mayr, V. V. Iglamov, V. I. Anisimov, and D. van der Marel, Phys. Rev. Lett. 101, 157406 (2008).
[15] S. K. Satija, J. D. Axe, G. Shirane, H. Yoshizawa, and K. Hirakawa, Phys. Rev. B 21, 2001 (1980).
[16] M. V. Eremin, D. V. Zakharov, H.-A. Krug von Nidda, R. M. Eremina, A. Shuvavev, A. Pimenov, P. Ghigna, J. Deisenhofer, and A. Loidl, Phys. Rev. Lett. 101, 147601 (2008).
[17] B. Lake, D. A. Tennant, C. D. Frost, and S. E. Nagler, Nature Materials 4, 329 (2005).
[18] I. V. Alexandrov, Theory of Magnetic Relaxation in Liquid and Solid Non-Metallic Paramagnetics (Nauka, Moscow, 1975).
[19] J. B. Parkinson and R. Loudon, J. Phys. C, 1, 1568 (1968).