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1997 J. Phys.: Condens. Matter 9 4495

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COMMENT

Comment on ‘Thermodynamic properties and the magnetic neutron scattering cross-section of an atom in a solid’

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Received 25 September 1996, in final form 21 November 1996

Abstract. We point out that the recent paper ‘Thermodynamic properties and the magnetic neutron scattering cross-section of an atom in a solid’ (S W Lovesey 1996, *J. Phys.: Condens. Matter* 8 5915) is based on a sum rule which is reliable only in the high-temperature limit. Indeed low-temperature experimental data cannot be favourably fitted.

A two-level model described by H_0 is chosen [1] to describe an isolated atom and a Weiss molecular-field description of the magnetic environment is provided by the choice

$$V = \lambda_x J_x + \lambda_y J_y + \lambda_z J_z \quad (1)$$

assuming

$$\langle 1|J_\alpha|1\rangle = \langle 2|J_\alpha|2\rangle = 0 \quad \alpha = x, y, z \quad \langle 1|J_\alpha|2\rangle = 0 \quad \alpha = x, y \quad \langle 1|J_z|2\rangle \neq 0 \quad (2)$$

where $|j\rangle$ with $j = 1, 2$ are the two non-degenerate eigenstates of H_0 of energy E_1 and E_2 , respectively.

In section 4 of [1] the author claims that ‘Average values of products of J_α are obtained from the partition function $Z = 2 \exp\{-(E_1 + E_2)/2T\} \cosh(\epsilon/2T)$ by differentiating it the requisite number of times with respect to λ_α which appears in ϵ ’, where $\epsilon = \sqrt{\Delta^2 + |2\langle 1|V|2\rangle|^2}$ with $\Delta = E_1 - E_2$. The above statement is wrong when $[H_0, V] \neq 0$. Using the relationship between the weights attached to elastic and inelastic scattering events and the second derivative of the partition function: $\mu_0^2 + T\chi = g^2(T^2/Z)(\partial^2 Z/\partial \lambda_z^2)$, the author says: From the second derivative of the partition function we find the estimate

$$g^2 \langle J_z^2 \rangle = \mu_0^2 + T\chi \quad (3)$$

Equation (3) is equation (4.1) of [1], referred to by the author as a *sum rule* because on the basis of assumption (2) and of the incorrect relationship between $\langle J_\alpha^2 \rangle$ and the second derivative of the partition function Z one has $\langle J_x^2 \rangle = \langle J_y^2 \rangle = 0$ and $\langle J_z^2 \rangle = J(J + 1)$. All results of [1] spring from equation (3) as confirmed by the author in section 6, where he claims ‘We have demonstrated that a sum rule for the weights, which is an exact statement, permits the calculation of the thermodynamic quantities in terms of Δ and the critical temperature, say.’.

We state that equation (3) is reliable only in the high-temperature limit. The trouble is in the evaluation of $\langle J_\alpha^n \rangle$ (with $n > 1$) as the n th derivative of the partition function since, when $[H_0, V] \neq 0$, as in the present case, this relationship is inconsistent. In particular, for $n = 2$ the correct relationship is

$$\langle J_\alpha^2 \rangle = \frac{1}{\beta^2 Z} \frac{\partial^2 Z}{\partial \lambda_\alpha^2} - \frac{2}{Z} \sum_{n=2}^{\infty} (-1)^n \frac{\beta^n}{(n+2)!} \sum_{l=1}^{n-1} (n-l+1) \text{Tr}(J_\alpha C_{l,\alpha} \mathcal{H}^{n-l}) \quad (4)$$

where $\mathcal{H} = H_0 + V$, $C_{l,\alpha} = [\mathcal{H}^l, J_\alpha]$. We give the proof of equation (4). The second derivative of the partition function

$$Z = \text{Tr} e^{-\beta \mathcal{H}} = \text{Tr} \left[\sum_{n=0}^{\infty} (-1)^n \frac{\beta^n}{n!} \mathcal{H}^n \right] = \sum_{n=0}^{\infty} (-1)^n \frac{\beta^n}{n!} \text{Tr}(\mathcal{H}^n) \quad (5)$$

with respect to λ_α reads

$$\frac{\partial^2 Z}{\partial \lambda_\alpha^2} = \sum_{n=2}^{\infty} (-1)^n \frac{\beta^n}{n!} \text{Tr} \left(\frac{d^2 \mathcal{H}^n}{d \lambda_\alpha^2} \right) \quad (6)$$

where

$$\frac{d^2 \mathcal{H}^n}{d \lambda_\alpha^2} = \sum_{l=1}^{n-1} \sum_{m=0}^{l-1} \mathcal{H}^m J_\alpha \mathcal{H}^{l-1-m} J_\alpha \mathcal{H}^{n-1-l} + \sum_{l=0}^{n-2} \sum_{m=0}^{n-2-l} \mathcal{H}^l J_\alpha \mathcal{H}^m J_\alpha \mathcal{H}^{n-2-l-m}. \quad (7)$$

The cyclic property of the trace allows us to write

$$\begin{aligned} \frac{\partial^2 Z}{\partial \lambda_\alpha^2} &= \sum_{n=2}^{\infty} (-1)^n \frac{\beta^n}{n!} \left[\sum_{l=1}^{n-1} \sum_{m=0}^{l-1} \text{Tr}(J_\alpha \mathcal{H}^{l-1-m} J_\alpha \mathcal{H}^{n-1-l+m}) + \sum_{l=0}^{n-2} \sum_{m=0}^{n-2-l} \text{Tr}(J_\alpha \mathcal{H}^m J_\alpha \mathcal{H}^{n-2-m}) \right] \\ &= \sum_{n=2}^{\infty} (-1)^n \frac{\beta^n}{n!} 2 \sum_{l=0}^{n-2} (n-l-1) \text{Tr}(J_\alpha \mathcal{H}^l J_\alpha \mathcal{H}^{n-2-l}). \end{aligned} \quad (8)$$

From the identity $\text{Tr}(J_\alpha C_{l,\alpha})=0$ it follows

$$\begin{aligned} \frac{\partial^2 Z}{\partial \lambda_\alpha^2} &= \sum_{n=2}^{\infty} (-1)^n \frac{\beta^n}{(n-2)!} \text{Tr}(J_\alpha^2 \mathcal{H}^{n-2}) + 2 \sum_{n=4}^{\infty} (-1)^n \frac{\beta^n}{n!} \sum_{l=1}^{n-3} (n-1-l) \text{Tr}(J_\alpha C_{l,\alpha} \mathcal{H}^{n-2-l}) \\ &= \beta^2 \left[\text{Tr}(J_\alpha^2 e^{-\beta \mathcal{H}}) + 2 \sum_{n=2}^{\infty} (-1)^n \frac{\beta^n}{(n+2)!} \sum_{l=1}^{n-1} (n-l+1) \text{Tr}(J_\alpha C_{l,\alpha} \mathcal{H}^{n-l}) \right]. \end{aligned} \quad (9)$$

Dividing by $\beta^2 Z$ equation (4) is obtained.

Note that the corrections with respect to equation (4.1) are negligible in the high-temperature limit, but are dramatic in the low-temperature limit of interest for the author to study actual compounds [2,3]. The expectation for the value of the gap at zero temperature $\epsilon(0)$ obtained from the *sum rule* (4.1) of [1] is

$$\frac{\Delta}{\epsilon(0)} = \sqrt{1 - \frac{\tanh x}{x}} \quad (10)$$

with $x = \Delta/2T_c$, T_c being the temperature at which $\epsilon = \Delta$.

As for the compound studied in [2] the author expects $\epsilon(0) = 1.64$ meV, whereas the expectation of the mean field approximation used in [2] is $\epsilon(0) = 0.65$ meV. Unfortunately any comparison with experiment is lacking. However, for PrFe_2Ge_2 and PrFe_2Si_2 studied in [3] measurements of the gap are available both in the paramagnetic region (Δ) and in the ordered region ($\epsilon(0)$). For PrFe_2Ge_2 the experimental value of $\Delta = 0.8$ meV and the critical temperature $T_N = 14.2\text{K}$ [3] lead to $\epsilon(0) = 4.3$ meV using equation (10), whereas the experimental value is $\epsilon(0) = 2.7$ meV [3]. As for PrFe_2Si_2 equation (10) gives $\epsilon(0) = 3.5$ meV taking the experimental $\Delta = 2.4$ meV and the critical temperature $T_N = 7.7$ K [3]. The experiment gives $\epsilon(0) = 2.8$ meV [3].

References

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