October 1971

BNL 16247

CONF-711108--10

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EXCHANGE INTERACTIONS IN EuO AND EuS*

L. Passell
Brookhaven National Laboratory, Upton, N. Y. 11973

O. W. Dietrich† and J. Als-Nielsen
Danish Atomic Energy Commission, Research Establishment Risø
Roskilde, Denmark

ABSTRACT

We have used inelastic neutron scattering techniques to measure the spin-wave dispersion curves within the first Brillouin zone of polycrystalline EuO and EuS. Least squares fitted values of the n.n. and n.n.n. exchange constants J_1 and J_2 are in fair agreement with previously reported results in the case of EuS. In EuO, however, we find J_2 to be ferromagnetic in agreement with a recent estimate by Kasuya but in contradiction to the previously measured antiferromagnetic n.n.n. coupling.

The divalent europium chalcogenides EuO and EuS are among the few known examples of simple Heisenberg ferromagnets and as such they offer unique opportunities for studying the mechanism of the exchange interaction between localized spins. Kasuya[1] has recently considered a variety of exchange processes which are thought to occur in these materials and has attempted to make numerical estimates of the exchange constants J_1 and J_2 which characterize respectively the interactions between nearest and next-nearest neighbor magnetic ions.

Meaningful evaluation of Kasuya's calculations, however, presupposes well-determined values of J_1 and J_2. The presently accepted values of the exchange constants in EuO and EuS come from two types of experiments: measurements of the magnetic contribution to the heat capacity[2,3] and measurements of the magnetization at low temperatures using NMR techniques.[4,5] In both methods the values of J_1 and J_2 result from fitting low temperature spin-wave theory to the measurements and as is usual with such procedures, there are questions of sensitivity and possibilities for systematic errors which are difficult to evaluate. Kasuya's estimates as well as the recent magnetization measurements of Menyuk, Dwight and Reed[6] have raised doubts concerning the value of the exchange constant J_2 in EuO which experiments[2,4] indicate is antiferromagnetic. Both the theoretical estimate and an extrapolation from measurements on other europium chalcogenides based on the magnitudes of the interatomic distances, however, suggest a ferromagnetic next-nearest neighbor coupling.[1]

*Work supported by U. S. Atomic Energy Commission.

†Guest scientist Brookhaven National Laboratory

We have found that by using thin slab samples of polycrystalline EuO and EuS prepared with separated Eu^{153} (to minimize neutron absorption) we can observe the scattering of neutrons by spin waves in both materials.[7] From these experiments we are able to determine the form of the "polycrystalline averaged" spin-wave dispersion curves within the entire first Brillouin zone. An analysis of these curves, which will be described below, gives us directly the values of J_1 and J_2.

By assuming only exchange interactions the spin-wave energies are expressed in the form[8]

$$\hbar\omega(\vec{q}) = 2S\ [\ J(0) - J(\vec{q})\ ]. \qquad (1)$$

Here $J(\vec{q}) = \sum_{\vec{R}} J(\vec{R}) \exp(i\vec{q}\cdot\vec{R}) = J_1 \sum_{\vec{R}_1} \exp(i\vec{q}\cdot\vec{R}_1) + J_2 \sum_{\vec{R}_2} \exp(i\vec{q}\cdot\vec{R}_2)$,

where we have included interactions only between nearest (R_1) and next-nearest neighbors (R_2). In both materials the Eu^{2+} ions form a f.c.c. lattice for which Eq. (1) reduces to the expression

$$\hbar\omega(\vec{q}) = 2S\ [\ 12J_1 + 6J_2 - J_1 \sum_{\vec{R}_1} \cos(\vec{q}\cdot\vec{R}_1) - J_2 \sum_{\vec{R}_2} \cos(\vec{q}\cdot\vec{R}_2)\ ]. \qquad (2)$$

Since we used polycrystalline samples, all directions of $\vec{q}$ are equally probable. Hence, for a given $|\vec{q}|$, we observe a distribution of spin-wave energies, i.e. a "line shape" which can be computed by averaging Eq. (2) over all directions of $\vec{q}$.

We specifically considered the contribution of the dipolar forces to the spin-wave energies as outlined in Ref. 8. At small values of $\vec{q}$ the dipolar contributions are significant in certain directions, but averaging over all directions made the dipolar contributions negligible within the q-range covered by the measurements.

We also considered the influence of a small magnetic anisotropy. Following an analysis by Lovesey[9], we concluded that the observed anisotropies in EuO[10] and EuS[11] had no significant effect on the resulting exchange constants.

The measurements were performed with a triple-axis spectrometer operated in the constant q mode using an incoming neutron energy of 13 meV. The energies corresponding to the peaks of the observed line shapes at each q were least squares fitted to the peak positions of the polycrystalline average of Eq. (2) using J_1 and J_2 as adjustable parameters. Figures 1 and 2 show the results. The open circles are the observed positions of the peak intensity; the calculated positions appear as the solid lines. We have also included the dispersion curves along a few representative symmetry directions computed using the best fitting values of J_1 and J_2. The inserts show the

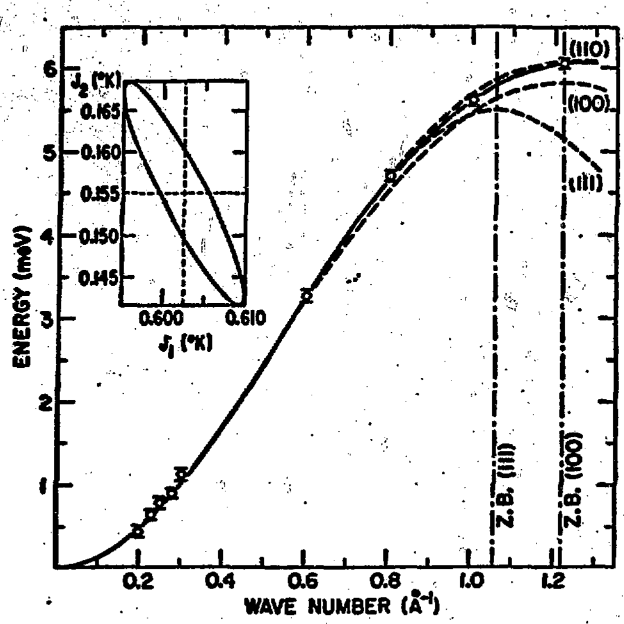

Figure 1. Spin-wave dispersion in polycrystalline EuO.

covariance ellipses for the two parameters resulting from the least squares fitting.[12] It is obvious from the fits that third-neighbor interactions are negligible in both EuO and EuS.

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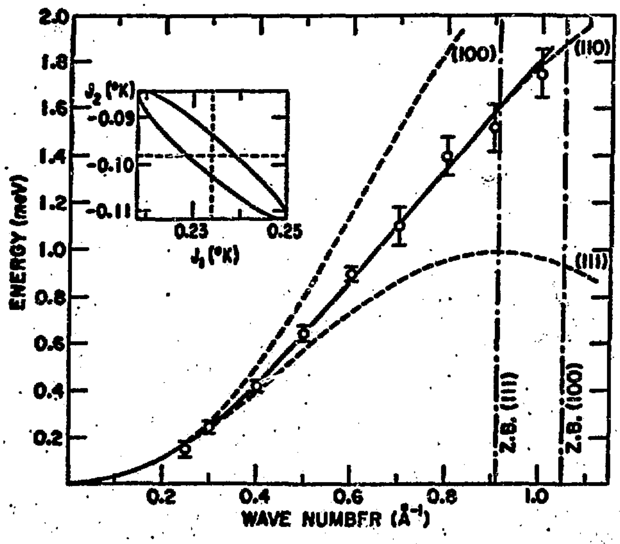

Figure 2. Spin-wave dispersion in polycrystalline EuS.

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Table I

Exchange Constants in °K

	EuO		EuS	
	J_1	J_2	J_1	J_2
This exp.	0.602±0.008	0.155±0.014	0.234±0.016	-0.098±0.014
Sp. Ht.	0.76±0.02[2]	-0.084±0.015[2]	0.172±0.019[3]	-0.013±0.032[3]
NMR	0.75±0.0025[4]	-0.0975±0.004[4]	0.20 ±0.01[5]	-0.08 ∓0.02[5]

Listed in Table I are the values of J_1 and J_2 obtained from these measurements together with the values reported from the heat capacity and the NMR magnetization measurements. For EuS, the agreement is almost within the limits of the errors. The present measurements confirm that J_2 is negative not only by virtue of the fits to the positions of peak intensity but also because the observed line shapes are broad. It is evident in Figure 2 that this is to be expected in polycrystalline material particularly near the zone boundary because exchange interactions of opposite sign give a large zone-boundary anisotropy of the spin-wave energies.

In EuO, however, our results are not in agreement with those obtained previously. In particular, we find both J_1 and J_2 to be positive as expected by Kasuya.[1] The conclusion that J_2 is positive is reinforced by the fact that the line shapes in EuO are observed to be significantly narrower than those in EuS reflecting the fact that the spin-wave dispersion is more isotropic when both exchange constants are of the same sign. This is evident in Figure 1.

We wish to express our appreciation to Dr. S. W. Lovesey for calculating for us the contribution of the magnetocrystalline anisotropy to the spin-wave energy.

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12. The statistical probability that the true values of J_1 and J_2 are within the covariance ellipse is $1-\exp(-0.5) = 0.39$ and the probability that they are within the outer rectangle is 0.68.