tive value for E_0; to obtain this result they seriously underestimate the kinetic energy. Thus they obtain a single-particle wave function which is too "wide" (this can be seen from their small value of A), and this is the reason why they obtain too large a value for J.

In conclusion, we wish to point out that it is quite possible for this approach to yield a much better value of E_0 than we have reported here. We have only used an analytic form for $f(r)$, although it is quite feasible to derive and solve a differential equation for f. In this way one might obtain a 10% improvement in both the kinetic and potential energies. Since E_0 is an order of magnitude smaller than either of these, a significant improvement is possible.

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EVIDENCE FOR FIELD-DEPENDENT ZERO-POINT SPIN-WAVE EFFECTS IN ANTIFERROMAGNETS*

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In this note we report the observation and analysis of nonlinear behavior in the low-temperature magnetic field dependence of the transverse magnetization, and corresponding magnetic field dependence of the perpendicular differential susceptibility, of antiferromagnetic EuTe and $CoCl_2$. We feel that these effects arise primarily from the zero-point spin-wave contribution to the free energy in accord with the theory of Kanamori and Yosida.[1]

A method in common use for the estimation of exchange parameters[2] utilizes the initial perpendicular susceptibility and presumes it to be a constant up to the saturation field, in accord with molecular field theory. However, a nonlinearity in the magnetization curve reduces the accuracy of this method; e.g., such a procedure overestimates the saturation field by about 10% in the case of EuTe. A zero-point correction to the initial susceptibility, in the evaluation of exchange parameters, has occasionally been considered but its magnitude is not precisely known.[3]

The magnetization[4] and the differential susceptibility[5] were examined to 200 kOe. EuTe powders were used because the anisotropy field [presumably in the (111) layer planes[6] of the fcc structure] is extremely low, i.e., about 200 Oe.[7] Above 4 kOe, all crystallites are magnetizing in the transverse mode. Monocrystals of $CoCl_2$ were measured with the field perpendicular to the c axis. Here, the transverse mode is obtained above 2 kOe.[8]

In Fig. 1 we present the magnetization and differential susceptibility for a EuTe powder sphere at 2.1°K. The magnetization curve is similar to one published by Busch et al.[9] during our study. Of principal interest is the upward concavity from the dashed line drawn to fit the

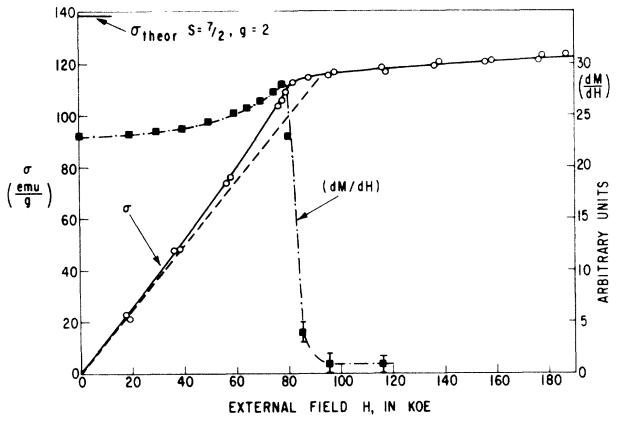

FIG. 1. Magnetization, σ, in emu/g and differential susceptibility, dM/dH, in arbitrary units vs external field for a EuTe powder sphere at 2.1°K.

lower field points. This also appears in the earlier data[9] without mention thereof. The curve of dM/dH reveals an increase of 22% from its initial value. The saturation field, H_S, determined from the peak in dM/dH, is 75 kOe (internal). For comparison we replot dM/dH in Fig. 2(a), normalized to its initial value (above the spin flop[7]), with a field scale normalized to H_S. In contrast to the discontinuity expected, the transition spread of 7 kOe cannot arise from the presumably dominant dipolar anisotropy because the dipolar energy of the saturation configuration is isotropic. We ascribe it to saturation field inhomogeneities caused by stoichiometry deviations (e.g., some Eu^{3+}), evidenced by the low saturation moment and the recently recognized sensitivity of the exchange to trivalent impurities.[10]

In Fig. 2(b) we show the normalized dM/dH curve for a monocrystal slab of $CoCl_2$ at 4.2°K. The saturation field is 34 kOe (internal). This better defined sample exhibits a sharper transition with a 1.5-kOe spread.

In the magnetic structure[6] of EuTe one must consider the nearest-neighbor (nn) exchange interactions between the corner and face-centered ions, and the antiferromagnetic next-nearest-neighbor (nnn) exchange interaction between the ions at the cube corners. Empirical analyses[6,7,11] indicate that the nn exchange is small compared to the nnn exchange interaction. This nn exchange, as well as the anisotropy, is neglected in our theoretical analysis.

The spin-wave calculation proceeds in a manner directly analogous to that by Kanamori and Yosida.[1] The spin-wave excitation frequencies are

$$\hbar\omega_{\vec{k}}^{\pm} = 2JSz\{[1 \pm \gamma_{\vec{k}}][1 \pm \gamma_{\vec{k}}(2H^2/H_S^2 - 1)]\}^{1/2}. \quad (1)$$

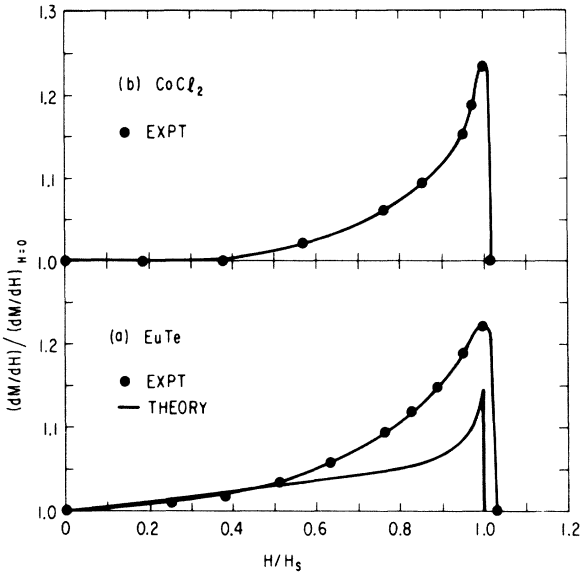

FIG. 2. Normalized differential susceptibility curves $(dM/dH)/(dM/dH)_{H=0}$ vs normalized field H/H_S where H_S, the saturation field, is determined from the peak observed in (dM/dH): (a) EuTe, experiment and theory; (b) $CoCl_2$, experiment.

Here J is the nnn exchange interaction, z is the number of nnn, $\gamma_{\vec{k}} = z^{-1}\sum_{\delta}\exp(i\vec{k}\cdot\vec{\delta})$, where the sum is over nnn, H is the applied magnetic field, and $H_S = 4SJz/g\mu_B$. At low temperature we neglect the spin-entropy contribution to the free energy. Therefore,

$$F = -NJzS(S+1) - N\mu_B gSH^2/2H_S + \tfrac{1}{2}\hbar\sum_{\vec{k}}(\omega_{\vec{k}}^{+} + \omega_{\vec{k}}^{-}). \quad (2)$$

The first two terms in Eq. (2) can be obtained in the molecular field theory, while the last term corresponds to the spin-wave modifications in the form of a zero-point energy. It is this term that produces a nonlinear magnetization and a nonconstant differential susceptibility.

An intuitive description of the effect may be obtained by realizing that the initial transverse configuration of the spins characterizes a non-Néel ground state of the antiferromagnet which manifests itself in a zero-point reduction of the sublattice magnetization. The sublattices rotate with increasing field until a ferromagnetic state is obtained at saturation. The ferromagnetic state does not suffer from a zero-point reduction of the magnetization. Thus, the moment in the direction of the field does not vary linearly with the sine of the rotated angle, but also must contain

a gradual suppression of the zero-point reduction as the ferromagnetic state is achieved.

The calculation of this effect depends upon the reliability of the spin-wave method in giving accurate values for the zero-point magnetization reduction. The magnetization and differential susceptibility are the negative of the first and second derivatives, respectively, of the free energy with respect to the magnetic field. The calculation of the zero-point contributions entails the evaluation of integrals over the first Brillouin zone appropriate to the magnetic sublattice. These have been done numerically with a GE-225 computer. The first Brillouin zone appropriate to the *ABCA* stacking sequence of the (111) layer planes is an oblique hexagonal prism tipped at an angle of $\theta = \arcsin\frac{1}{3}$ from the vertical with $c = \pi\sqrt{3}/a_0$ and $a = 4\pi\sqrt{2}/3a_0$. Here a_0 is the cubic lattice spacing.

The results of the calculation of the susceptibility for EuTe, normalized to the value at zero field, are indicated by the theoretical curve in Fig. 2(a). We have made use of the empirical value[6] of the nnn exchange constant, $J = 0.124°K$. We see that the theoretical and experimental curves are in accord for low values of the relative field but tend to deviate somewhat in the higher regions, although maintaining a similar shape. The experimental curve for $CoCl_2$ bears a stronger similarity in shape to the theoretical curve although, of course, the magnitudes differ. The calculation pertinent to $CoCl_2$ will be presented elsewhere, with other analyses of $CoCl_2$ experiments.

Although the imperfectly defined chemical and structural state of the EuTe sample precludes a refined analysis, the discrepancy between theory and experiment cannot be ignored. Another source of nonlinear transverse magnetization is effective biquadratic exchange arising from the strain dependence of the exchange energy.[12-14] Following Kittel,[12] the magnetization field relation is

$$h = m(1+2\alpha) - 4\alpha m^3. \tag{3}$$

Here h is the field normalized to the saturation field in the absence of the biquadratic term, m is the relative net magnetization, and α is the ratio of biquadratic to bilinear exchange terms times S^2. The normalized differential susceptibility is

$$\frac{(dm/dh)}{(dm/dh)_{m=0}} = \left[1 - \frac{12\alpha m^2}{1+2\alpha}\right]^{-1}. \tag{4}$$

We have examined the discrepancy between theory and experiment for EuTe in Fig. 2(a) in terms of the exchange striction analysis,[14] invoking only nnn exchange. The equilibrium lattice contraction so derived is in good agreement with recent x-ray measurements by Rodbell, Osika, and Lawrence.[15] The value of α required is, however, about an order of magnitude smaller than that employed[6] to describe the temperature dependence of the sublattice magnetization.

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