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► **To cite this version:**

P. Schwob, G. Everett. EFFECTIVE EXCHANGE INTEGRAL ($J_1 + J_2$) IN EuS FROM STANDING SPIN WAVE RESONANCE. *Journal de Physique Colloques*, 1971, 32 (C1), pp.C1-1066-C1-1067. 10.1051/jphyscol:19711383 . jpa-00214422

HAL Id: jpa-00214422

<https://hal.science/jpa-00214422>

Submitted on 4 Feb 2008

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EFFECTIVE EXCHANGE INTEGRAL ($J_1 + J_2$) IN EuS FROM STANDING SPIN WAVE RESONANCE (*)

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Résumé. — La méthode de la résonance des ondes de spin stationnaires a été appliquée à des couches minces de EuS à 1,1 °K dans la configuration perpendiculaire. L'énergie des ondes de spin est proportionnelle au carré du vecteur d'onde k dans le régime observé ($k < 0,2 \pi/a$). L'intégrale effective d'échange ($J_1 + J_2$) est déterminée à $0,088 \pm 0,010$ °K. Cette valeur est inférieure à celle de Passenheim et collaborateurs [7] déduite de la chaleur spécifique. L'origine de cette différence est attribuée à des effets de corrélation, analogues à ceux calculés par Silbergliitt et collaborateurs [8] pour CrBr₃.

Abstract. — The standing spin wave resonance has been measured in thin films of EuS at 1.1 °K in the perpendicular configuration. The magnon dispersion relation is found to be quadratic in the observed region of k ($k < 0.2 \pi/a$). The effective exchange energy ($J_1 + J_2$) determined is 0.088 ± 0.010 °K. This value is lower than that calculated by Passenheim et al. [7] from specific heat measurements. This discrepancy is attributed to correlation effects, analogous to those calculated by Silbergliitt et al [8] for CrBr₃.

The excitation spectrum of a Heisenberg ferromagnet can be derived from its Hamiltonian :

$$H_{ij} = - g\beta H \sum_j S_j - 2 \sum_{i>j} J_{ij} S_i S_j \quad (1)$$

where β is the Bohr magneton and $- 2 J_{ij}$ is the exchange interaction between the spins S_i and S_j at the lattice sites i and j . By applying the Holstein-Primakoff transformation to (1) and restricting J_{ij} to nearest and next nearest neighbors (J_1 and J_2 resp.) the single particle dispersion relation for magnons (spin waves) is calculated :

$$\hbar\omega_k = g\beta H + 2 S J_1 \sum_{\mathbf{l}_1} [1 - \exp(i\mathbf{k}\mathbf{l}_1)] + 2 S J_2 \sum_{\mathbf{l}_2} [1 - \exp(i\mathbf{k}\mathbf{l}_2)] \quad (2)$$

where \mathbf{l}_1 and \mathbf{l}_2 denote the set of nearest and next nearest neighbors [1]. In the long wave length approximation, $\mathbf{k}\mathbf{l} \ll 1$, (2) reduces for a face centered cubic lattice to

$$\hbar\omega_k = 2 S(J_1 + J_2) a^2 k^2 + g\beta H \quad (3)$$

where a is the lattice parameter.

The experimental determination of the parameters J_1 and J_2 is of primary importance for an understanding of the nature of the exchange coupling. Macroscopic experiments such as measurements of the specific heat or of the magnetization as a function of temperature provide us with an averaged value of the exchange integrals over a certain temperature range, assuming the validity of the dispersion relation (2) over the entire Brillouin zone. It is possible only from a microscopic experiment to verify the detailed form of the dispersion relation and to provide values for the exchange interaction. Such magnon spectroscopy can be done by either inelastic neutron scattering or by photon absorption. The latter experiment, the standing spin wave resonance, was first observed by

Seavy and Tannenwald in thin films of permalloy [2].

Kittel [3] has shown theoretically that a homogeneous rf field in an insulating ferromagnetic film can excite those spin wave modes which have nodes at the film surface and having a net transverse magnetization different from zero. This means that the film thickness L is equal to an odd multiple of half wavelengths of the spin wave. Thus, absorption of rf power should be observed at values of the magnetic field H_k which are given by the following equation :

$$(H_{up} - H_k)^{1/2} = (2(J_1 + J_2) S/g\beta)^{1/2} \cdot a\pi/L \cdot n \quad (4)$$

where n is an odd number and we have made use of the long wavelength approximation (3). H_{up} is the uniform precession field (ferromagnetic resonance) and corresponds to $k = 0$.

We have used the method of standing spin wave resonance to investigate the dispersion relation for low k values and to determine the effective exchange interaction in the fcc insulating ferromagnetic compound EuS. Dispersion relations near the center of the Brillouin zone cannot be determined precisely by neutron diffraction and in addition the europium compounds are not well suited for neutron diffraction investigations due to the high nuclear absorption. Vacuum distilled EuS was evaporated onto fused quartz substrates heated to about 500 °C in a vacuum of 3 to $5 \cdot 10^{-6}$ torr. The thickness of the films was determined by weight, by optical interferometry or with an oscillating quartz crystal thickness monitor to an accuracy of 10 % or better. The absorption of rf power at a frequency of 23.63 GHz and at 1.1 °K was measured in a static magnetic field perpendicular and parallel to the film plane. From the ferromagnetic resonance fields we determined the saturation magnetization of the films to be $4 \pi M = 15.11$ kG which is very close to the theoretical value of 15.30 kG. This indicates that the films are equal in quality to the best bulk specimens reported to the present. In the perpendicular configuration the additional absorption peaks due to the excitation of standing spin waves

(*) Work supported in part by the U. S. Atomic Energy Commission, AEC Report No. UCR 34 P77-27.

could be well resolved, a typical example is shown in figure 1. In figure 2 we have plotted the experimental values of $(H_{up} - H_k)^{1/2}$ for two films as a function of the mode index n . It is seen that the quadratic dispersion relation is very well obeyed for all spin waves observed.

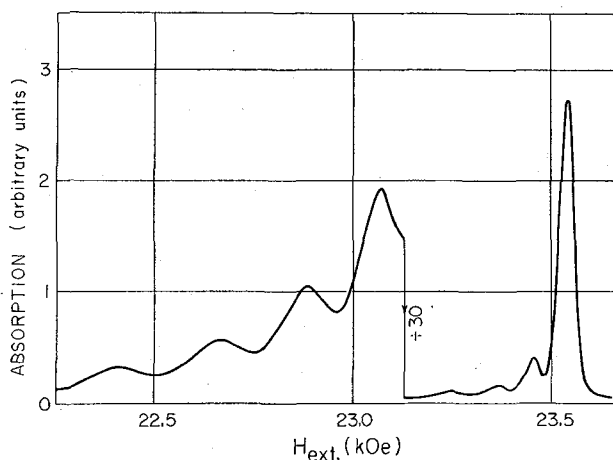


FIG. 1. — Microwave absorption versus external magnetic field of an EuS film (624 Å) at 1.1 °K.

Soohoo [4] has pointed out that the simple relationship $k = n\pi/L$ is changed if the pinning of the surface spins is incomplete, i. e. if the spin wave amplitude is not completely zero at the surface. Under this condition one would observe an apparent deviation from the quadratic dispersion relation. Incomplete pinning is probably indicated in figure 2 for the thicker film because the straight line intersects the abscissa with a non zero value. For the higher k values, however, this effect seems to be completely negligible and does not affect the determination of the slope. The origin of this surprisingly strong pinning lies probably in the strong change of the crystalline electric field at the surface and also in the magnetic dipolar energy. The latter has been shown by Benson et al. [5] to cause pinning for the case of very thin films.

Sparks [6] has recently emphasized that the choice of the mode indices is dependent on the spatial inhomogeneities of the film magnetization. Judging from the high saturation magnetization of our samples we conclude that this effect is very small. In addition we want to point out that we have been able to resolve the $n = 1$ mode in the spectrum of the 258 Å film, in

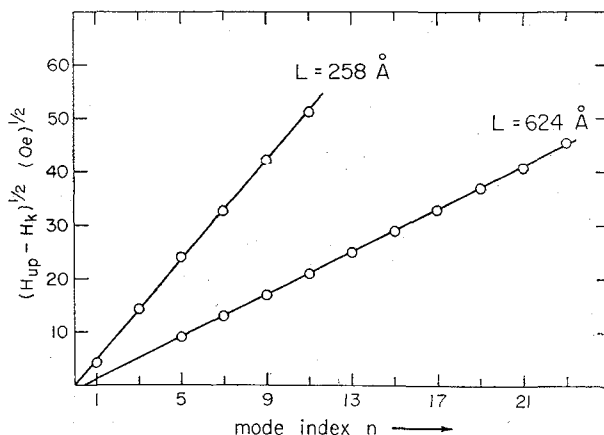


FIG. 2. — Square root of the experimental spin wave energy as a function of the mode index for two EuS films at 1.1 °K.

agreement with Spark's conclusion that the first observed peak should be labeled $n = 1$.

From the slope of the straight lines shown in figure 2 and from the film thicknesses we calculate the value of $(J_1 + J_2)/k_B$ to be 0.080 °K (258 Å) and 0.084 (624 Å). Together with the results of the two other films [0.098 °K (302 Å) and 0.089 °K (529 Å)] we determine an average of

$$(J_1 + J_2)/k_B = 0.088 \pm 0.010 \text{ °K.}$$

This number has to be compared with the effective exchange integral determined by other methods. The most recent specific heat measurements carried out by Passenheim et al. [7] in the same temperature region and in magnetic fields up to 10 kG, resulted in the values $J_1 = +0.20$, $J_2 = -0.06$ °K and therefore $(J_1 + J_2) = 0.14 \pm 0.02$ °K. This number agrees roughly with our result but the discrepancy is larger than the combined experimental uncertainties. Lattice strains, which are undoubtedly present in the films, can also be ruled out as the origin of the disagreement, because they would contribute differently for films of different thicknesses, whereas our results are essentially the same for all films. Our present conclusion therefore, is that the actual magnon dispersion relation differs from the form given by equation (2) at high k values. This conclusion is strongly supported by recent theoretical and experimental work by Silberglitt et al. [8] on the spin wave spectrum of CrBr_3 . These authors showed that correlation effects increase considerably the spin wave energies at high k values. Exchange integrals determined with eq. (2) from the specific heat would, therefore, be too high.

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