SPIN-WAVE CONTRIBUTION TO THE SPECIFIC HEAT OF EuS†

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Ferromagnetism has recently been reported in EuS and in other insulating europium salts.[1-3] EuS has the NaCl structure, the europium ion being in an S state with $S=\frac{7}{2}$. The nature of the ferromagnetic exchange coupling has not been clear since direct exchange through overlap of the $4f$ electron wave functions is unlikely to be important. A possible indirect exchange coupling mechanism has been proposed.[4] Specific heat measurements on EuS were undertaken in order that the results might be compared with the predictions of spin-wave theory.

About 6 grams of EuS powder were obtained from R. L. Wild and R. D. Archer, University of California, Riverside. The powder was compressed around a copper wire (to be clamped between the jaws of a mechanical thermal switch to the helium bath) to which a carbon resistance thermometer was attached with a small amount of tin. A manganin wire resistance heater was wound around the specimen. Two procedures were used in obtaining the specific heat data. During two runs, measured amounts of heat were supplied to the specimen and the temperatures before and after the heating periods were determined. During another run heat was supplied at a constant rate, and the average time rate of change of thermometer resistance was measured over a series of small intervals and used to calculate the corresponding time rate of change of temperature over each interval. No significant difference was observed in the final results from the two methods. The light weight and high specific heat of the specimen made the temperature drift during heater-off periods quite small, in most cases negligible. The heat capacity of the addenda, always less than 1% of the total, was calculated and subtracted from the total observed heat capacity.

A rough estimate of the Debye θ for EuS can be made if it is assumed that EuS, which is unstable at high temperatures, would have about the same melting temperature as other rare earth sulfides. The result is $\theta \approx 200$ degrees, which indicates that it is reasonable to assume a T^3 dependence for the lattice contribution to the specific heat below 4°K. Figure 1 shows a plot of $C/T^{3/2}$ versus $T^{3/2}$, which would yield a straight line if the specific heat of EuS could be represented by the sum of a lattice term AT^3 and a spin-wave term $BT^{3/2}$.[5] The curvature cannot be ignored in the present case. A least-squares fit to the experimental data was made using a polynomial

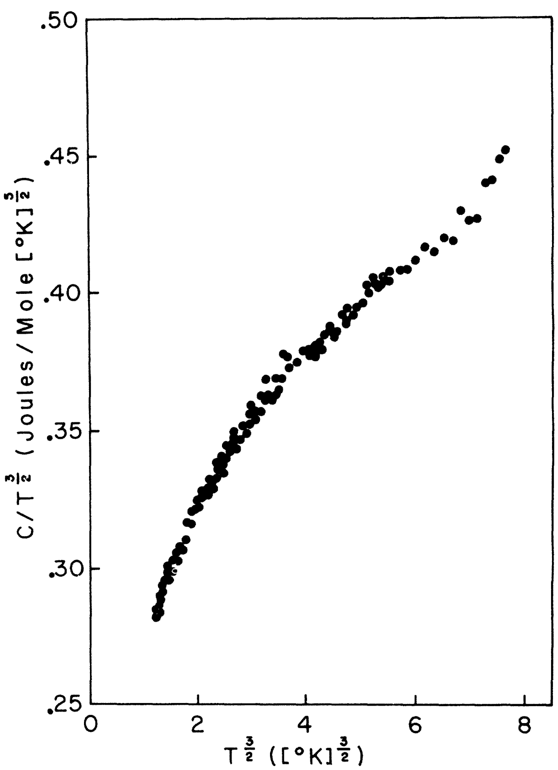

FIG. 1. $C/T^{3/2}$ versus $T^{3/2}$ for EuS.

$$C/R = AT^3 + BT^{3/2} + DT^{5/2}.$$

The coefficients were determined to be $A \approx 0$, $B = 2.82\times10^{-2}$, $D = 6.65\times10^{-3}$, the root-mean-square deviation of C/R being 3.5×10^{-3}. Figure 2 shows the experimental points; the curve represents the above equation for C/R. If the Debye θ is about 200 degrees, the lattice contribution would be less than one-half of one percent of the total specific heat below 4°K.

A value for the exchange integral J can be obtained by equating the coefficients of the $T^{3/2}$ terms from experiment and from spin-wave theory as given by, say, Van Kranendonk and Van Vleck.[5] The result is $J = 1.23\times10^{-5}$ eV. The Curie temperature is 17 ± 1°K;[2,6] thus $kT_C/J \approx 119$. This may be compared with a value computed from the work of Brown and Luttinger[7] concerning the de-

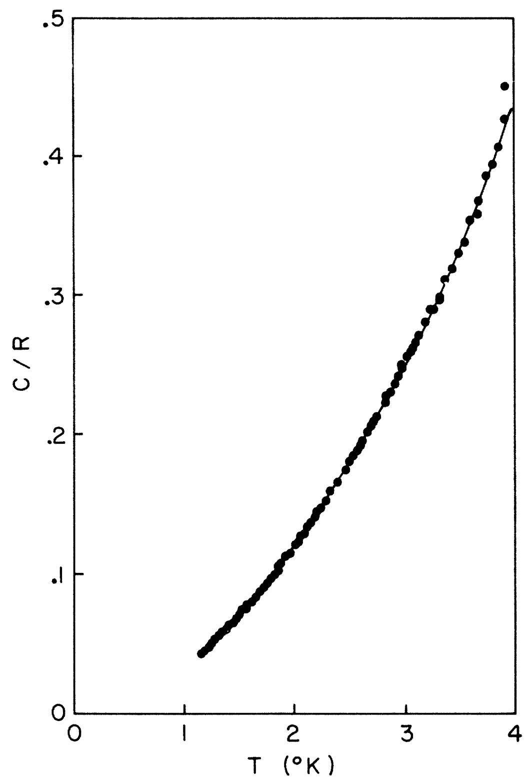

FIG. 2. C/R versus T for EuS. The curve is represented by $C/R = 2.82\times10^{-2}T^{3/2} + 6.65\times10^{-3}T^{5/2}$.

termination of the Curie temperature of a Heisenberg ferromagnet. The result obtained from their work using the Kramers-Opechowski method is $kT_C/J = 107.9$ for a fcc lattice with $S = \frac{7}{2}$. The agreement is reasonably good and is much better than is found in metallic systems. By comparison, Brockhouse[8] has determined J for cobalt from neutron scattering experiments. His value leads to $kT_C/J = 7.6$, whereas Brown and Luttinger give $kT_C/J = 13.0$ for a fcc lattice with $S = 1$.

Dyson[9] and Tahir-Kheli and ter Haar[10] have given expressions for the coefficient of the $T^{5/2}$ term in a spin-wave specific heat, showing that the ratio of the coefficient of the $T^{5/2}$ term to the coefficient of the $T^{3/2}$ term is inversely proportional to the Curie temperature. If the exchange integral parameter is evaluated by equating coefficients of $T^{3/2}$ terms from experiment and from theory, the theoretical value of the coefficient of the $T^{5/2}$ term for EuS can be calculated to be 5.2×10^{-3}.

The agreement between theory and experiment is impressive, both in the determination of the $T^{5/2}$ term in the specific heat and in the relation between the exchange integral and the Curie temperature. One concludes that the spin system in EuS can be described at least phenomenologically by a simple Heisenberg spin Hamiltonian. This conclusion is particularly significant since the fundamental assumptions used in deriving the spin Hamiltonian are probably not justified.

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