Second-Neighbor Exchange Integral in EuS†

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Experimental measurements of the specific heat of EuS in the temperature region 1–4°K have been analyzed to determine the first- and second-neighbor exchange integrals. The results are: $J_1 = (1.48\pm0.16)\times10^{-5}$ eV; $J_2 = (-0.11\pm0.28)\times10^{-5}$ eV.

IN two recent papers, McGuire *et al.* have discussed the ferromagnetic properties of some divalent europium compounds.[1,2] In the second of these papers, they present some interesting speculations concerning the exchange integrals in these materials. In order to account for the probable antiferromagnetism of EuTe (whereas other compounds such as EuO, EuS, and EuSe are ferromagnetic), they suppose that the exchange integral connecting nearest-neighbor Eu^{++} ions is positive, but that the second-neighbor exchange is negative. They propose that the magnitude of the first-neighbor interaction decreases on going from EuO through the series to EuTe, but that the magnitude of the second-neighbor interaction remains constant or increases slightly. In this note, we analyze the information concerning the exchange interaction between second neighbors in EuS which can be obtained from the results of specific-heat measurements on this material.[3] Our conclusion is that the second-neighbor exchange is, in magnitude, less than 25% of the first-neighbor exchange, and may have the opposite sign.

In reference 3, a measurement of the specific heat of EuS was reported, and analyzed theoretically with respect to the series obtained by Dyson[4] for a Heisenberg ferromagnet. Dyson showed that at low temperatures, the specific heat of spin waves may be expressed as:

$$C/R = \alpha T^{3/2} + \beta T^{5/2} + \gamma T^{7/2} + \delta T^4 + \cdots. \tag{1}$$

The first three terms of this series are obtained for a system of noninteracting spin waves; the term involving T^4 is a correction for the interaction of spin waves with each other. This interaction term is relatively small compared to the others if the spin on each site is large (it is 7/2 in EuS) and may be neglected at reasonably low temperatures. We may, therefore, consider the spin-wave system to be one of noninteracting bosons in the temperature range from 1 to 4°K to which the measurements pertain.

It was found in that work that an excellent empirical

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[1] T. R. McGuire, B. E. Argyle, M. W. Shafer, and J. S. Smart, Appl. Phys. Letters **1**, 17 (1962).

[2] T. R. McGuire, B. E. Argyle, M. W. Shafer, and J. S. Smart (to be published).

[3] D. C. McCollum and J. Callaway, Phys. Rev. Letters **9**, 377 (1962). The specific heat EuS in the temperature range 1–4°K is dominated by spin waves. The lattice contribution is negligible.

[4] F. J. Dyson, Phys. Rev. **102**, 1230 (1956).

fit to the experimental specific-heat data could be obtained with $\alpha = 2.82\times10^{-2}$; $\beta = 6.65\times10^{-3}$; and all higher coefficients zero. If one assumes, in conformity with the usual form of the Heisenberg theory, that the exchange interaction connects only nearest-neighbor magnetic ions, this quantity can be determined to have the value 1.23×10^{-5} eV. Then the formula given by Dyson for β yields $\beta = 5.2\times10^{-3}$.

This result is in sufficiently good agreement with the experimentally determined β to given one confidence that spin-wave theory does apply to EuS, but the discrepancy is definitely outside the experimental error and would be particularly noticeable in the higher portion of the temperature region considered. Subsequently, McGuire *et al.*[2] proposed that the second-neighbor exchange integral might not be negligible, and we decided to attempt a more complete analysis of the specific-heat data to see if some limits could be placed on this quantity. It should be noted that such an analysis has frequently been considered in connection with the specific heats of crystal lattices; however, in that case the complexity of lattice vibration spectra renders inversion of the specific-heat data impractical. The present analysis is made possible by the fact that the spin-wave spectrum in EuS is much simpler than the phonon spectrum.

Regardless of the nature of the exchange interaction responsible for ferromagnetic alignment in the europium compounds, the spin-wave spectrum must have the full symmetry of the crystal. This means that the spin-wave dispersion relation $E(q)$ may be expanded in a Fourier series,

$$E(\mathbf{q}) = E_0 - 2S \sum_n J_n e^{i\mathbf{q}\cdot\mathbf{R}_n}. \tag{2}$$

The R_n are direct lattice vectors; the J_n are coefficients which are referred to as exchange integrals, although no commitment to the Heisenberg theory of ferromagnetism is implied; and S is the spin of an ion. The J_n will depend only on the magnitude of the R_n. In the absence of an external magnetic field, we may set

$$E_0 = 2S \sum_n J_n.$$

In order to calculate the specific heat of a system of noninteracting spin waves with $E(q)$ given by Eq. (2), it is necessary to perform the integral:

$$C = \frac{\Omega K}{(2\pi)^3} \int \left(\frac{E}{KT}\right)^2 \frac{e^{E/KT}}{(e^{E/KT}-1)^2} d^3q \tag{3}$$

in which Ω is the volume of a unit cell, K is Boltzmann's constant, and T is the absolute temperature.

The terms of the Dyson series for the specific heat, Eq. (1) which pertain to noninteracting spin waves may be found in the following way: The density of states, $n(E)$ is introduced in order to convert Eq. (3) into an integral over energy,

$$C = \Omega K \int \left(\frac{E}{KT}\right)^2 \frac{e^{E/KT}}{(e^{E/KT}-1)^2} n(E) dE. \qquad (4)$$

Then the density of states is expanded as a power series in the energy:

$$n(E) = \sum_m b_m E^{m+1/2}. \qquad (5)$$

This expression is inserted in (4), and the integration is performed with the upper limit made infinite. One obtains a set of relations connecting the effective exchange integrals with the coefficients α, β, and γ, of Eq. (1). As examples, we give the first two such relations for a face-centered cubic lattice with first- and second-neighbor interactions only.

$$J_1 + J_2 = \frac{K}{2S}\left(\frac{c_0}{4\alpha}\right)^{2/3}, \quad J_1 + 2J_2 = \frac{K}{2S}\beta c_1\left(\frac{c_0}{4\alpha}\right)^{7/3}, \quad (6)$$
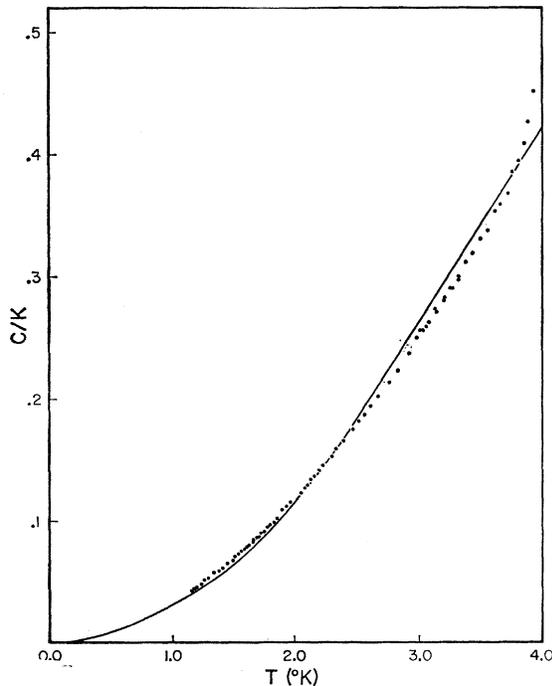

FIG. 1. The specific heat of EuS is shown as a function of temperature. The smooth curve was computed from Eq. (3) with $J_1 = 1.47 \times 10^{-5}$ eV; $J_2 = -0.10 \times 10^{-5}$ eV. The points represent the experimental data of reference 3.

where c_0 and c_1 are numerical constants given by

$$c_0 = \frac{15\zeta(5/2)}{32\pi^{3/2}}; \quad c_1 = \frac{4096\pi^{3/2}}{105\zeta(7/2)}. \qquad (7)$$

Evidently, J_1 and J_2 could be determined if α and β were known, and knowledge of γ would likewise enable a determination of J_3, etc.

There is, however, a serious difficulty in this procedure: If the temperature is high enough for the second and higher terms of (1) to be important, it is quite likely that corrections resulting from the finite volume of the Brillouin zone will also be significant. In this case, the upper limit of integration in (4) with $n(E)$ given by (5) is finite, and terms of the form $e^{-\text{const}/T}$ appear in C. Moreover, the series (5) converges only for energies less than that of the first critical point,[5] so that under these circumstances, use of the expansion of $n(E)$ is not justified.

These considerations are important in the analysis of experimental results concerning the specific heat of EuS even in the temperature range of liquid helium. We have, therefore, adopted the following procedure. First and second neighbors only are considered in Eq. (2). Trial values are assumed for J_1 and J_2 in this equation, and the integral (3) is performed numerically on an electronic computer for a number of temperatures, using the exact Brillouin zone. The resulting specific heat is then compared with experiment, and the work repeated until a best fit, in the sense of least squares is obtained. No approximations other than the neglect of the interactions of spin waves with each other, and the restriction to first and second neighbors are made.

The following results for the exchange integrals were obtained:

$$J_1 = (1.48 \pm 0.16) \times 10^{-5} \text{ eV},$$

$$J_2 = (-0.11 \pm 0.28) \times 10^{-5} \text{ eV}.$$

The calculated specific heat is shown as a function of temperature in Fig. 1, where it is compared with the experimental results.

The second-neighbor exchange is seen to be considerably smaller than the first-neighbor exchange, and quite possibly negative, in general agreement with the suggestion of McGuire et al.[2] It is, in fact, too small in magnitude to be determined with precision from the available experimental data. Additional experiments, including the temperature dependence of the spontaneous magnetization might be of considerable assistance in determining these quantities more precisely.

[5] J. Callaway and A. J. Hughes, Phys. Rev. 128, 134 (1962).