

teraction, for which the  $r_{12}^{-1}$  of Eq. (2) may be replaced by  $\delta(r_{12})$ . We employ the unscreened interaction because we do not anticipate strong screening over dimensions small with respect to those of the local moment.

<sup>5</sup>An exchange integral, evaluated for simple plane waves as in Eq. (2), does not display such strong oscillations as seen in Fig. 1. The evaluation (as we have done) of Eq. (3) with exchange integrals based on plane waves orthogonalized to the Gd ion's shells is in some senses inconsistent with Eq. (3) and its simple plane-wave functional character, though one may argue its appropriateness when viewing  $\rho$  away from the Gd site. Orthogonalization effects on  $J$  and elsewhere must be accounted for in an ultimate treatment of the problem, and in anticipation of such a treatment we have chosen to use  $J(Q)$  based on orthogonalized functions. The effect of inserting orthogonalized plane-wave functions into Eq. (1) is significant and will be reported in a further publication.

<sup>6</sup>Two observations should be made concerning the  $Q$  dependence of  $J$ . First, there is a singularity at  $2k_F$  (barely observable in the present case) due to the  $Q$  sampling. Second,  $J$  oscillates with decreasing amplitude with increasing  $Q$ . These oscillations are of much more significant magnitude than the oscillations

in the tail of the form factor (see Fig. 1).

<sup>7</sup>Free-electron theory normally yields a choice of  $k_F$  between 0.5 and 1.0 a.u. for most metals.

<sup>8</sup>The  $\rho(r)$  are plotted for unnormalized conduction-electron orbitals, but assuming common normalization, the three  $\rho(r)$  are scaled appropriately for direct comparison.

<sup>9</sup>These were obtained by observing the range in which  $\rho(0)$  oscillates [due to the varying sign of  $J(Q)$ ] as the upper limit of integration is varied. In addition, we have crudely estimated the effect on  $\rho(0)$  of going from our  $J(Q)$  sampling to a suitably averaged  $J(Q)$ .

<sup>10</sup>For the experimental situation considered by Giovannini, Peter, and Schrieffer,<sup>2</sup> see, for example, D. Shaltiel, J. H. Wernick, H. J. Williams, and M. Peter, Phys. Rev. **135**, A1346 (1964); G. Low, Proceedings of the International Conference on Magnetism, Nottingham, England, 1964 (Institute of Physics and the Physical Society, University of Reading, Berkshire, England, 1965); P. Craig, B. Mozer, and R. Segnan, to be published.

<sup>11</sup>P. W. Anderson and A. M. Clogston, Bull. Am. Phys. Soc. **2**, 124 (1961); J. Kondo, Progr. Theoret. Phys. (Kyoto) **28**, 846 (1962); S. Koide and M. Peter, Rev. Mod. Phys. **36**, 160 (1964); and R. E. Watson, S. Koide, M. Peter, and A. J. Freeman, to be published.

### DENSITY-OF-STATES EFFECTS IN THE MAGNETIC STIFFNESS OF 3d-3d TRANSITION-METAL ALLOYS

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In Figs. 1(a) and 1(b) we give neutron-scattering results for the spin-wave stiffness constant [ $D(0)$  in the spin-wave energy term  $D(T)q^2$ ] of bcc 3d-3d transition-metal alloys, and compare them with a quantity obtained from an approximate density-of-states curve of these materials. Bearing in mind the uncertainties that attend a derivation of the density-of-states function, the qualitative resemblance of the two figures is rather remarkable. We give below what we believe to be the explanation of it.

A year ago we published results for both bcc and fcc phases of the Fe-Ni system, and interpreted them with apparent success in terms of the Heisenberg Hamiltonian.<sup>1</sup> According to that discussion the stiffness may be computed from the mean spin  $\bar{S}$  and an effective exchange parameter which is such that  $\mathcal{J}_{\text{eff}}(\vec{r}-\vec{r}')\bar{S}^2 = \langle \mathcal{J}_{\vec{r}\vec{r}'} S_{\vec{r}} S_{\vec{r}'} \rangle$ , where  $\mathcal{J}_{\vec{r}\vec{r}'}$  can be any of the three types of coupling AA, AB, and BB. We

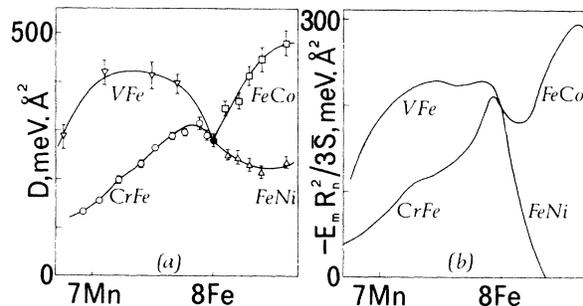


FIG. 1. (a) The observed spin-wave stiffness parameter  $D(0^\circ\text{K})$  for 3d-3d alloys; (b) an approximate prediction of this parameter using the total magnetic energy  $E_m$  deduced from the density of states.

now find that the same interpretation cannot be made for the Cr-Fe system, and it therefore appears that our previous fit may have been due merely to the fact that we were matching in each phase a three-parameter quadratic form to a smoothly varying set of only six or

seven points.

The Cr-Fe series affords a particularly clear-cut test of the idea that a phenomenological Heisenberg Hamiltonian will suffice to explain the data. It so happens that with increasing concentration of Cr, the spin of the Cr atom declines steadily and passes through zero at  $40 \pm 2\%$ .<sup>2</sup> According to our previous discussion the value of  $D$  where one of the spins is zero is simply  $2a^2 J_{AA} S_A c_A$ , where  $J^{(n)}$  is the  $n$ th spatial moment of  $\mathcal{J}$  and suffices  $A$  refer to the element of nonzero spin. Thus on the Heisenberg picture the stiffness is expected to differ from the value for the pure element,  $2a^2 J_{AA}^{(2)} S_A$ , only insofar as  $S_A$  and the concentration  $c_A$  have changed, and a parameter-free test of the model can be made. When this test is applied to Cr-Fe alloys, the prediction is found to be in error by  $18 \pm 4\%$ .<sup>3</sup>

This result and the associated failure of the Heisenberg picture to explain the results for Cr-Fe destroys confidence in the simple analysis, and we have sought a band-theoretic rationalization of the data.

In the case of the bcc  $3d$  alloys, a density-of-states curve is available that is believed to have semiquantitative significance insofar as Shimizu, Takahashi, and Katsuki have derived from it specific heats, magnetic susceptibilities, and their temperature dependences, in addition to a number of other phenomena in rough agreement with experiment.<sup>4</sup> Recently, Shimizu and Katsuki<sup>5</sup> have used an improved curve, and the ideas of the rigid-band model, to derive values as a function of electron number for a variety of quantities such as the exchange splitting and the total internal magnetic energy  $E_m$  of the ground state. They draw attention in particular to the fact that the interband splitting deduced—some  $1.56 \text{ eV atom}^{-1}$  for iron—is an order of magnitude greater than  $E_m$ , while within a factor of two  $E_m$  agrees with  $kT_C$  over the whole range of electron number we have studied, as it should.

These conclusions hold a number of points of interest for spin-wave theory. We recall that according to band theory the elementary excitations of a Bravais ferromagnet are, in addition to the spin-wave branch, a manifold of one-electron excitations raised at  $q=0$  by an energy equal to the band splitting. In our alloys this manifold is far above  $kT_C$ , which nowhere substantially exceeds  $0.1 \text{ eV}$ . Thus certainly for temperatures well below  $kT_C$  the

only excitations strongly in evidence in the material will be those corresponding to the single spin-wave branch. If this state of affairs were to continue to be true almost up to the Curie temperature, the relationships between  $D$ ,  $E_m$ , and  $T_C$  would not necessarily be drastically different from those which would obtain in an insulator subject to Heisenberg coupling.

In an exchange-coupled insulator, where  $E_m = -J^{(0)} Z S^2$ , the stiffness would be given by  $-E_m R_n^2 / 3S$  times the ratio  $J^{(2)} / J^{(0)}$  of the second to the zeroth spatial moment of the exchange coupling. ( $R_n$  is the nearest-neighbor separation.) To explore the possibility that some such relationship might hold approximately in the  $3d$ - $3d$  alloys, we have plotted  $-E_m R_n^2 / 3S$ , using a band-theory  $E_m$  determined in the manner of Shimizu and Katsuki, as a solid line in Fig. 1(b). It does indeed have the qualitative behavior of our experimental  $D$ . As it would be quite reasonable to expect that  $J^{(2)} > J^{(0)}$  in  $3d$  metals, insofar as these parameters can be given a meaning in the present argument, it is not surprising that the theoretical  $D$  in Fig. 1 is low by a factor  $\sim 2$ . More striking is the fact that the arbitrary use of  $J^{(2)} / J^{(0)} = 1.8$  would produce agreement within about  $\pm 20\%$  over the whole series of alloys, except for the Fe-Ni system for which the theoretical curve is obviously wrong as it gives a  $T_C$  falling to zero.

On both sides of the point for pure iron the theoretical curve bifurcates, and this division is reflected in our results. According to band theory, the existence of different  $D$ 's for the two alloys of a given electron number derives from the different exchange splittings in the two alloys, which are known to have different Fermi levels because their saturation magnetizations are not the same. The severe quantitative discrepancy in the case of Fe-Ni could be removed by only slight changes in the shape of the density-of-states peak near 7 electrons  $\text{atom}^{-1}$ , this region being one in which the density-of-states curve is known to be unreliable; the desirable changes improve the agreement with experiment for susceptibility in the controversial region.

It would seem to follow that the spectrum of excitations in these substances is such that the spin-wave dispersion relation and the phenomena that depend on it behave in a phenomenological sense like those of an insulator, the quantitative details being determined by

their band structure.

We are grateful to Sir Nevill Mott for a conversation about the work.

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<sup>1</sup>M. Hatherly *et al.*, Proc. Phys. Soc. (London) **84**, 55 (1964).

<sup>2</sup>C. G. Shull and M. K. Wilkinson, Phys. Rev. **97**, 304

(1955).

<sup>3</sup>W. Marshall and G. A. Murray have recently shown in unpublished work that our formula for the stiffness of an insulator requires the addition of a further term, effectively taking the discussion to second order in perturbation theory; the effect in the present context is to increase the discrepancy with experiment.

<sup>4</sup>M. Shimizu, T. Takahashi, and A. Katsuki, J. Phys. Soc. Japan **19**, 1856 (1964), and references therein.

<sup>5</sup>M. Shimizu and A. Katsuki, Proceedings of the International Conference on Magnetism, Nottingham, England, 1964 (Institute of Physics and the Physical Society, University of Reading, Berkshire, England, 1965).

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## NUCLEAR SPIN RELAXATION IN *p*-TYPE InSb AT LIQUID-HELIUM TEMPERATURES\*

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The spin-lattice relaxation rate  $1/T_1$  of In<sup>115</sup> nuclei in the semiconductor InSb has been studied and explained for samples ranging in net acceptor impurity concentration from  $10^{14}/\text{cm}^3$  to  $10^{17}/\text{cm}^3$ . The temperature dependence of the relaxation rate between 1.0 and 4.2°K was determined as well as the magnetic field dependence between 300 Oe and 10 kOe. Three distinct relaxation mechanisms were found, which are operative singly in each of three different concentration regions. The relaxation mechanism occurring in the intermediate-concentration region is of a new type, characterized by an inverted temperature dependence of the relaxation rate ( $1/T_1$  increases as the temperature decreases) and an  $H^{-1/4}$  magnetic field dependence. This relaxation mechanism is attributed to the dipolar coupling between In<sup>115</sup> nuclei and nearby bound paramagnetic holes associated with acceptor impurities, the coupling being modulated by the hopping of holes among impurity sites, typical of impurity conduction in a compensated semiconductor.<sup>1</sup> Spin diffusion among In<sup>115</sup> nuclei permits the nuclear spins distant from the paramagnetic holes to relax. The relaxation mechanism at low concentration is due to modulation of the same dipolar coupling by the spin-lattice relaxation of the bound holes, with subsequent In<sup>115</sup> spin diffusion, and the high-concentration relaxation mechanism is probably due to free holes associated with impurity-band conduction.

The samples were all cut from the same gold-

doped single crystal which became contaminated during growth by an unknown shallow acceptor which is responsible for the samples' electrical properties. The compensation  $N_D/(N_A - N_D)$  for the samples of net acceptor concentration between  $7 \times 10^{14}/\text{cm}^3$  and  $3 \times 10^{16}/\text{cm}^3$  lies between 2.0 and 1.6, and for the low-concentration samples is about 6. Figure 1 illustrates the dependence of the relaxation rate measured at 5 kOe on net acceptor concentration. The temperature dependence is also shown at a few of the temperatures at which measurements were taken. The three concentration regions are roughly distinguishable from the figure. The low-concentration region, in which  $1/T_1$  is characterized by linear dependence on net acceptor concentration and by a weak, but normal-sense, temperature dependence, encompasses impurity concentrations between  $\sim 10^{14}/\text{cm}^3$  and  $\sim 10^{15}/\text{cm}^3$ . The intermediate-concentration range, in which the striking property is the inverse temperature dependence of  $1/T_1$ , extends from  $\sim 2 \times 10^{15}/\text{cm}^3$  to  $\sim 2 \times 10^{16}/\text{cm}^3$ . The high-concentration range, where  $1/T_1$  again has a temperature dependence in the normal sense, is taken as above  $\sim 3 \times 10^{16}/\text{cm}^3$ . Since it can be easily shown that none of the usual intrinsic relaxation mechanisms<sup>2</sup> is capable of yielding relaxation times as short as those observed in the low-concentration region, it is reasonable to hypothesize that the relaxation there arises from the Bloembergen mechanism which entails fast-relaxing paramagnet-