

tions, at least in the system GdNi<sub>2</sub>.

Our results on the cubic Laves-phase compounds GdPt<sub>2</sub> and GdRh<sub>2</sub> are similar to those of GdNi<sub>2</sub> although the anomalies in  $\rho(T)$  and  $d\rho/dT$  near  $T_c$  are not as striking as in the case of GdNi<sub>2</sub>. We obtain values for  $T_c$  of about 23°K in GdPt<sub>2</sub> and 70°K in GdRh<sub>2</sub> compared with 36 and 74°K, respectively, obtained by Crangle and Ross.<sup>7</sup>

In summary, our experiment gives the first clearly quantitative agreement with the molecular-field description of the spin-disorder scattering near the critical point, and we conclude that this agreement illustrates the validity of such a theory in a system such as GdNi<sub>2</sub> where there is clearly a well-localized Gd moment. We believe that the difference of the critical behavior in these compounds from that in Ni<sup>4</sup> or PdFe alloys<sup>8</sup> probably is due to the fact that, in these gadolinium compounds, the magnetic  $f$  electrons are well localized. Therefore, a local "s-d" exchange interaction, a feature common to the theories discussed earlier,<sup>1,2,5</sup> is a better representation of the true situation than in the case of Ni or PdFe alloys where the  $d$  electrons are not well localized. One is led to make a distinction<sup>9</sup> between the statistical treatment of the problem and the model interaction used. It is quite possible that the major source of the qualitative difference between theory and experiment lies not so much with the details of the sta-

tistical treatment of the critical spin fluctuations as with the model interaction used to describe the scattering of the conduction electrons.

There is clearly a need for further theoretical study of this point.

We would like to thank J. H. Wernick of Bell Telephone Laboratories for providing the samples used in this work and J. Krupp for assistance in the calculations for Fig. 2.

---

<sup>†</sup>Work supported in part by the National Science Foundation and the Research Corporation.

<sup>1</sup>P. G. de Gennes and J. Friedel, *J. Phys. Chem. Solids* **4**, 71 (1958).

<sup>2</sup>D. J. Kim, *Progr. Theoret. Phys.* **31**, 921 (1964).

<sup>3</sup>L. S. Ornstein and F. Zernicke, *Proc. Acad. Sci. Amsterdam* **17**, 793 (1914).

<sup>4</sup>P. P. Craig, W. I. Goldburg, T. A. Kitchens, and J. I. Budnick, *Phys. Rev. Letters* **19**, 1334 (1967).

<sup>5</sup>M. E. Fisher and J. S. Langer, *Phys. Rev. Letters* **20**, 665 (1968).

<sup>6</sup>E. A. Skrabek and W. E. Wallace, *J. Appl. Phys.* **34**, 1356 (1963).

<sup>7</sup>J. Crangle and J. W. Ross, in *Proceedings of the International Conference on Magnetism, Nottingham, England, 1964* (The Institute of Physics and The Physical Society, London, England, 1965), p. 240.

<sup>8</sup>J. A. Mydosh, J. I. Budnick, M. P. Kawatra, and S. Skalski, *Phys. Rev. Letters* **21**, 1346 (1968); M. P. Kawatra, S. Skalski, J. A. Mydosh, and J. I. Budnick, to be published.

<sup>9</sup>The importance of this distinction was pointed out to us by D. J. Kim.

## TWO-MAGNON SCATTERING OF NEUTRONS

R. A. Cowley, W. J. L. Buyers, and P. Martel

Atomic Energy of Canada Limited, Chalk River Nuclear Laboratories, Chalk River, Ontario

and

R. W. H. Stevenson

Department of Natural Philosophy, Aberdeen University, Aberdeen, Scotland

(Received 26 May 1969)

We report the experimental observation of the inelastic scattering of slow neutrons by two-magnon processes and present theoretical calculations which account for the shape and wave-vector dependence of the spectrum.

Two-magnon processes in antiferromagnets have recently attracted considerable interest not only in infrared absorption<sup>1</sup> and Raman scattering<sup>2,3</sup> experiments, but also in theoretical studies. Elliott et al.<sup>4,5</sup> have shown that the shape of the two-magnon spectrum observed in these optical experiments is influenced by magnon-magnon interactions, and Fleury<sup>6</sup> has strikingly confirmed their predictions in RbMnF<sub>3</sub>. In this Letter we report the observation of neutron inelastic scattering by pairs of magnons in the antiferromagnet cobalt fluoride. Neutron scattering differs from optical experiments because the wave-vector dependence of the scattering may be observed, and because two-magnon scattering arises from the correlation between pairs of spins rather than from the four-spin correlations sug-

gested by Elliott *et al.*<sup>4,5</sup> for optical experiments. The nature of the interaction between the neutrons and the magnons is also better understood than that between the photons and the magnons. The experiments were performed on cobalt fluoride as it might be expected to show strong magnon-magnon interactions and to exhibit bound two-magnon states because of its narrow spin-wave bandwidth.<sup>7</sup>

The neutron-scattering cross section  $S(\vec{Q}, \omega)$  for two-magnon processes has been evaluated within the noninteracting spin-wave approximation. For a spin-only antiferromagnet the scattering arises from fluctuations in the  $z$  components of pairs of spins and, at low temperatures, has been obtained by an extension of the work of Nagai and Yoshimori<sup>6</sup> as

$$S(\vec{Q}, \omega) = (g\mu_B)^2 |f(\vec{Q})|^2 [(Q_x^2 + Q_y^2) / |\vec{Q}|^2] \sum_{ij} |\sinh(\theta_i \mp \theta_j)|^2 \delta(\omega - \omega_i - \omega_j) \Delta(\vec{Q} - \vec{q}_i - \vec{q}_j), \quad (1)$$

where  $\vec{Q}$  and  $\omega$  are the wave vector and frequency transfer in the experiment. The form factor of the magnetic ions is  $f(\vec{Q})$ , while  $g\mu_B$  is the product of the gyromagnetic ratio of the neutron and the Bohr magneton. The summation over  $i$  and  $j$  is over all the one-magnon states described by frequency  $\omega_i$  and wave vector  $\vec{q}_i$ , while  $\theta_i$  is the phase factor which enters into the expression for the spin-wave variables in terms of the single-sublattice excitation operators.<sup>8</sup> When  $\vec{Q} - \vec{q}_i - \vec{q}_j$  is equal to a reciprocal-lattice vector  $(h, k, l)$  for which  $h+k+l$  is even, the minus sign in the function  $\sinh(\theta_i \mp \theta_j)$  is taken, and if  $h+k+l$  is odd, the plus sign applies. The two-magnon scattering is therefore most intense in zones containing magnetic reciprocal-lattice vectors.

The experiment was performed with the single crystal of  $\text{CoF}_2$  mounted in a similar manner to that used in our earlier experiments.<sup>9</sup> The neutron scattering was measured with a triple-axis crystal spectrometer controlled so as to obtain the intensity of scattered neutrons as a function of frequency transfer for a fixed predetermined momentum transfer.<sup>10</sup> Some typical results are shown in Fig. 1. In the distributions at the lower temperatures, 12, 4.7, and 1.6°K, there is a peak whose frequency is about  $(3.2 \pm 0.2) \times 10^{12}$  Hz. The rise in the intensity at low frequencies in these distributions is due to one-magnon scattering within the ground-state doublet of  $\text{CoF}_2$ , the  $AB$  transition described earlier.<sup>9</sup> The increase in the intensity at high frequencies is from scattering by the  $AC$  transitions.<sup>9</sup> Clearly both of these are far more intense than the peak studied in these experiments. This peak is believed to arise from two-magnon scattering, firstly because of its considerable temperature dependence (Fig. 1), and secondly because of the crystal-momentum dependence of the scattering. The peak was observed around the magnetic lattice point  $(1, 0, 0)$ , but was absent both at the nuclear lattice points  $(2, 0, 0)$  and  $(1, 0, 1)$ , and also at the magnetic lattice point  $(0, 0, 1)$ . This behavior is

in agreement with that predicted by Eq. (1). The peak is also believed to arise from two-magnon processes because its frequency and shape are in agreement with the calculations described below.

In Fig. 2 we show the results for several wave vectors around the reciprocal lattice point  $(1, 0, 0)$ , with a background, similar to that shown by the dashed lines in Fig. 1, subtracted. Although this background is of necessity somewhat arbitrary, we believe the results shown in Fig. 2 are substantially correct for the intensity, shape, and frequency dependence of the two-magnon scattering throughout the Brillouin zone.

At first sight the results of our measurements

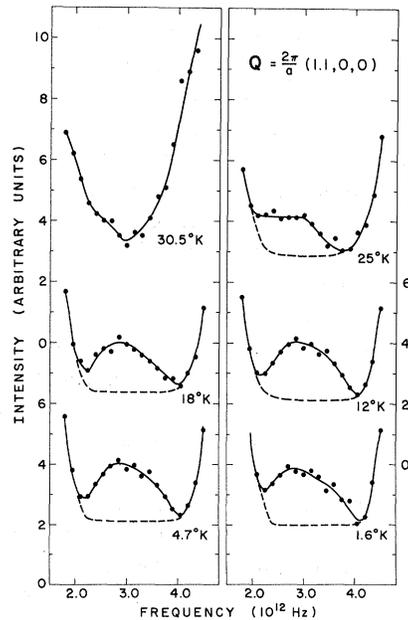


FIG. 1. Typical neutron groups observed from two-magnon scattering in  $\text{CoF}_2$  as a function of temperature. The dashed line shows our estimate of the background under the peaks. The right-hand scale applies only to the results at 18 and 12°K.

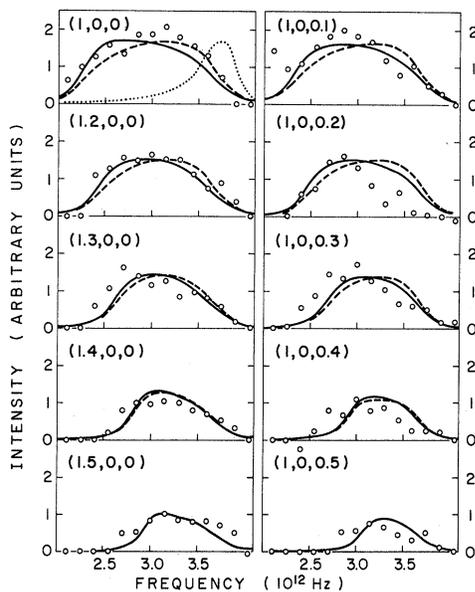


FIG. 2. The two-magnon scattering at 4.7°K, with the background subtracted (Fig. 1), for various wave-vector transfers. The solid (dashed) curves are the results of the spin-wave model with (without) interactions, and the dotted curve is the two-magnon density-of-states approximation. The spin-wave calculations are normalized with a single scale factor to the results. The density-of-states curve (dotted) has been reduced to a convenient size as it grossly disagrees with the observed magnitude of the two-magnon intensity.

are surprising because a large peak in the two-magnon spectrum would be expected at a frequency of  $3.8 \times 10^{12}$  Hz, twice the average frequency of the zone-boundary magnons.<sup>9</sup> Indeed the combined density-of-states model for the scattering, obtained by deleting the  $\sinh$  function in Eq. (1), does show a peak at this frequency (Fig. 2). Our experiment shows, therefore, that the density-of-states approximation fails to describe the two-magnon scattering.

The reason for this failure may be understood in the following way. The  $z$  component of the spin may be written in the spin-wave approximation as  $S_z = S - a^\dagger a$ . Two-magnon scattering of this type is possible at low temperatures only if both the creation and annihilation of local spin deviations, described by operators  $a^\dagger$  and  $a$ , are able to create magnons. In the antiferromagnet, scattering occurs because the annihilation of a spin deviation is able to create magnons.<sup>8</sup> An exception is the case of zone-boundary magnons which cannot be created by annihilation of a spin deviation. Consequently no scattering occurs in the region near twice the frequency of these modes. For lower frequencies the amount of

scattering is determined by the factor  $\sinh(\theta_i \mp \theta_j)$  in Eq. (1). In simple ferromagnets, two-magnon scattering of this type is absent because the annihilation of a spin deviation is not able to create magnons of any wave vector.

The two-magnon scattering was calculated from Eq. (1). The one-magnon properties of  $\text{CoF}_2$  were obtained from a spin- $\frac{1}{2}$  model which gave good agreement with the observed one-magnon frequencies,<sup>7,9</sup> and which involved Heisenberg nearest-neighbor exchange and anisotropic next-nearest-neighbor exchange ( $J_{xx} = J_{yy} \neq J_{zz}$ ). The calculations were performed mostly with a mesh of 4096 points and a frequency resolution of  $0.1 \times 10^{12}$  Hz. The results, which are given as the dashed curves of Fig. 2, show that this noninteracting-magnon model gives a good account of our measurements. The integrated intensity of the one-magnon scattering for a wave-vector transfer  $(1.5, 0, 0)2\pi/a$  on this model was calculated to be 5.8 times larger than the integrated intensity of the two-magnon scattering near  $(1, 0, 0)2\pi/a$ . This is also consistent with the experimental ratio of  $4.7 \pm 1.5$ , where the large error is mostly due to the uncertainty in the background. Calculations taking account of the effect on the scattered intensity of the complex nature of the wave functions in cobalt fluoride<sup>7</sup> gave the calculated ratio as 3.5.

A further calculation was performed to assess the importance of magnon-magnon interactions using a modified Ising form for the exchange interaction. The two-magnon scattering in this approximation, which will be fully discussed in a later publication, is shown by the solid curves in Fig. 2. It only differs slightly from the results of the noninteracting theory, and the slightly better agreement with experiment is barely significant. Much greater accuracy will be required in the measurement of the two-magnon scattering before the difference between the calculations is measurable.

In conclusion, we have observed two-magnon scattering of neutrons in cobalt fluoride. The results show, not surprisingly, that a simple density-of-states approach is unrealistic but that calculations based on either an interacting- or noninteracting-magnon model with the actual neutron-magnon interaction do give good agreement with our measurements at low temperatures. The temperature dependence of the two-magnon scattering has also been found to be more marked (Fig. 1) than that of the one-magnon scattering.<sup>9</sup> A quantitative description of these latter mea-

surements must await further theoretical developments.

<sup>1</sup>S. J. Allen, R. Loudon, and P. L. Richards, Phys. Rev. Letters **16**, 463 (1966).

<sup>2</sup>P. A. Fleury and R. Loudon, Phys. Rev. **166**, 514 (1968).

<sup>3</sup>J. W. Halley and I. Silvera, Phys. Rev. Letters **15**, 654 (1965).

<sup>4</sup>R. J. Elliott, M. F. Thorpe, G. F. Imbush, R. Loudon, and J. B. Parkinson, Phys. Rev. Letters **21**, 147

(1968).

<sup>5</sup>R. J. Elliott and M. F. Thorpe, to be published.

<sup>6</sup>P. A. Fleury, Phys. Rev. Letters **21**, 151 (1968).

<sup>7</sup>R. A. Cowley, P. Martel, and R. W. H. Stevenson, Phys. Rev. Letters **18**, 162 (1967).

<sup>8</sup>O. Nagai and A. Yoshimori, Progr. Theoret. Phys. (Kyoto) **25**, 595 (1961).

<sup>9</sup>P. Martel, R. A. Cowley, and R. W. H. Stevenson, Can. J. Phys. **46**, 1355 (1968).

<sup>10</sup>B. N. Brockhouse, in Inelastic Scattering of Slow Neutrons from Solids and Liquids (International Atomic Energy Agency, Vienna, Austria, 1961), p. 113.

### EXACT RESULTS IN THE KONDO PROBLEM: EQUIVALENCE TO A CLASSICAL ONE-DIMENSIONAL COULOMB GAS

P. W. Anderson

Cavendish Laboratory, Cambridge University, Cambridge, England,  
and Bell Telephone Laboratories, Murray Hill, New Jersey 07974

and

G. Yuval

Cavendish Laboratory, Cambridge University, Cambridge, England

(Received 28 January 1969)

We demonstrate an exact equivalence between a Kondo problem and the thermodynamics of a classical one-dimensional gas with alternating charges and a logarithmic potential. This classical gas has a critical point.

We demonstrate an asymptotically exact equivalence<sup>1</sup> between the simplest Kondo problem, the ground state of a spin  $S = \frac{1}{2}$  interacting with an otherwise free electron gas, and the thermodynamics of a certain one-dimensional classical gas. The central point is an asymptotic expression (i.e., for  $E_F t \gg 1$ ) for the probability amplitude for a succession of spin flips. The interaction in the classical gas is of logarithmic (two-dimensional Coulomb) type for which simpler cases have been solved exactly by Dyson, Wilson, and Gunson.<sup>2</sup> Some physical results from the classical gas problem are available already—e.g., that the antiferromagnetic case has no mean spin—and these will be discussed in a succeeding communication. Generalizations to finite  $T$ , higher spin, and more physical models seem not forbidding.

We divide the Hamiltonian as follows:

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1, \quad (1)$$

$$\mathcal{H}_0 = \sum_{k,\sigma} \epsilon_k n_{k\sigma} + JS_z \sum_{kk'\sigma\sigma'} c_{k\sigma}^\dagger (s_z)_{\sigma\sigma'} c_{k'\sigma'}, \quad (2)$$

$$\mathcal{H}_1 = J \sum_{k,k'} c_{k\sigma}^\dagger c_{k'\sigma'} [S_+(s_-)_{\sigma\sigma'} + S_-(s_+)_{\sigma\sigma'}], \quad (3)$$

The eigenstates of  $\mathcal{H}_0$  are

$$\Psi_\dagger = \alpha \prod_k c_{k\dagger}^\dagger \prod_{k'} d_{k'}^\dagger |\text{vac}\rangle, \quad \Psi_\ddagger = \beta \prod_k d_{k\dagger}^\dagger \prod_{k'} c_{k'\ddagger}^\dagger |\text{vac}\rangle, \quad (4)$$

where  $c_k^\dagger$  are scattering states appropriate to the potential  $+\frac{1}{4}J$  [phase shift  $\delta_+ \simeq -\frac{1}{4}\pi J\rho(E_F)$ ] and  $d_k$  are scattering states appropriate to  $-\frac{1}{4}J$  ( $\delta_- \simeq -\delta_+$ ). For  $J$  antiferromagnetic,  $\delta_+$  is negative.

The lowest eigenstate with  $S_z = +\frac{1}{2}$  we denote by  $\alpha\Psi_{0\dagger}$ . We calculate

$$F(t) = \langle \alpha\Psi_{0\dagger} | \exp(i\mathcal{H}t) | \alpha\Psi_{0\dagger} \rangle = \sum_\omega |\langle \alpha\Psi_{0\dagger} | \Psi_\omega \rangle|^2 e^{i\omega t}, \quad (5)$$

where  $\Psi_\omega$  are the exact eigenstates. We assume that the Fourier transform  $F(\omega)$  must have a branch