GENERALIZED SUSCEPTIBILITY OF NICKEL

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Measurements of the magnetic inelastic neutron scattering from paramagnetic nickel at $1.6T_C$ give a good absolute fit to calculations for a simple free-electron gas with a random-phase approximation exchange-enhancement factor in the region of 3.

We give herein an analysis of recent measurements of the magnetic inelastic scattering of neutrons from paramagnetic nickel in terms of electron-gas theory. It suggests that the degree of exchange enhancement operating at $1.6T_C$ increases the static susceptibility by a factor of only about 3, and gives a value for the effective intra-atomic Coulomb repulsion integral at that temperature of approximately 0.4 eV.

The experiment, which has already been briefly described,[1] consisted in measurements of the magnetic scattering cross section

$$\frac{\partial^2\sigma}{\partial\Omega\partial E}(\vec{K},\omega)=\frac{1}{\hbar}(r_0\gamma)^2\frac{k'}{k}|F(\vec{K})|^2\sum_{\alpha\beta}(\delta_{\alpha\beta}-\hat{K}_\alpha\hat{K}_\beta)S_{\alpha\beta}(\vec{K},\omega) \tag{1}$$

(here written in conventional notation) for a single crystal of Ni^{60} at 1020 ± 10°K over the whole range of $\vec{K}$ and throughout an interval of energy transfers from 0.02 to 0.12 eV. According to Izuyama, Kim, Kim, and Kubo,[2]

$$S_{\alpha\beta}(\vec{K},\omega)=\frac{\hbar}{\pi g^2\mu^2}(n_k+\tfrac{1}{2}\pm\tfrac{1}{2})\,\mathrm{Im}\chi_{\alpha\beta}(\vec{K},|\omega|), \tag{2}$$

where we have written the thermodynamic factor in terms of a Bose-Einstein occupation value

$$n_k=[1-\exp(-|\hbar\omega\beta|)]^{-1}, \tag{3}$$

with $\pm\hbar\omega$ the transfer of energy to the magnet.

Doniach[3] has expressed $\mathrm{Im}\chi_{zz}$ for the unenhanced free-electron gas at absolute zero in the form

$$\mathrm{Im}\chi_{\mathrm{non}}(\vec{K},\omega)=\frac{\pi}{8}g^2\mu^2N(E_F)\frac{k_F}{K}\{(1-x_+^2)\theta(x_+^2)-(1-x_-^2)\theta(x_-^2)\}, \tag{4}$$

with $N(E)$ the density of states per atom per spin state, k_F the electron wave vector at the Fermi surface,

$$x_\pm=\frac{\hbar^2K^2\pm2m^*\hbar\omega}{2\hbar^2k_FK},$$

and

$$\theta(y)=1,\ y<1;\qquad \theta(y)=0,\ y\geq1. \tag{5}$$

$\mathrm{Im}\chi_{\mathrm{non}}$ is zero at $\omega=0$, and has maxima along the locus $\hbar\omega/E_F=(K/k_F)^2$ or $2(K/k_F)-(K/k_F)^2$ whichever is the greater (Fig. 1).

In random-phase approximation, exchange interaction leads in the standard manner to an enhanced

$$\chi(\vec{K},\omega)=\chi_{\mathrm{non}}(\vec{K},\omega)/[1-2I_{\mathrm{eff}}\chi_{\mathrm{non}}(\vec{K},\omega)/g^2\mu^2], \tag{6}$$

where I_{eff}, in the application to nickel, at any rate, is the effective intra-atomic Coulomb repulsion[4];

$$\alpha=1/[1-I_{\mathrm{eff}}N(E_F)] \tag{7}$$

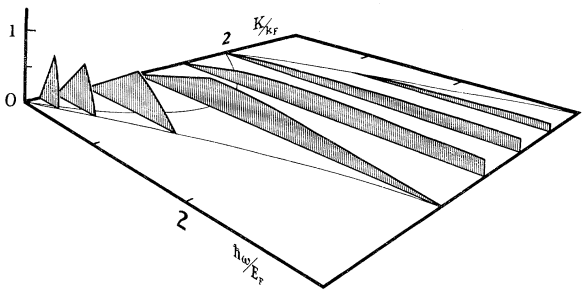

FIG. 1. The imaginary part of the generalized susceptibility of a noninteracting free-electron gas at absolute zero, in units of $g^2\mu^2N(E_F)$.

is the so-called "enhancement factor" by which $\chi(0,0)$ is increased. A further effect of exchange enhancement in $\mathrm{Im}\chi$ is to shift the line of maxima to lower ω, physically as Doniach points out because the more nearly "critical" the system, the slower must be the dominant fluctuations. Hubbard[5] has discussed the fact that in nickel, other sources of enhancement beside Coulomb repulsion should have a relatively small influence on I_{eff}; furthermore the principal one, intra-atomic exchange, would be expected in this substance to lead to almost the same value of I_{eff} anyway, so that nickel is peculiarly well suited for making a test of the general concepts underlying Eq. (6).

To give a treatment for nickel corresponding to that of Eq. (4), $\chi(\vec{K},\omega)$ should strictly speaking be computed from the Lindhard formula with due regard to the presence of degenerate d bands and to the actual Fermi surface (FS) of nickel. Now if, to fix ideas, we were to say that the number n of d-s holes per atom in paramagnetic nickel were as great as 1.2, the appropriate FS could be said to have withdrawn from the boundary of the first Brillouin zone essentially to the same extent that 0.6 of a hole withdraws the d-s FS in the ferromagnetic phase; that is to say, the FS would have broken right away from the Brillouin-zone boundary and would be shaped like a knobbly sphere of radius $k_F \approx 1.4$ Å^{-1} about the center point Γ.[6,7] In such circumstances, paramagnetic nickel would be almost an ideal substance on which to test the free-electron theory of susceptibility. It is likely, however, that the number of d-s holes is much more nearly 0.6, in which case the paramagnetic FS must be constructed by expanding the knobbly spheres of the repeated-zone scheme until they interconnect via a rather complex system of tubes and protuberances.[8] Even if the latter is the case, however, the dominant "caliper dimension" governing the broad features of χ through the Lindhard formula will be not too far from that of the knobbly sphere, and from arguments of simple continuity it might be supposed that where surface is lost, density of states is gained, so that the net result of computing the Lindhard expression should not be too sensitive to the exact form of the FS, certainly for the smaller K's and ω's. Moreover the majority of the d-s holes, being at the very top of a band, are in a roughly parabolic density of states.

These arguments are loose, for want of a more closely detailed description of the band structure than we have, but they raise the possibility of finding some "effective" equivalent electron gas by direct application of Eq. (6).

Any such comparison with free-electron gas theory should for a further reason be made only for the smaller K's, perhaps up to $\frac{1}{2}K_{\max} \approx 0.9$ Å^{-1} in the present case. For at larger K's the umklapp terms that render χ periodic in $\vec{K}$ will certainly cause departures from free-electron behavior.

Figure 2 shows the appropriate measured susceptibilities. At $K = 0.5$-0.9 Å^{-1} the points out to 0.05 eV do cluster quite clearly along straight lines radiating from the origin of ω as predicted, and at larger energies they tend to bend over towards the horizontal. A set of curves was fitted by writing χ in terms of E_{F}, k_{F}, and I_{eff} as independent variables, proper account being taken[3] of the elevated temperature which broadens the energy distribution of Fig. 1 by an amount $\approx kT$. As seen in Table I, a k_{F} in the anticipated range has indeed emerged

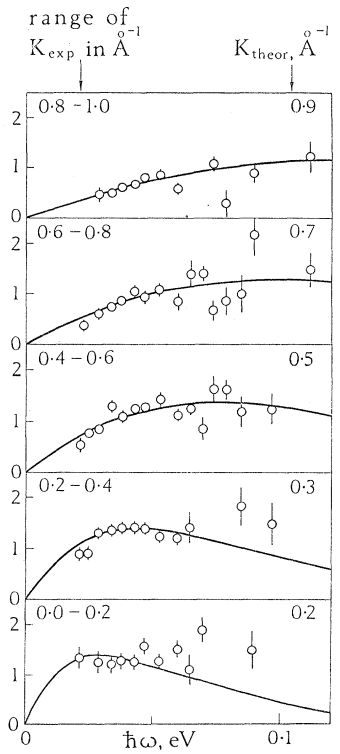

FIG. 2. Absolute values of $\mathrm{Im}\chi_{zz}(K,\omega)/g^2\mu^2$ in eV^{-1} atom^{-1} deduced from the neutron scattering of nickel at $1.6T_c$, together with curves calculated for an effectively equivalent free-electron gas.

Table I. Constants of an "equivalent free-electron gas" accounting for the generalized susceptibility of nickel at 1.6 T_c

E_F	0.4 ± 0.15 eV
k_F	1.3 ± 0.2 Å^{-1}
$N(E_F)$	1.6 ± 0.4 states eV^{-1} atom^{-1} (spin state)$^{-1}$
m^*/m	16 ± 5
n	0.84 ± 0.2 holes atom^{-1}
I_{eff}	0.4 ± 0.2 eV
α	3 ± 1

from this procedure, while the derived quantities m^*, $N(E_F)$, and n take values in general agreement with those obtained for nickel by other methods.

The I_{eff} we find is in the range of values that might be expected from the original discussions of Hubbard and Kanamori, Hubbard's estimate[5] for nickel being ~0.8 eV. Our I_{eff} corresponds to the $U_{\text{eff}}{}^{d-d}\sum_\mu(\delta n_\mu)^2/(\delta n)^2$ of Hodges, Ehrenreich, and Lang[9] [their Eq. (4.1)], for which their calculation gives a value of 0.66 eV. In fact, I_{eff} in the present simple model of Eq. (6) must have a temperature dependence.[4] To discuss it, Wohlfarth[10] writes Eq. (7) as $\alpha = [1 - \bar{I}]^{-1}$, with $\bar{I}$ the product of I_{eff} and $\int_0^\infty N(E) \times (df/dE)dE$. He finds that solely on the basis of the temperature smearing of the FS, $\bar{I}$ would fall from unity at T_c to about 0.83 at our temperature of $1.6T_c$. According to Edwards,[11] I_{eff} for a parabolic band should be reduced by a factor ~0.9 in the same temperature interval owing to an increase in the number of intermediate states available during a two-particle scattering process. From these considerations one would expect in the present investigation an α of roughly $[1-0.75]^{-1} = 4$, which is consistent with what we observe. Our effective $N(E_F)$ of 1.6 is, indeed, lower than the 2.1 calculated by Hodges et al.[7,9] (for paramagnetic nickel at 0°K) by approximately the reduction factor suggested by Wohlfarth.

It is notable that the enhanced static susceptibility $\chi(0,0)$ corresponding to a free-electron gas with the constants in our Table I is only 3×10^{-4} emu mole^{-1}, a third of the actual value[12] for nickel at 1020°K. By the Kramers-Kronig relations, $\chi(0,0)$ is the small-K limit of $\int\omega^{-1}\text{Im}\chi d\omega$, the true value of which at small K must therefore be greater than the value for a free-gas model. As seen in the lower diagrams of Fig. 2 this is indeed confirmed, the experimental points at the higher frequencies refusing to fall to zero within the range over which we have taken measurements. Thus the most appropriate "equivalent electron gas" for discussing the dynamics of this material would seem, curiously enough, not to dominate the static properties owing to a strong contribution to the latter from electron transfers of comparatively high energy.

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