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HAL Id: jpa-00228607
https://hal.archives-ouvertes.fr/jpa-00228607
Submitted on 1 Jan 1988

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EXCHANGE SPIN WAVES AND THEIR MANIFESTATION IN TWO-MAGNON ABSORPTION AND RAMAN SCATTERING

V. V. Eremenko, S. A. Zvyagin, Yu. G. Pashkevich, V. V. Pishko, V. L. Sobolev and S. A. Fedorov

Abstract. — Spin waves whose activation energy in exchange approximation remains finite and is defined by exchange magnetic interaction integrals are called exchange spin waves (ESW). In antiferromagnets with $n$ magnetic sublattices, collinear in exchange approximation the SW spectrum exhibits $n-2$ exchange branches. Peculiarities of the ESW manifestation in the two-magnon absorption (TMA) and Raman scattering (RS) are traced in rhombic antiferromagnets.

Dispersion dependences of the SW spectrum, $\omega(k; H)$ have been studied in antiferromagnetic CuCl$_2$·2H$_2$O which has well-known parameters characteristic of its magnetic structure. DC magnetic field was applied along the easy magnetization axis (a-axis). The SW spectrum have been studied for all magnetically ordered phases possible for this field orientation. The Dzyaloshinski-Meriya interaction leading to weak canting of sublattice magnetizations and the arising weak ferro- and antiferromagnetism vectors is responsible for the appearance of coupling between oscillations of the same symmetry corresponding to exchange and acoustic SW spectrum branches. For uniform precession of magnetization in oscillations corresponding to exchange modes weak antiferromagnetism vectors have the largest oscillation amplitude. The oscillation amplitudes of the magnetization vector and the principal antiferromagnetism vector involve a small parameter $D_1^{-1}$ ($D$ is the Dzyaloshinski-Moriya interaction, $I$ is the exchange constant). The latter makes it different to observe exchange modes (due to weak signals) by antiferromagnetic resonance (AFMR) and RS. Frequency-field dependences of the AFMR spectrum in CuCl$_2$·2H$_2$O were studied experimentally at $T = 2$ K under applied magnetic fields of 0-10 T. The $D$ constant has been determined [1] by studying the region of strong interactions of exchange and acoustic modes of the same symmetry existing for $H_{sf} \leq H \leq H_f$ ($H_{sf}$ is the spin-flop transition field, $H_f$ the spin-flip transition field). Similar interaction of oscillations with the same symmetry exists also in the spin waves, the wave vector region with this interaction being dependent on an applied magnetic field. In fields $0 < H < H_{sf}$ (collinear phase) and $H_{sf} \leq H \leq H_f$ the CuCl$_2$·2H$_2$O spectrum consists of four branches, two of them being exchanged ones, and two acoustic. In a field $H_f$ the second-order phase transition of the spin-flip type takes place. In this case the main antiferromagnetism vector vanishes and a magnetically ordered phase (spin-flip phase) is realized in which a magnetic unit cell coincides with a crystallographic one, the number of sublattices is halved, and there are only two SW spectrum branches instead of four.

Critical points (CP) of the SW spectrum where the group velocity is zero have been studied theoretically in detail. The existence of new dynamic CP due to the SW interaction has been demonstrated. The general criterion for the existence of such CP in the magnon spectrum of multisublattice magnets has been stated. The following conditions are required for their appearance. First, in the absence of interaction the SW branches should cross in a given spatial point of $k$-space, i.e. the so-called “cross situation" should appear. Secondly, in this point group velocities should be directed along the wave vector and opposite to each other. Taking into account the interaction between SW spectrum branches (in our case, it is the Dzyaloshinsky-Moriya interaction) results in pushing these branches apart and arising dynamic CP. The peculiarities of density of SW states inherent to multi-sublattice magnets have been studied systematically. The calculation of the effective mass tensor components has been performed near all possible CP. It has been established that some effective mass tensor components, as well as additions to densities of SW states near CP due to the interaction between spectrum branches depend on its value.

Polarization peculiarities of TMA have been studied in detail in magnetically ordered CuCl$_2$·2H$_2$O phases in an applied magnetic field parallel to the easy magnetization axis. TMA in the spin-flip phase has been analysed on that exchange mode whose TMA amplitude does not involve the small $D_1^{-1}$ parameter. The absorption intensity maximum arises at frequencies near the double SW energy with the wave vector being at the Brillouin zone boundary and in magnetic fields corresponding to appearing of zero derivative $\partial \omega(k; H) / \partial k$.

The TMA spectra have been obtained from CuCl$_2$·2H$_2$O single crystals cut along the crystallographic axes at $T = 2$ K.
In the frequency range from 12 to 15 cm\(^{-1}\) weak TMA bands are located on the pronounced AFMR line sideband of the acoustic mode that makes it difficult to observe them. It should be noted, that according to theoretical calculations, it is this frequency range where TMA is to be observed due to dynamic CP arising from "cross situation". We succeeded observing TMA bands in the frequency range of 17 - 23 cm\(^{-1}\) [2]. In accordance with the calculations, two exchange branches can arise in this range. The appearing of zero derivative of the dispersion curve of one of them leads to its higher TMA intensity. The observed absorption band location for the incident emission frequency \(\omega = 22\) cm\(^{-1}\), dc field orientation \(\mathbf{H}/a\) and polarization \(\mathbf{H}_\parallel/a\) is in agreement with the theory for the case of TMA on the exchange branch with zero derivative \(\partial \omega (k, H)/\partial k\). Variations of the emission polarization direction result in the TMA intensity decrease that is in good agreement with the theory as well. It should be noted that in this case the contribution to TMA on the exchange magnons from acoustic magnon states can be neglected due to a great energy difference between exchange and acoustic branches.

While studying RS the expansion of permittivity is, unlike the generally accepted approach, not over spins of separate ions, but over irreducible representations of the magnetic symmetry group. The first expansion term describes the contribution to the scattering tensor due to linear magneto-optical effects, and the second one due to quadratic magneto-optical effects. The tensor form of the one-magnon scattering can easily be obtained if the types of irreducible spin combinations taking part in oscillations of a given magnon mode are known [3]. The comparison between scattering intensities on exchange and acoustic modes of the same symmetry in rare-earth orthoferrites that the scattering intensity on the exchange mode in \((D/I)^{1/2}\) is less than that on acoustic one [3]. Similar relation is fulfilled for the spin-flop phase of CuCl\(_2\)-2H\(_2\)O under an applied magnetic field, where exchange and acoustic modes of the same symmetry exist. However, in the region of a strong interaction between exchange and acoustic modes, scattering intensities become magnitudes of the same order. In the case of scattering on the exchange modes in the collinear phase, the scattering intensity is independent of a magnetic field [4].

Exchange and acoustic mode frequencies can be of the same order of magnitude in quasi-one-dimensional and quasi-two-dimensional multisublattice magnetics with the anomalous relation of exchange intersublattice integrals. In this case scattering intensities on the entire magnetic modes seem to be the same order of magnitudes in a wide range of fields.

In [5] the contribution to the one-magnon scattering tensor on acoustic magnons was studied in EuTe from the exchange scattering mechanism related to quadratic magneto-optical effects in an applied magnetic field. This mechanism can be shown to contribute always to the RS tensor if the magnetic structure is noncollinear, noncollinearity being due to both the Dzyaloshinsky-Moriya interaction and a magnetic field. Moreover, there are cases when the given scattering mechanism contributes to off-diagonal components of the scattering tensor. However, in antiferromagnets collinear in the exchange approximation, the contribution from the exchange scattering mechanism is always attenuated by the small \(DI^{-1}\) factor.

We studied Raman scattering in a four-sublattice exchange-non-collinear antiferromagnet with the magnetic structure of the UO\(_2\) type. It has three acoustic and one exchange modes oscillations of which are transformed by the total symmetry representation. The scattering tensor on exchange modes is diagonal, each its component is formed by the exchange scattering mechanism. In this case the scattering tensor has no small factor, and the scattering intensity on exchange magnons is much higher than that on acoustic ones.