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MAGNETIC EXCITATIONS AND SPECIFIC HEAT FOR INTERMETALLIC COMPOUNDS INVOLVING THULIUM

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Abstract. – A Green’s function theory is developed for studying magnetic excitations and specific heat of thulium intermetallic compounds. Green’s functions and the internal energy expressions appropriate for the cubic Laves structure are given. Numerical results for TmAl₂ are presented.

1. Green’s function formalism

The cubic Laves structure has a magnetic lattice with diamond structure. This structure can be constructed from 2 fcc sublattices A and B, one shifted with respect to the other. We assume nearest-neighbor and next-nearest-neighbor ferromagnetic exchange, so that the Hamiltonian can be written

\[ \mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1 \]

\[ \mathcal{H}_0 = \sum_i \left[ V_A - 2JzMA S^z_{Ai} - 2J'z'M_B S^z_{Ai} \right] + NzJM_A^2 + N'_z J'M_AM_B + (B \text{ terms}) \]

\[ \mathcal{H}_1 = - \sum_{ij}^{AA} J_{ij}^{AA} \left[ \frac{1}{2} (S^+_{Ai}S^-_{Aj} + S^-_{Ai}S^+_{Aj}) + S_{Ai}S_{Aj} \right] \]

\[ - \sum_{ij}^{AB} J_{ij}^{AB} \left[ \frac{1}{2} (S^+_{Ai}S^-_{Bi} + S^-_{Ai}S^+_{Bi}) + S_{Ai}S_{Bi} \right] \]

\[ + (BB \text{ terms}) + (BA \text{ terms}) \]

\[ S_{Ai} = (S^z_{Ai} - \langle S^z_{Ai} \rangle). \]

In these expressions \( J' \) is the nearest-neighbor exchange, \( J \) is the next-nearest-neighbor exchange, \( z' \) is the number of nearest-neighbors, and \( z \) is the number of next-nearest neighbors. \( V_A \) is the crystal-field potential which for TmAl₂ can be written in the form given by Lea, Leask and Wolf [1]. Collective excitations are found from the temperature Green’s functions which are calculated using perturbation theory and the chain-diagram approximation. This approximation is equivalent to the equation of motion RPA theory described by Buyers [2]. Coupled equations involving inter- and intrasublattice Green’s functions are involved. These are decoupled by introducing “acoustical” and “optical” Green’s functions [3]. When the crystal-field has orthorhombic symmetry, or when it has cubic symmetry with axis of magnetization in the (110) direction, then there is a coupling between Green’s functions involving the pairs \((S^+, S^-)\) and \((S^-, S^-)\) and between the pairs \((S^+, S^+)\) and \((S^-, S^+)\). Transverse and longitudinal Green’s functions decouple. For the cubic crystal-field with \( z \)-axis in the (001) direction, used here, the pairs \((S', S+), (S', S-)\) give zero contribution, so that within the chain-diagram approximation, the system can be solved to give

\[ G_{++}^{0/\alpha} (q, \omega) = \frac{g_{++} (\omega)}{1 - g_{++} (\omega) (J (q) \pm |J' (q)|)} \]

and

\[ G_{zz}^{0/\alpha} (q, \omega) = \frac{g_{zz} (\omega)}{1 - g_{zz} (\omega) (J (q) \pm |J' (q)|)} \]

and the non-interacting Green’s functions \( g_{ij} (\omega) \) can be found in the literature [2]. \( J' \) and \( J \) are the Fourier transforms of nearest-neighbor and next-nearest-neighbor exchange. \( J' \) can be a complex number. In these equations the + sign is associated with the acoustical Green’s function and the − sign is associated with the optical Green’s function. Fourier coefficients of acoustical and optical correlation functions can be defined from corresponding Green’s functions by the fluctuation-dissipation theorem [4]

\[ C_{ij}^{(0)} (q, \omega + i\eta) = \int_{-\infty}^{\infty} d\omega [1 - e^{-\beta\omega}]^{-1} \frac{1}{\pi} \text{Im} C_{ij}^{(0)} (q, \omega + i\eta) \]

where the limit \( \eta \) goes to zero is implied. Straightforward algebra allows the thermal average of the Hamiltonian to be expressed in terms of \( C_{ij}^{(0)} (q) \)

\[ \langle \mathcal{H} \rangle = \langle \mathcal{H}_0 \rangle \]

\[ - \sum_q \frac{1}{2} J_q \left( C_{++q}^0 + C_{+q}^0 + C_{-q}^0 + C_{++q}^0 \right) \]

\[ + J_q \left( C_{zzq}^0 + V_{zzq}^0 \right) \]

\[ + \frac{1}{2} \left| J_q' \right| \left( C_{+q}^0 + C_{-q}^0 + 2C_{zzq}^0 \right) \cos 2\phi \]

\[ - \frac{1}{2} \left| J_q' \right| \left( C_{-q}^0 + C_{-q}^0 + 2C_{zzq}^0 \right) \cos 2\phi \]

(8)
The angle \( \phi \) is defined from \( J_4 = J_4^* e^{i\phi} \). The specific heat can be calculated by differentiating this expression with respect to temperature.

2. Discussion

Figure 1 presents the experimental specific heat for TmAl_2 and the theoretical curve in the paramagnetic phase calculated using equation (8). The specific heat of LuAl_2 has been subtracted in order to account for the contribution of phonons and conduction electrons [7]. For this calculation the following parameters were used: \( x = 0.458 \), \( W = 0.399 \) K, \( (J_z + J'_z) = 0.190 \) K and \( \frac{J'}{J} = 10.0 \). \( x \) and \( W \) are Lea, Leask, and Wolf parameters for the cubic crystal-field [1]. The sum of exchange parameters is fixed by the mean-field transition temperature \( T_c = 3.4 \) K. The ratio \( J'/J \) is chosen to be consistent with other intermetallic compounds of the same structure since it is not known by any independent experiment. These crystal-field parameters give a \( \Gamma_8^{(1)} \) ground state with \( \Gamma_1, \Gamma_4, \Gamma_3, \Gamma_6^{(2)} \), and \( \Gamma_2 \) excited states. The crystal-field alone accounts very well for the specific heat in the paramagnetic phase down to about 1.5 \( T_c \) with the chain-diagram approximation accounting for the remainder of the paramagnetic specific heat.

The insert to figure 1 shows the lower-lying branches of dispersion curves for the collective excitations at \( T = 3.6 \) K for two high symmetry directions. The branches at about 3.7 K originate from a crystal-field transition between \( \Gamma_1 \) and \( \Gamma_4 \) states. The branches showing stronger dispersion originate from a crystal-field transition between the \( \Gamma_6^{(1)} \) ground state and the \( \Gamma_4 \) state. There is one more set of branches corresponding to a transition out of the ground state at about 40 K, however its intensity is lower than the 14 K branches by a factor of 3. There is no soft mode, and the thermodynamic properties in the paramagnetic phase just above \( T_c \) are due almost entirely to the \( \omega = 0 \) modes corresponding to transitions within degenerate multiplets.

![Dispersion Curves](image)

Fig. 1. – Magnetic specific heat for TmAl_2. The solid line gives the Green’s function result. The broken line gives the mean-field approximation. The insert shows dispersion curves for TmAl_2 at \( T = 3.6 \) K with \( q \) expressed in units of \( 2\pi/\lambda \) where \( \lambda \) is the lattice constant.