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Magnetic Behaviour of Band Electrons

RYOGO KUBO, TAKEO IZUYAMA AND DUK-JOO KIM
University of Tokyo, Tokyo, Japan
AND
YOSUKE NAGAOKA
Kyoto University, Kyoto, Japan

Two problems are studied in this report on the magnetic behaviour of band electrons. One is the neutron diffraction phenomena due to band electrons, and the other is the dynamical effect for the so-called *s-d* interaction. In the first problem the correlation effect between the electrons is taken into account by means of the ladder approximation. It is shown that the band model is able to explain the neutron diffraction phenomena, such as magnetic diffuse scattering in ferromagnetic states, critical scattering and magnetic scattering in the paramagnetic region. In the second problem the dynamical effects on the spin waves in the *s-d* model are examined.

1. Neutron Diffractions

Some years ago, some investigators, Elliott and Marshall[1] among others, made successful theories which explain very well the neutron diffraction experiments on iron in terms of a localized spin model. We shall show in the following that these neutron diffraction experiments can be understood equally well by the itinerant electron model, so that the neutron diffraction data do not necessarily give support to the unique validity of the localized spin model, contrary to the view expressed by previous investigators. It is easily shown that the interacting electrons in ferromagnetic states give the sharp Bragg spots. Further, the band theory is able to explain magnetic diffuse scattering in ferromagnetic states, critical scattering and magnetic scattering in the paramagnetic region, if the Coulomb interaction of electrons is properly taken into account.

The differential cross-section of the neutron scattering by electron spins is expressed in terms of the function $S_{\alpha\beta}(\boldsymbol{\kappa}, \omega)$ ($\alpha, \beta = x, y$ or z) describing the pair correlation between spins in the electron system. The correlation function $S_{\alpha\beta}(\boldsymbol{\kappa}, \omega)$ is related to the dynamical susceptibility $\chi_{\alpha\beta}(\boldsymbol{\kappa}, \omega)$ of this electron system by the formula,

$$S_{\alpha\beta}(\boldsymbol{\kappa}, \omega) = i\frac{\chi_{\alpha\beta}(\boldsymbol{\kappa}, \omega+i0) - \chi_{\alpha\beta}(\boldsymbol{\kappa}, \omega-i0)}{e^{\hbar\omega/k_BT}-1}$$

where $\boldsymbol{\kappa}$ corresponds to the change of the wave vector of a neutron due to the magnetic scattering and $\hbar\omega$ is the change of the neutron energy by scattering.

For mathematical convenience, the $3d$ electrons are assumed to be in a single band and the overlaps of the Wannier functions are neglected. Then, expressing $\boldsymbol{\kappa}$ as a sum of a reciprocal lattice vector $\boldsymbol{K}$ that is closest to $\boldsymbol{\kappa}$ and a wave vector $\boldsymbol{q}$ in the first Brillouin zone, we get

$$\chi_{\alpha\beta}(\boldsymbol{\kappa}, \omega) = |F(\boldsymbol{\kappa})|^2 \chi'_{\alpha\beta}(\boldsymbol{q}, \omega)$$

$$\chi'_{\alpha\beta}(\boldsymbol{q}, \omega) = i \times \int_0^\infty dt\, e^{-i\omega t} \left[\left(\sum_k \tilde{a}^+_{k+q}(t)\, \tilde{\sigma}_\alpha \tilde{a}_k(t),\ \sum_l \tilde{a}^+_l \tilde{\sigma}_\beta \tilde{a}_{l+q} \right) \right]$$

where $\tilde{\sigma}$ is the Pauli spin matrix, $\tilde{a}_k = \begin{pmatrix} a_{k\uparrow} \\ a_{k\downarrow} \end{pmatrix}$ the destruction operator for the Bloch orbital with wave vector $\boldsymbol{k}$, defined as $\tilde{a}_{k+K} = \tilde{a}_k$ (mod $\boldsymbol{K}$), $\boldsymbol{K}$ being any reciprocal lattice vector, $F(\boldsymbol{\kappa})$ the form factor given by the Wannier function, and the sum is carried over $\boldsymbol{k}$ in the first Brillouin zone. The reduced susceptibility $\chi'_{\alpha\beta}(\boldsymbol{q}, \omega)$ is calculated under the following approximations:

(i) The matrix elements of the Coulomb interaction in the Bloch orbital representation are taken to be a constant v. (ii) The electron self-energy is approximated by the exchange self-energy and the remaining correlation effects of electrons are treated by means of the ladder approximation (or the R.P.A.[2]). Then we obtain

$$\chi'_{-+}(q,\omega)=\frac{-\Gamma_{-+}(q,\omega)}{1+v\Gamma_{-+}(q,\omega)}$$

and

$$\chi'_{zz}(q,\omega)$$
$$=-\frac{\Gamma_{\uparrow}(q,\omega)+\Gamma_{\downarrow}(q,\omega)-2v\Gamma_{\uparrow}(q,\omega)\Gamma_{\downarrow}(q,\omega)}{1-v^2\Gamma_{\uparrow}(q,\omega)\Gamma_{\downarrow}(q,\omega)}$$

where

$$\Gamma_{-+}(q,\omega)\equiv\sum_k\frac{f_{k+q,\downarrow}-f_{k,\uparrow}}{\varepsilon_{\downarrow}(k+q)-\varepsilon_{\uparrow}(k)-\hbar\omega}$$

and

$$\Gamma_{\uparrow}(q,\omega)\equiv\sum_k\frac{f_{k+q,\uparrow}-f_{k,\uparrow}}{\varepsilon(k+q)-\varepsilon(k)-\hbar\omega}, \text{ etc.}$$

Here $f_{k,\mu}$ is the occupation probability of electrons in the state characterized by the wave vector k and spin direction μ, and $\varepsilon_\mu(k)$ is the energy of this state including the exchange self-energy;

$$\varepsilon_\mu(k)\equiv\varepsilon(k)-v\sum_\kappa f_{k+\kappa,\,\mu}$$

At $T<T_c$, $\chi'_{-+}(q,\omega)$ has poles corresponding to spin waves as well as those due to Stoner excitations. Proper consideration of the former leads to the following magnetic diffuse scattering;

$$\frac{d^2\sigma}{d\Omega d\omega}=\left(\frac{2g_0e^2}{m_0c^2}\right)^2\frac{k'}{k}\,|F(\kappa)|^2\,2(1+\hat{\kappa}_z^2)$$
$$\times\frac{N}{e^{\beta\hbar\omega}-1}\delta(\omega-\omega(q)),$$

where N is the total number of electrons, m_0 the mass of an electron, $g_0=1.911$, $\hat{\kappa}$ the unit vector in the direction of κ, k the wave vector of incident neutrons, and k' is that of scattered neutrons. In the above the dispersion relation for the spin waves is taken to be $\omega=\omega(q)$. The scattering cross section due to Stoner excitations which must be added to this result is neglected here, because it gives extremely small diffuse spots as shown by Elliott[3]. The above result is substantially equal to that given by the localized model, because $\omega(q)$ is proportional to q^2 for sufficiently small values of q.

The magnetic critical scattering (at $T>T_c$) is obtained as a natural consequence of our calculation. Adopting the effective mass approximation for the holes in the d-band, we expand

$$\Re e\,\Gamma(q,\omega)\equiv R(q,\omega)$$

as

$$R(0,0)+\left(\frac{mV}{24\pi^2\hbar^2k_F}\right)q^2$$
$$+\left(\frac{m^3V}{2\pi^2\hbar^4k_F}\right)\frac{\omega^2}{q^2}+\left(\frac{m^3V}{12\pi^2\hbar^4k_F{}^3}\right)\omega^2+\cdots,$$

and note that

$$\Im m\,\Gamma(q,\omega)\begin{cases}=\left(\dfrac{V}{2\pi}\dfrac{m^2}{\hbar^3}\right)\dfrac{\omega}{q}, & \text{if } |\omega|<v_Fq,\\ =0, & \text{if } |\omega|>v_Fq,\end{cases}$$

where m is the effective mass, and V is the volume of the system. Thus we obtain

$$\frac{d\sigma}{d\Omega}=\int d\omega\frac{d^2\sigma}{d\Omega\,d\omega}$$
$$=\left(\frac{2g_0{}^2e^2}{m_0c^2}\right)^2|F(\kappa)|^2\frac{C}{B(T-T_c)+q^2}.$$

The constants are defined by

$$v_F=\hbar k_F/m,\qquad E_F=\hbar^2k_F{}^2/2m$$

$$B=\frac{8(3\pi^2)^{1/3}}{3}\frac{k_B{}^2T_cm^2}{\hbar^4(N/V)^{2/3}}\left(1+\frac{\pi^2}{12}\frac{k_B{}^2T_c{}^2}{E_F{}^2}\right)^{-2},$$

$$C=54\pi^2\frac{\hbar k_BT_cv_F}{E_F{}^2}\left(\frac{N}{V}\right)N.$$

Now v may be fixed by the condition that

$$1+vR(0,0)=0\quad(\text{at } T=T_c),$$

where

$$R(0,0)=-\frac{3N}{4E_F}\left\{1-\frac{\pi^2}{12}\left(\frac{k_BT}{E_F}\right)^2\right\}$$

corresponds to the Pauli susceptibility of a degenerate electron gas. According to the analysis of low temperature specific heat, the effective mass of d-electrons in iron is about 12 times of free electron mass, and that in nickel is about 28. Then the constant B is 6.1×10^{12} for Fe and 5.0×10^{13} for Ni. The calculated B for Fe is in nice agreement with the value given by Lowde's experiment[4].

The critical scattering below the Curie point is also given by our treatment. The cross section in this case is expressed as

$$\frac{d\sigma}{d\Omega}=\left(\frac{g_0e^2}{m_0c^2}\right)^2|F(\kappa)|^2k_BT_c\left\{\frac{(1-\hat{\kappa}_z{}^2)C}{2B(T_c-T)+q^2}\right.$$
$$\left.+(1+\hat{\kappa}_z{}^2)\frac{C_1(T,q)}{q^2}\right\},$$

where

$$C_1(T,q)\begin{cases}=C, & \text{if } k_B\{(T_c-T)T_c\}^{1/2}\ll v_Fq\\ =\dfrac{N}{\omega_2}\propto\left(1-\dfrac{T}{T_c}\right)^{-1/2}, & \text{if } k_B\{(T_c-T)T_c\}^{1/2}\gg v_Fq.\end{cases}$$

In the above, ω_2 is the coefficient of the q^2 term in the spin wave spectrum; i.e., $\omega(q) = \omega_2 q^2 + 0(q^4)$.

At somewhat higher temperatures above the Curie point, the magnetic scattering is called the paramagnetic scattering. From the view-point of the localized spin model, the paramagnetic scattering is the scattering by nearly free paramagnetic ions. In the band model, however, there are no such paramagnetic ions. So it has been asserted that the observed paramagnetic scattering may be supposed to favour the localized spin model. It should be noted, however, that the temperatures of the experiments so far made are not high enough; the paramagnetic ions in the localized spin model cannot be regarded as free ions at such temperatures. Then the short range order effect is rather important. On account of this effect, our band theoretical calculation gives the paramagnetic scattering shown in Fig. 1.

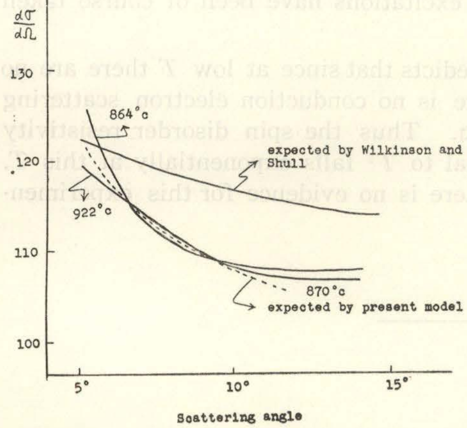

Fig. 1. Paramagnetic scattering for iron.

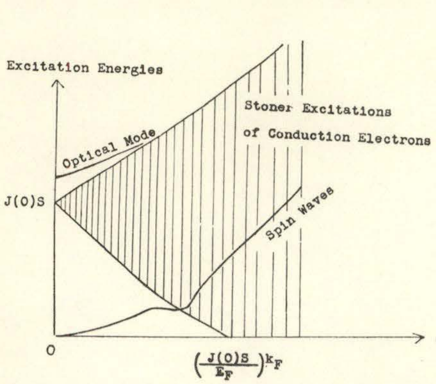

Fig. 2. Excitation energies of one-pair states generated by spin-flop processes.

2. Dynamical *s-d* Model

Magnetic properties of the *s-d* system may be represented in terms of $\bar{\chi}(\boldsymbol{q}, \omega)$ which is substantially equal to the dynamical susceptibility of the itinerant electrons. We have examined so far the spin waves in an *s-d* system. The results of our dynamical treatment differ from those of usual static one[5] mainly in the following points:

(i) The spin waves with wave numbers larger than $(J(0)S/E_F)k_F$ dissociate into Stoner excitations of conduction electrons[6]. Here, $J(q)$ is the Fourier component of the exchange integral between a conduction electron and a localized *d*-spin, and S is the magnitude of the total spin at a lattice site.

(ii) The frequency spectrum of the spin waves with wave numbers approximately equal to $(J(0)S/E_F)k_F$ departs appreciably from the q^2-law, though the departure depends sensitively on the magnitude and shape of $J(q)$.

(iii) An optical mode of spin density waves appears in addition to the usual spin waves described above.

References

1 R. J. Elliott and W. Marshall: Rev. Mod. Phys. **30** (1958) 75. W. Marshall: Lectures on Neutron Diffraction (Harvard Univ. 1959), unpublished notes. R. J. Elliott and R. D. Lowde: Proc. Roy. Soc. **A230** (1955) 46. R. D. Lowde: Proc. Roy. Soc. **A235** (1956) 305.

2 P. A. Wolff: Phys. Rev. **120** (1960) 814.

3 R. J. Elliott: Proc. Roy. Soc. **A235** (1956) 289.

4 R. D. Lowde: Rev. Mod. Phys. **30** (1958) 69.

5 T. Kasuya: Prog. Theor. Phys. **16** (1956) 45.

6 C. Kittel and A. H. Mitchell: Phys. Rev. **101** (1956) 1611. A. H. Mitchell: Phys. Rev. **105** (1957) 1439.

DISCUSSION

P. G. de Gennes: Would your results be qualitatively modified if the divergence of the Coulomb matrix element at small momentum transfers was taken into account?

T. IZUYAMA: The replacement of the Coulomb matrix element by the constant v may be regarded as an approximation for the Fourier component of the screened Coulomb interaction. If you use the original Coulomb matrix elements which are divergent at small momentum transfers, you must introduce polarization diagrams which lead to the screening for the long range part of the Coulomb force. We have introduced the screened Coulomb interaction in the original Hamiltonian and omitted the polarization diagrams in our calculation. Thus our calculation is consistent.

C. KITTEL: It would be interesting to examine the neutron scattering in the limit of zero effective mass: One might perhaps expect the diffraction to be largely inelastic in this limit.

D. KIM: We have not yet considered this limiting case.

R. J. ELLIOTT: Does your theory give a different answer to the usual one for the scattering of neutrons from the Stoner excitations?

T. IZUYAMA: Dr. Elliott already considered the Stoner excitations for the band theoretical interpretation of the ferromagnetic diffuse scattering. The effect of these excitations was found to be very small. Accordingly we have omitted these excitations in the calculation of the ferromagnetic diffuse scattering. In the other topics the Stoner excitations or the individual particle excitations have been of course taken into account.

R. J. ELLIOTT: The *s-d* interaction picture predicts that since at low T there are no spin waves of wave vector $k > (JS/E_F)\, k_F$ there is no conduction electron scattering between the Fermi surfaces of different spin. Thus the spin disorder resistivity which is, in the spin wave region, proportional to T^2 falls exponentially at this T. In Fe we once estimated this to be 5°K but there is no evidence for this experimentally.