OPTICAL OBSERVATION OF MAGNETIC-FIELD-INDUCED SPIN ALIGNMENT IN ANTIFERROMAGNETIC EuTe†

J. Feinleib
Lincoln Laboratory, Massachusetts Institute of Technology, Lexington, Massachusetts 02173

and

C. R. Pidgeon
Francis Bitter National Magnet Laboratory,* Massachusetts Institute of Technology, Cambridge, Massachusetts 02139

(Received 18 September 1969)

The effects of spin alignment induced by a large external magnetic field have been observed in the optical reflectivity spectra of antiferromagnetic EuTe at 1.5 K. By comparison with the reflectivity spectra of ferromagnetic EuO, EuS, and EuSe, we conclude that we have observed the transition from an antiferromagnetic superlattice band splitting to the ferromagnetic exchange splitting that occurs with complete spin alignment at $H \sim 80$ kOe.

A striking change in the reflectivity of antiferromagnetic EuTe has been observed as a function of magnetic field, giving direct evidence for the first time of the changeover of the energy-band structure of an antiferromagnetic material, with superlattice band splittings, to the spin-polarized band structure of the ferromagnetic europium chalcogenides (EuO, EuS, and EuSe). The latter materials exhibit a characteristic polarization-dependent sharp triplet structure in the E_1 reflectivity peak in a small "domain-orienting" magnetic field at temperatures well below the Curie point.[1,2] At zero and very low fields EuTe exhibits a broad polarization-independent triplet structure of the E_1 peak. At high fields, as the sublattice spins cant up towards the field direction, this changes to a sharp polarization-dependent triplet structure whose components grow in strength with increasing field up to a saturation field ~80 kOe. At this point the triplet structure is identical in form to that of EuO, EuS, and EuSe, showing that complete parallel spin alignment has occurred. The central peak for the high- and low-field triplets is shifted in energy by ~0.2 eV. We conclude, as discussed below, that the low-field structure is characteristic of the effects of antiferromagnetic spin ordering and the high-field structure of ferromagnetic spin ordering. Such a direct observation of the change in band structure for the two cases has not previously been reported, although it has long been predicted theoretically.[3]

The experiments were performed on single crystals of EuTe grown from the melt.[1,2] Near-normal-incidence reflectivity was measured from cleaved (100) faces, oriented with the magnetic field perpendicular to the face in the Faraday configuration. Left and right circularly polarized light was used over the spectral range 2.0 to 3.0 eV. The results of these measurements at 1.5°K are shown in Fig. 1. This figure shows the reflectivity structure, above the 2.2-eV absorption edge, of the first prominent peak E_1 as a function of magnetic field up to 100 kOe. The E_1 peak is seen to have a polarization-independent triplet structure at zero field. As the field increases up to 40 kOe, this structure broadens, and near 60 kOe there is a change to an entirely different type of polarization-dependent triplet structure. The lowest energy peak of this triplet has a pure σ_R polarization while the highest energy peak is pure σ_L. The central

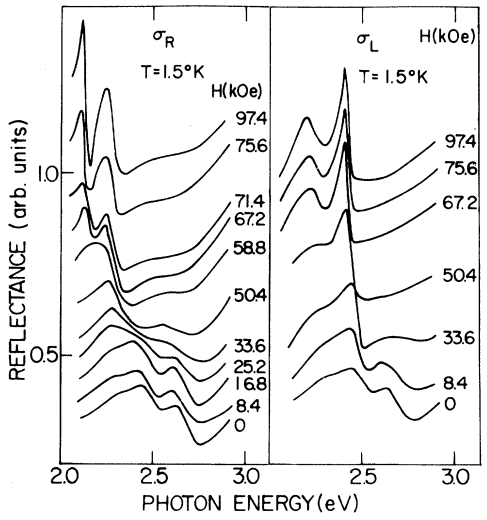

FIG. 1. Reflectivity of E_1 peak of EuTe at 1.5°K as a function of external magnetic field for σ_R and σ_L polarization. The reflectance scales apply to the 97.4-kOe curves, the others having been shifted for clarity.

peak is composed of both polarizations although it has a greater σ_R component.

The peaks of this triplet structure grow in magnitude but have only a slight shift with photon energy as the magnetic field is increased above 40 kOe. As the field is increased from 80 kOe up to the highest fields, no further changes are observed in the positions or magnitudes of the E_1 peaks. Below the onset of the sharp triplet structure, the peaks of the doublet structure are observed to have mixed σ_L and σ_R composition, and have a small red shift with field. When compared with the polarized reflectivity measurements of EuO, EuS, and EuSe, it is evident that the E_1 structure of EuTe at high fields has identical form to that observed in the ferromagnets (in the ferromagnetic case a small external field was necessary to orient the domains). The central peak in EuTe at 1.5 K is at the highest energy for this series: EuO (1.25), EuS (1.9), EuSe (2.1), and EuTe (2.2 eV), and follows the same sequence as the absorption edges.[4] The splittings of the side peaks from the central one are in the neighborhood of 0.25 eV for the first three materials, and 0.15 eV for EuTe. We do not consider the E_2 peak here since it is clearly more complex (as discussed elsewhere for the ferromagnetic case[5]), but it again shows a saturation effect at about 80 kOe. The data on the E_1 peak are summarized in Fig. 2, where the peak positions as a function of magnetic field are also plotted. Representative reflectivity spectra are plotted on the same scale for comparison.

EuTe has been determined to be antiferromagnetic below 8°K with a spin structure of the second kind (like MnO).[6,7] The magnetic ordering in the series of europium chalcogenides changes with increasing anion atomic weight and lattice constant[8]: EuO (5.15 Å, $T_C = 69.4$ K), EuS (5.96 Å, $T_C = 16.5$ K), EuSe (6.19 Å, $T_N = 4.6$ K), and EuTe (6.60 Å, $T_N = 7.8$ K). From the Heisenberg model and the molecular-field approximation, McGuire and Shafer[9] were able to explain the Curie and Néel temperatures, T_C and T_N, in terms of two competing exchange interactions, J_1 and J_2, between nearest-neighbor and next-nearest-neighbor europium ions, respectively. On this model J_1 is found to be positive and decreases rapidly with increasing lattice constant, whereas J_2 is negative and remains approximately constant. J_1 dominates at the EuO end of the series, but J_1 and J_2 become comparable for EuSe, and J_2 dominates for EuTe. In fact, for ferromagnetic EuO the total exchange energy is

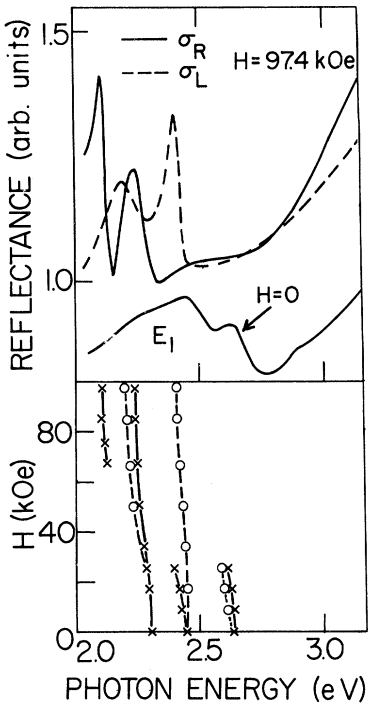

FIG. 2. Polarized reflectivity of EuTe for $H = 97.4$ kOe and $H = 0$ at 1.5°K. The $H = 0$ curve has been shifted for clarity. Also shown are the positions of the peaks in the E_1 multiplet as a function of magnetic field.

$E_{tot} \propto 12J_1 + 6J_2$, whereas for EuTe it depends only on J_2:

$$E_{tot} \propto -6J_2. \quad (1)$$

Thus, EuSe is ferromagnetic below 2.8 K, becomes antiferromagnetic with complex spin-structure changes up to about 5 K, and can be made ferromagnetic at these temperatures with a field ~8 kOe applied.[9] By contrast, EuTe is antiferromagnetic at 1.5 K even with fields ~50 kOe applied.[8,9]

We have previously shown that the E_1 structure change in EuO, EuS, and EuSe with magnetic ordering is caused by the ferromagnetic exchange splitting of the $5d(t_{2g})$ final state, and that the transition involved is $4f^7(^8S_{7/2}) - 4f^6(^7F_J)5d(t_{2g})$.[1,2] The ferromagnetic exchange splits the t_{2g} state into spin-up and spin-down components which are spin mixed by spin-orbit interaction. For antiferromagnetic EuTe, the final $5d(t_{2g})$ state is also split by exchange interaction, but the splitting is more complex.[3] In this case, the band is split by the alternating-spin superlattice of the f electrons on the Eu^{2+} ion. By comparison with the nonmagnetic state of the crystal, the antiferromagnetic state has additional splittings of

the energy bands because of the symmetry change of the crystal when the superlattice of the spin structure is taken into account. Physically this corresponds to the energy splitting associated with the two spin configurations of the conduction-band wave functions for $\vec{k}$ close to the new zone boundary. However, there is no exchange splitting at $\vec{k}=0$ for this case (by contrast to the ferromagnetic case where the exchange splitting extends approximately uniformly over the entire zone), since the two spin configurations of the conduction-band wave functions are associated with the same energy here. In the lower-energy configuration, the spin wave functions alternate so as to be aligned with the f electrons on each site; in the higher energy one, the spin wave functions are oppositely aligned. Since each band component has up- and down-spin parts, we do not expect a polarization dependence of transitions. This is in accordance with E_1 structure we observe at very low fields. By a similar argument to that for the ferromagnetic case,[1,2] we associate the triplet observed here with the antiferromganetic superlattice splitting at the new zone boundary. The magnitudes of the splittings observed in the low-field (antiparallel) and high-field (parallel) limits, ~0.15 eV, are approximately the same because the physical potential causing the splitting in the two cases arises from the same exchange interaction.

In addition to the basic antiferromagnetic superlattice band splitting, there is superimposed a net ferromagnetic interaction induced by the magnetic field, making the spectra more complex. This comes about above the spin-flop field, when the sublattice magnetizations are perpendicular to the magnetic field and then slowly cant up toward the magnetic-field direction. The spin-flop field in EuTe has been determined to be about 4 kOe.[9] This low value results from the cubic symmetry of the NaCl structure. If we make the approximation of zero anisotropy field, then we may estimate the applied saturation field H_S required for spin alignment from the following considerations. The energy density in a simple molecular-field model[10] is

$$E=(6J_1+6J_1\cos 2\theta+6J_2\cos 2\theta)(7/2)^2 -(7/2)g\beta H\cos\theta, \quad (2)$$

where 2θ is the angle between the sublattice spin directions and $(7/2)g\beta$ is the magnetic moment of each ion. Putting $\partial E/\partial\theta=0$ for the configuration of minimum energy, and then setting $\theta=0$ for the condition of complete spin alignment, we have the field required for saturation

$$g\beta H_S=84(J_1+J_2). \quad (3)$$

Taking J_1 and J_2 from Ref. 9, we obtain the estimate $H_S\sim 70$ kOe. For this simple model, this is in reasonable agreement with our high-field magneto-optical data, where the amplitudes and positions of the E_1 peaks saturate at $H\sim 80$ kOe.

In conclusion we should point out that it has been suggested that exciton effects[11] may be important in the $4f$ to $5d$ transitions, and presumably it is possible to have exciton states at different parts of the zone. However, if this is the case, it is still true that the wave functions associated with these states will be determined partly by the narrow d band (i.e., partially localized d state) in the model proposed here, so that the effects observed are still a direct manifestation of the transition from ferromagnetic exchange splitting to antiferromagnetic superlattice splitting of the d band. We may note here that the exchange splitting of the E_1 structure, which is 0.15 eV, is much larger than the Zeeman energy corresponding to an external magnetic field of 100 kOe. The effect of the intraionic exchange, therefore, is to enhance greatly the magneto-optical effects observed in this material. Thus, the technique can readily be used for the direct observation of the effects of magnetic ordering on band structure, as demonstrated here; in particular, we have uncovered an effect, amenable to simple experimental observation, which has long been proposed by theorists but never before observed directly.

The crystals of EuTe were grown by T. B. Reed and R. E. Fahey, to whom we are extremely grateful. We wish to thank D. Adler, J. Dimmock, K. Dwight, S. Foner, A. Misetich, and W. Scouler for helpful discussions.

---

†The Lincoln Laboratory portion of this work was sponsored by the Department of Air Force.

*Supported by the U. S. Air Force Office of Scientific Research.

[1]J. Feinleib, W. J. Scouler, J. O. Dimmock, J. Hanus, T. B. Reed, and C. R. Pidgeon, Phys. Rev. Letters 22, 1385 (1969).

[2]C. R. Pidgeon, J. Feinleib, W. J. Scouler, J. Hanus, J. O. Dimmock, and T. B. Reed, to be published; W. J. Scouler, J. Feinleib, T. B. Reed, and C. R. Pidgeon, in Proceedings of the Fifteenth Annual Conference on Magnetism and Magnetic Materials, Philadelphia, Pa., 1969 (to be published).

[3]J. C. Slater, Phys. Rev. 82, 538 (1951); D. Adler,

Phys. Rev. Letters 17, 139 (1966).
[4]S. Methfessel and D. C. Mattis, in Handbuch der Physik (Springer-Verlag, Berlin, Germany, 1968), Vol. XVIII, Pt. 1, p. 389.
[5]W. J. Scouler, J. Feinleib, J. O. Dimmock, and C. R. Pidgeon, to be published.
[6]G. Busch, P. Junod, M. Risi, and O. Vogt, in Proceedings of the International Conference on the Physics of Semiconductors, Exeter, England, 1962 (The Physical Society, London, England, 1962), p. 727.
[7]T. R. McGuire, B. E. Argyle, M. W. Shafer, and J. S. Smart, Appl. Phys. Letters 1, 17 (1962).
[8]G. Busch, J. Appl. Phys. 38, 1386 (1967).
[9]T. R. McGuire and M. W. Shafer, J. Appl. Phys. 35, 984 (1964).
[10]See, for example, C. Kittel, in Introduction to Solid State Physics (John Wiley & Sons, Inc., New York, 1966), 3rd ed., p. 484.
[11]T. Kasuya and A. Yanase, Rev. Mod. Phys. 40, 684 (1968).

MAGNONS AT LOW AND HIGH TEMPERATURES IN THE PLANAR ANTIFERROMAGNET K_2NiF_4

J. Skalyo, Jr., and G. Shirane
Brookhaven National Laboratory,* Upton, New York 11973

and

R. J. Birgeneau† and H. J. Guggenheim
Bell Telephone Laboratories, Murray Hill, New Jersey
(Received 4 November 1969)

The spin-wave spectrum of K_2NiF_4 has been measured at 5.0°K and found to be extremely anisotropic with no measurable dispersion in the direction perpendicular to the NiF_2 planes. A study of the temperature dependence of the two-dimensional magnons shows that even relatively long-wavelength spin waves ($\lambda \approx 110$Å) show little renormalization or lifetime effects up to $\approx 1.1 T_N$.

The study by Birgeneau, Guggenheim, and Shirane[1] of the quasielastic scattering of neutrons on the group of compounds typified by K_2NiF_4 has recently provided definitive proof of their two-dimensional character. In particular, long-range two-dimensional correlations were observed to exist over a wide range of temperatures above the phase transition temperature ($T_N = 97.1$°K in K_2NiF_4). From the form of quasielastic scattering above and below T_N the investigators concluded that in K_2NiF_4 the phase transition at 97.1°K is two-dimensional in nature, the three-dimensional aspects being essentially redundant. More recent quasielastic measurements[2] show that above T_N the diffusive scattering is associated entirely with the z-z component of the wave-vector–dependent susceptibility $\chi^{\alpha\alpha}(\vec{q})$, thus indicating that even in the paramagnetic regime the anisotropy is playing a crucial role. In order to understand the nature of the phase transition it is necessary to have quantitative information about both the isotropy of the Hamiltonian and the relative size of intra- and interplanar interactions; these may be determined through a study of the magnons at low temperatures. In addition, considerable insight into the spin dynamics may be obtained by studying the thermal evolution of the magnons around T_N; it might be anticipated that this could be extremely unusual in a two-dimensional near-Heisenberg system. We have therefore undertaken a detailed study of the magnons in K_2NiF_4. In this Letter we report the results of this study at low temperatures (5°K) together with some preliminary measurements of the magnons around T_N.

From the two-dimensional character of K_2NiF_4, a highly anisotropic spin-wave spectrum is expected with measurements in the c direction giving an indication of interplanar exchange. The magnetic structure[3] is shown in Fig. 1 where the planar behavior is evident in that the NiF_2 planes are separated from one another by two KF planes. Furthermore, within the molecular-field approximation, there is no net coupling between nn planes in the Néel state. This structure results in two domains as the body-centered spin in Fig. 1 can be up or down.

Because of the existence of two domains, measurements were by necessity made simultaneously in both the [100] and [010] zones. The spin-wave spectrum along $[\zeta, 0, 0]$ and $[0, \zeta, 0]$ are the same if only intraplanar exchange exists. The spin-wave energy for intraplanar magnons has been obtained using the expression given by Keffer[4] for a uniaxial two sublattice antiferromagnet; for $[\zeta, 0, 0]$ the energies are

$$E(q_x) = \{[g\mu_B H_A - 4J_1 + 4(J_2 + 2J_3)\sin^2\tfrac{1}{2}q_x a]^2 - 16J_1^2\cos^2\tfrac{1}{2}q_x a\}^{1/2}. \quad (1)$$