

THERMODYNAMICS OF $CeAl_2$ AT LOW TEMPERATURES:
SPECIFIC HEAT AND SPIN-RELAXATION RATE.¹

O. Huddák

Inst. Physics, Czech Acad. Sci., Praha

J.L. Gavilano, H.R. Ott

Laboratorium für Festkörperphysik, ETH-Hönggerberg, Zürich

Received 31 July 1995, accepted 8 February 1996

Thermodynamic theory recently developed [1] predicts that at least a part of the observed [2] linear dependence in the spin relaxation rate and specific heat of the compound $CeAl_2$ may be explained by existence of new low frequency features in the spin excitation spectrum of the incommensurate magnetic structure at low temperatures. We show here that also the third order term T^3 in the specific heat is expected to be present at low temperatures within the frame of this theory.

Linear dependencies in the specific heat and the spin-lattice relaxation time were recently observed in [2] below T_N outside the critical temperature region in $CeAl_2$. Recently we concluded [1] that at least a part of the observed linear dependence in the spin-lattice relaxation rate and in the specific heat of the compound $CeAl_2$ may be explained by existence of new low frequency modes in the spin excitation spectrum of the incommensurate magnetic structure. This structure seems to be non-chiral spiral. The results of this paper summarize our recent results concerning the incommensurate structure in this compound and extend our calculations of the specific heat temperature dependence to the next lowest order. It is shown here that new type of modes contributes besides the linear contribution also by the cubic term T^3 .

Recent discussions about excitation spectra and response functions expected to occur in incommensurate modulated systems, [3]-[9], were extended to the magnetic system $CeAl_2$ in [2] and in [1]. The imaginary part of the transversal magnetic dynamic susceptibility decreases to zero linearly with frequency even in the case when a strong single ion easy anisotropy axis exists as a consequence of new low energy spin excitations which should be present in studied systems.

$CeAl_2$ belongs to those materials in which frozen-in incommensurate modulated transversal spin-wave was observed to exist as the ground state, [10]. Al nuclear-quadrupole-resonance studies, [2], of $CeAl_2$ at low temperatures reveal a linear temperature dependence of the spin-lattice relaxation rate (T^{-1}) and a large linear term in

¹Presented at 9th Czech and Slovak conference on magnetism, Košice, Slovakia, August 28-30 1995

the specific heat. It was argued in the paper [1] that magnetic low-energy excitations of incommensurate modulated structure in $CeAl_2$ may account for a large fraction of linear behaviour of both quantities. Here we extend our results to the next order term in the specific heat.

We start our description with the simplest exchange Hamiltonian relevant for our problem of description of such two modulated spirals

$$H = H_A + H_B + H_i, \quad (1)$$

where the two spirals, denoted by A and B, are described by the Hamiltonian ($X=A, B$):

$$H_X = - \int_{BZ} \left[\frac{1}{2} J(\mathbf{q}) (S_{X,\mathbf{q}}^+ S_{X,\mathbf{q}}^- + S_{X,\mathbf{q}}^- S_{X,\mathbf{q}}^+) + J(\mathbf{q}) S_{X,\mathbf{q}}^z S_{X,\mathbf{q}}^z \right] d^3q. \quad (2)$$

The interaction between the spiral magnetic moments A and B is probably quadratic one according to [2]. Its explicit form is unknown. However, we assume that new spin variables may be introduced such that they combine the A and B spins. As a result the original interaction energy H_i is expected to be splitted into two local anisotropy energy terms with respect to new spins.

It is convenient to introduce new independent spin variables in the following way:

$$\begin{aligned} \mathbf{m}_i &= \frac{1}{\sqrt{2}} (\mathbf{S}_{A,i} + \mathbf{S}_{B,i}), \\ \mathbf{M}_i &= \frac{1}{\sqrt{2}} (\mathbf{S}_{A,i} - \mathbf{S}_{B,i}). \end{aligned} \quad (3)$$

We assume that resulting anisotropy terms for spins \mathbf{m} and \mathbf{M} have the simplest possible forms:

$$H_i = -D \sum_i \mathbf{m}_i^x \mathbf{m}_i^x, \quad (4)$$

$$H_i = -D \sum_i \mathbf{M}_i^y \mathbf{M}_i^y. \quad (5)$$

The interaction constant D is positive.

The static and dynamic properties of both systems of spins, \mathbf{S}_A and \mathbf{S}_B , are mutually dependent. Using the new spin variables the total hamiltonian (1) now consists of two parts for \mathbf{m} and \mathbf{M} spins, respectively. The part for \mathbf{m} is found from (1) to be in the form:

$$H = - \int_{BZ} \left[\frac{1}{2} J(\mathbf{q}) (m_{\mathbf{q}}^+ m_{\mathbf{q}}^- + m_{\mathbf{q}}^- m_{\mathbf{q}}^+) + K(\mathbf{q}) m_{\mathbf{q}}^z m_{\mathbf{q}}^z \right] d^3q, \quad (6)$$

where

$$K(\mathbf{q}) \equiv J(\mathbf{q}) + D.$$

The part for \mathbf{M} is found from (1) to be in the form:

$$H = - \int_{BZ} \left[\frac{1}{2} J(\mathbf{q}) (M_{\mathbf{q}}^+ M_{\mathbf{q}}^- + M_{\mathbf{q}}^- M_{\mathbf{q}}^+) + K(\mathbf{q}) M_{\mathbf{q}}^y M_{\mathbf{q}}^y \right] d^3q. \quad (7)$$

The $J(\mathbf{q})$ interaction energy is such that together with strong local anisotropy D leads to an amplitude modulated state. Two spins, \mathbf{m} and \mathbf{M} , thus order with the same modulation wavevector.

The NQR and NMR spectra are sensitive as well to the static magnetic structure (lineshape) as to the dynamic properties (spin relaxation time). This latter quantity, $\frac{1}{T_1}$, may be calculated using the definition given in [1] for Al nucleus sites

$$\frac{1}{T_1} = \frac{k_B T}{\hbar N_{\text{site}}} \sum_{\nu} |A_{\nu}(q)|^2 \frac{\text{Im} \chi^{\nu\nu}(q, \omega)}{\omega}, \quad (8)$$

where the resonance frequency $\omega \rightarrow 0$. Here A_{ν} is the Fourier transform of the hyperfine coupling, the sum on ν in (8) is over components of the diagonal ($\nu = x, y$), N_{site} is the number of the lattice sites. We have found an explicit expression for (8) in the limit of low temperatures. It is sufficient in this limit to calculate the susceptibility without taking into account thermally activated processes due to temperature T prefactor in (8).

The spin relaxation rate is thus found to be

$$\begin{aligned} \frac{1}{T_1} &\approx \frac{T}{\kappa}, \\ \frac{1}{\kappa} &\equiv \frac{k_B \hbar m \gamma \omega^2 \hbar I}{D^{*2}}, \\ \eta &\equiv D^{*2} \frac{1}{N_{\text{site}}} \sum_q \frac{\text{Im} \chi^{+-}(q, \omega)}{m \omega}, \end{aligned} \quad (9)$$

Here D^* is an energy constant, [1], and

$$(\hbar \omega \hbar I)^2 \equiv \langle |A_{\nu}(q)|^2 \rangle.$$

For our set of parameters we have found $\eta = 0.518$. Note, that the spin relaxation rate (9) depends linearly on temperature in those incommensurate systems in which the amplitude \mathbf{m} and the modulation wavevector \mathbf{Q} are only weakly temperature dependent. This situation occurs also in the case of $CeAl_2$ compound below the critical temperature T_N but outside the critical region.

The specific heat excess contribution due to low frequency spin excitations behaves in qualitatively different way depending on whether the commensurate or incommensurate modulation is stabilized in the ground state. In the later case we have found within the approximation used in our paper and using general formulas from [6] - [9] that there exists a nonzero q -dependent density of states of low frequency spin excitations, denoted here by $Z(\omega, q)$. In the limit of small frequencies

$$\lim_{\omega \rightarrow 0} Z(\omega, q) = Z_0(q) > 0.$$

In the limit of finite frequencies the specific heat excess, C , due to new kind of spin excitations at low frequencies, is given up to the third order in temperature by

$$C = \gamma T^+ + \delta T^3$$

$$\begin{aligned}
 \gamma &\equiv \frac{k_B R Z_0 H}{D^*} \\
 \delta &\equiv \frac{k_B^3 R Z_2 H'}{D^{*3}} \\
 H &\equiv \int_0^\infty \frac{x^2 \exp(-x)}{(1 - \exp(-x))^2} dx, \\
 H' &\equiv \int_0^\infty \frac{x^4 \exp(-x)}{(1 - \exp(-x))^2} dx.
 \end{aligned}
 \tag{10}$$

R is the usual constant. Analytical form of Z_0 is given in [1]. This specific heat excess is linearly and cubically temperature dependent if the amplitude m and the modulation wavevector Q are again both only weakly temperature dependent. The cubic term has its origin in the second order frequency contribution to the density of states

$$Z(\omega) \approx Z_0 + Z_2 \omega^2 + \dots$$

In future it will be interesting to compare experimentally observed values of γ and δ constants with those predicted by our theory.

References

- [1] O. Hudák (a), J.I. Gavilano, H.R. Ott: *Z. Phys. B: Cond. Matter* (to appear)
- [2] J.I. Gavilano, J. Hunziker, O. Hudák, T. Sileator, F. Hülliger, H.R. Ott: *Phys. Rev. B* **47** (1993) 3438
- [3] H. Liu: *J. Magn. Mater.* **22** (1980) 93
- [4] P.-A. Lindgaard: *J. Magn. Mater.* **31-34** (1983) 603
- [5] T. Ziman, P.-A. Lindgaard: *Phys. Rev.* **33** (1986) 1976
- [6] S.W. Lovesey: *J. Phys. C: Solid State Physics* **21** (1988) 2805
- [7] M.A. Brackstone, S.W. Lovesey: *J. Phys. C: Condens. Matter* **1** (1989) 6793
- [8] Ch. J. Lantwin: *Z. Phys. B: Condens. Matter* **79** (1990) 47
- [9] S.W. Lovesey, G.I. Watson, D.R. Westhead: *Int. J. Mod. Phys.* **5** (1991) 1313
- [10] B. Barbara, D. Gignoux, C. Vittier: *Lectures on Modern Magnetism*, Science Press and Springer-Verlag, Beijing and Heidelberg, 1988, ch. 3
- [11] N. Bulut, D.W. Howe, D.J. Scalapino, N.E. Bickers: *Phys. Rev. B* **41** (1990) 1797