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To cite this version:
B. Hennion, J. Pierre. Magnetic excitations in HoZn and ErCu compounds. Journal de Physique Colloques, 1979, 40 (C5), pp.C5-141-C5-142. <10.1051/jphyscol:1979552>. <jpa-00218969>

HAL Id: jpa-00218969
https://hal.archives-ouvertes.fr/jpa-00218969
Submitted on 1 Jan 1979

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Magnetic excitations in HoZn and ErCu compounds

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Résumé. — Nous avons étudié les excitations magnétiques dans le composé antiferromagnétique ErCu à 4,3 K et dans le composé ferromagnétique HoZn entre 4,2 et 30 K. Nous en déduisons les interactions interatomiques et l'ordre de grandeur des couplages magnétoélastiques ou biquadratiques.

Abstract. — We have studied the magnetic excitations in the antiferromagnet ErCu at 4.3 K and in the ferromagnet HoZn between 4.2 and 30 K. We deduce the interatomic interactions and the order of magnitude of the magnetoelastic or quadrupolar couplings.

1. Introduction. — The aim of present work is to study the propagation and range of indirect interactions in cubic CsCl-type compounds: what type of band electrons mediates indirect couplings, are the interactions isotropic or not, may higher order exchange and magnetoelastic couplings be revealed through the dispersion relations? These questions have been thoroughly investigated in pure rare earths, both experimentally and theoretically [1, 2]. We analyze here experiments performed on ferromagnetic HoZn ($T_c = 74$ K) and antiferromagnetic ErCu ($T_N = 16$ K).

2. HoZn compound. — The magnetic excitations were previously studied at 4.2 [3] and two branches were followed through the Brillouin zone (BZ). HoZn undergoes at 23 K a first order spin rotation from [110] to [111] direction, driven by crystal field effects. We have studied the magnetic excitations by inelastic neutron scattering on IN2 triple axis spectrometer (Laue-Langevin Institute-Grenoble) in both orthorhombic and rhombohedral phases (figure 1). Additional branches appear above 21 K near the BZ center $\Gamma$, they arise from couplings between excited levels; their intensity vanish rapidly as the branch flattens.

Two main branches remain through the rest of the BZ, with a shape similar to that encountered at 4.2 K. A significant lowering occurs for the second branch at 23 K, associated with the lowering of the second excited level in the rhombohedral phase, which is responsible for the phase transition (lower free energy for this phase at $T > 23$ K).

The analysis is done within a dynamical susceptibility formalism [4]. The single ion hamiltonian includes within the molecular field (MF) model 1) crystal field as determined in the paramagnetic range, 2) Heisenberg exchange defined by the molecular field constant $\lambda = -3\theta/J(J+1)$, 3) second order magnetoelastic and quadrupolar couplings defined by two parameters $B_2$ and $B_3$ [3, 6]. The couplings are isotropic when $B_3 = 12 B_2$.

The parameters $\theta$, $B_2$, $B_3$ influence mainly the mean excitation energies and may be deduced after
a few try-and-error steps; the values $\theta = 72 \pm 2\,^\circ$, $B_2 = 4 \pm 2\,mK$, $B_3 = 15 \pm 15\,mK$ give a satisfactory fit from 4.2 to 30 K. These coefficients are in agreement with the analysis of static experiments (magnetization and magnetostriction) by Morin and Schmitt [5].

Inter-ion couplings are deduced by a least square fit of the dispersion curves at 21 K (table I), six parameters are found sufficient. Their determination agrees with the previous one at 4.2 and matches very well the experimental data at 25 K. Thus Heisenberg interactions are not strongly modified by the spin rotation and higher order interactions have a small influence on the dispersion. The energy contribution of these last terms obtained from the values of $B_2$ and $B_3$ is more than one order of magnitude smaller than Heisenberg contribution, thus such terms may be neglected in the inter-ion Hamiltonian within a first approximation.

Table I. — Exchange interactions $I$ (mK) with different neighbours (l, m, n). A negative value means a ferromagnetic coupling. Standard error : $\pm 2\,mK$.

<table>
<thead>
<tr>
<th>lmn</th>
<th>100</th>
<th>110</th>
<th>111</th>
<th>200</th>
<th>210</th>
<th>211</th>
</tr>
</thead>
<tbody>
<tr>
<td>HoZn</td>
<td>87</td>
<td>+2.3</td>
<td>-21</td>
<td>+9</td>
<td>-6.4</td>
<td>-11.5</td>
</tr>
<tr>
<td>ErCu</td>
<td>+18</td>
<td>+7.4</td>
<td>-22</td>
<td>-14</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

3. **ErCu compound.** — ErCu orders with a C-type antiferromagnetic structure; propagation vector $\mathbf{q} = (\frac{1}{2}, \frac{1}{2}, 0)$, quadratic magnetic cell. Two branches of excitations were followed at 4.3 K along the symmetry directions of the crystallographic BZ (figure 2). The contributions of different antiferromagnetic domains could not be separated, although a broader neutron group is observed near $M$, which is the center $M'$, or the corner $M''$ of a magnetic BZ depending on the domain orientation relative to the diffraction plane.

The theoretical treatment is similar to the ferromagnetic case. However 1) the local quantization axis is taken to coincide with the direction of the sublattice magnetization, 2) the dipolar and/or the pseudodipolar terms are taken into account, 3) the inter-ion hamiltonian is divided in two parts relative to interactions between ions on the same or different sublattices (6). Then the energies for transverse spin waves are obtained from:

$$
[1 - J_{\alpha} g^+g^- (\omega)] [1 - J_{\beta} g^+g^- (\omega)] - J_{\gamma}^2 g^+g^- (\omega) g^+g^- (\omega) = 0
$$

where $g^{\alpha\beta}$ are the single ion susceptibilities [4], $J_{\alpha}^\gamma$ and $J_{\beta}^\gamma$ are the Fourier transform of interactions between ions lying on same or different sublattices respectively.

The mean energies of dispersion branches are well reproduced by taking into account the dipolar terms and a term $B_3 = -0.6\,mK$ including magnetoelastic and quadrupolar interactions. Four inter-ion interactions are sufficient to fit the dispersion curves (table I).

4. Discussion. — The range of interactions is rather short for both compounds. Largest interactions occur with first neighbours (sign reversal between ErCu and HoZn) and third neighbours (ferromagnetic coupling). Similar results were obtained by Goebel et al. [7] on La$_{1-x}$Gd$_x$Zn and La$_{1-x}$Gd$_x$Ag from NMR experiments. It is not possible to explain the interactions via the simple Ruderman-Kittel coupling via free s-electrons [6]. Dormann [8] relates the spin reversal of the hyperfine field contribution from first neighbours to the variation of the interatomic distance. Band calculations [9] have shown a large d character near the Fermi level in these compounds. Thus indirect coupling between first neighbours may occur through the direct overlap of d electrons; but the d wave function has a large extent towards the alloyed metal and may lead to the propagation of interactions through the Zn (or Cu) atom.

Acknowledgments. — We thank P. Morin and D. Schmitt for their participation to this work and P. Florés for the technical help during the experiments performed at the Lalie-Langevin Institute.

References