D. Amit and E. P. Gross, Phys. Rev. 145, 130 (1966).
[2]G. W. Rayfield and F. Reif, Phys. Rev. 136, A1194 (1964).
[3]R. J. Donnelly, W. I. Glaberson, and P. E. Parks, Experimental Superfluidity (University of Chicago Press, Chicago, 1967); R. J. Donnelly and P. H. Roberts (to be published).
[4]W. I. Glaberson, D. M. Strayer, and R. J. Donnelly, Phys. Rev. Letters 20, 1428 (1968).
[5]J. L. Yarnell, G. P. Arnold, P. J. Bendt, and E. C. Kerr, Phys. Rev. 113, 1379 (1959).
[6]P. J. Bendt, R. D. Cowan, and J. L. Yarnell, Phys. Rev. 113, 1386 (1959).
[7]B. E. Springett, Phys. Rev. 155, 139 (1967). Springett has obtained the variation of μ_0 by estimating the curvature of the roton minimum reported by D. G. Henshaw and A. D. B. Woods, in Proceedings of the Seventh International Conference on Low Temperature Physics, Toronto, Canada, 1960, edited by G. M. Graham and A. C. Hollis Hallett (University of Toronto Press, Toronto, 1961), p. 539.
[8]C. Zipfel and T. M. Sanders, Jr., private communication.
[9]L. D. Landau and E. M. Lifshitz, Fluid Mechanics (Addison-Wesley Publishing Company, Inc., Reading, Mass., 1959), pp. 69-70.
[10]H. H. Tjerkstra, Physica 18, 853 (1952).
[11]H. E. Hall, Phil. Mag. Suppl. 9, 89 (1960).
[12]P. H. Roberts and R. J. Donnelly, to be published.
[13]S. V. Iordanskii, Zh. Eksperim. i Teor. Fiz. 48, 708 (1965) [translation: Soviet Phys.—JETP 21, 467 (1965).
[14]R. L. Douglas, Phys. Rev. 174, 255 (1968).

NEUTRON SCATTERING IN FERROMAGNETIC DILUTE ALLOYS

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A new basis for the analysis of the neutron elastic-diffuse-scattering data in ferromagnetic dilute alloys is formulated and Low and Holden's data on PdFe are analyzed from this viewpoint. We especially discuss the impurity-concentration dependence of the range of the conduction-electron spin polarization about the magnetic impurity.

From measurements of the neutron elastic diffuse scattering in ferromagnetic dilute alloys one obtains information concerning the spatial distribution of the conduction-electron spins as well as the spin on the impurity atom.[1,2] The alloys of Fe or Co in Pd are especially interesting since the conduction-electron spin polarization has been shown to have a very long range, ~10 Å.[3] This exceedingly long range of magnetic perturbation due to a magnetic impurity is attributed to the large exchange enhancement of the Pd matrix[4] and the interatomic nature of the electron-electron interaction.[5]

Another interesting aspect of the problem, although it has received less attention, is that the range of conduction-electron spin polarization around a magnetic impurity decreases sharply with increasing concentration of magnetic impurities, i.e., the neutron scattering experiment shows that increasing the Fe or Co concentration from ~0.5 to 4.0 at.% reduces the range from 10 to 1 Å.[3] In this Letter we present an analysis of the concentration dependence of the conduction-electron spin polarization in ferromagnetic dilute alloys. The basis of our analysis is as follows: In exchange enhanced metals, the spatial extent of the spin polarization around an impurity disturbance may decrease sharply as the spin splitting of the host matrix increases.[6] Increasing the impurity concentration in dilute ferromagnetic alloys leads to an increase in the spin splitting of the host matrix bands. We have derived an expression for the conduction-electron spin polarization which self-consistently includes the concentration dependence of the spin splitting of the host matrix and accounts for the sharp concentration dependence observed in the neutron-scattering experiments by Low and Holden.[3] In analyzing the neutron-scattering experiments we show that simple parabolic bands with 0.36 hole/atom[7] are a very poor approximation for the $3d$ hole bands of Pd. This is in agreement with recent band-calculation results.[8] Further, the fall-off with q of the susceptibility function for the actual band is expected to be significantly more rapid than that for a parabolic band.

The Fourier component of the conduction-electron spin polarization $\langle\sigma_z(q)\rangle$ $(q \neq 0)$ in terms of which the neutron scattering cross section will be described [see Eq. (6) below] is calculated starting with a finite number of magnetic impurities in the system. We obtain the following expression using the random phase approximation assuming the spins are ferromagnetically ordered

along the z axis:

$$\langle\sigma_z(q)\rangle = \tfrac{1}{2} l F_H(q) \frac{J(q)}{N} \langle S_z(q)\rangle \frac{\chi_{0+}^*(q) + \chi_{0-}^*(q) + 2U(q)\chi_{0+}^*(q)\chi_{0-}^*(q)}{1 - U(q)^2 \chi_{0+}^*(q)\chi_{0-}^*(q)}, \tag{1}$$

where

$$\chi_{0\sigma}^*(q) = \frac{\chi_{0\sigma}(q)}{1 - \mathcal{J}(q)\chi_{0\sigma}(q)} \quad (\sigma = + \text{ or } -). \tag{2}$$

In Eq. (1) the bands of the conduction electrons of the host metal are approximated by an l-fold degenerate band of the same structure, $J(q)$ is the s-d exchange integral, $F_H(q)$ is the form factor of the host matrix band, N is the total number of atoms in the system, $S_z(q) = \sum_i S_{iz} \exp(iqR_i)$, where S_i is the impurity Fe spin at the lattice point R_i, and $\chi_{0\sigma}(q)$ is the unenhanced longitudinal spin susceptibility of each $3d$ band:

$$\chi_{0\sigma}(q) = -\sum_K \frac{f_{k,\sigma} - f_{k+q,\sigma}}{\epsilon_{k,\sigma} - \epsilon_{k+q,\sigma}}, \tag{3}$$

where $\epsilon_{k,\pm} = \epsilon_k \mp [J(0)/N]\langle S_z(0)\rangle + U(0)n_\mp - \mathcal{J}(0)n_\pm$, and

$$f_{k\pm} = \{\exp[\beta(\epsilon_{k\pm} - \mu)] + 1\}^{-1}. \tag{4}$$

$U(q)$ is the Fourier component of the electron interaction (Coulomb repulsion) between electrons of opposite spins, including intra-atomic and interatomic and intraband and interband components, and $\mathcal{J}(q)$ is the electron interaction (exchange minus Coulomb) between electrons of parallel spins, again including intra-atomic and interatomic and intraband and interband components, n_σ is the number of conduction electrons with spin σ, and μ is the chemical potential.

Equation (1) is valid only for $q \neq 0$ and Eq. (4) is obtained for the $q = 0$ case. Equation (4) is the basis of the Stoner model. An important point to note about Eq. (1) is that here $\chi_{0\sigma}$ or $\chi_{0\sigma}^*$ is a function of the uniformly spin-split Fermi distribution function $f_{k\sigma}$. The fact that the more concentrated FePd system is ferromagnetic and the conduction-electron bands are spin split has not been taken into account properly in the usual formulation of the spin polarization problem where only a single impurity is introduced into the Pd matrix.[4,5] If we consider $\langle\sigma_z(q)\rangle$ in the one-impurity limit, Eq. (1) reduces to

$$g_H \mu_B \langle\sigma_z(q)\rangle = \frac{1}{g_H \mu_B} \frac{J(q)}{N} \langle S_z\rangle \left[l F_H(q) \frac{\tfrac{1}{2} g_H^2 \mu_B^2 \chi_0(q)}{1 - \{U(q) + \mathcal{J}(q)\}\chi_0(q)} \right], \tag{5}$$

where $\chi_0(q)$ is obtained from Eq. (3) by replacing the spin-dependent Fermi distribution function $f_{k\sigma}$ by the paramagnetic spin-independent Fermi distribution function f_k. Notice that the quantity in the bracket on the right-hand side of Eq. (5) is the paramagnetic susceptibility of the host matrix.

To analyze the neutron scattering data such as that of Low and Holden, we have derived a general form for the elastic diffuse scattering cross section:

$$\frac{d\sigma}{d\Omega} = \left(\frac{\gamma e^2}{2m_0 c^2} \right)^2 \sin^2\alpha \langle S_z\rangle^2 N_0 \left[g_I F_I(q) + g_H \frac{\langle\sigma_z(q)\rangle}{\langle S_z(q)\rangle} \right]^2 (1 - \sum_K \delta_{q,K}), \tag{6}$$

where in Eq. (6) the average is taken over a random distribution of impurities. [Notice that in Eq. (1) this average is not taken yet.] K is zero or the reciprocal lattice vector, g_I and g_H are, respectively, the g factor of the impurity and host-metal electron spin, $F_I(q)$ is the form factor of the impurity spin, $\langle S_z\rangle$ is the expectation of the z component of the individual impurity spin, N_0 is the number of the impurity, α is the angle between the z axis and the scattering vector of the neutron (Low and Holden normalized their data with $\sin^2\alpha = 1$), and $(\gamma^2 e/2m_0 c^2)^2 = 0.073$ b.

If we were to use Eq. (5) instead of Eq. (1) for $\langle\sigma_z(q)\rangle$ in Eq. (6), then $(d\sigma/d\Omega)/N_0$ would be independent of impurity concentration. This is quite

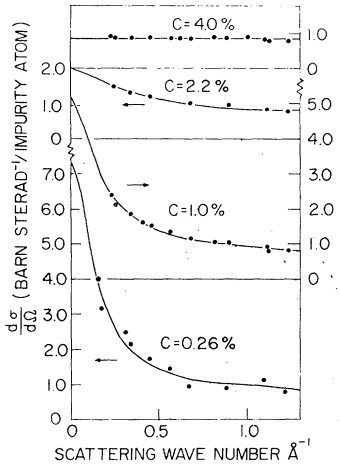

FIG. 1. The dots are the neutron-scattering data obtained by Low and Holden. The solid lines are a theoretical fit with $k_F = 0.5$ Å^{-1} and $B = 7$.

contrary to what is observed experimentally even for dilute alloys as shown in Fig. 1. To interpret the observed sharp changes in $d\sigma/d\Omega$ it has been suggested that in a system with a finite concentration of magnetic impurities, the long-range tails of the conduction-electron spin polarization would overlap. Accordingly, the conduction-electron spin polarization per impurity could decrease due to the nonlinearity of electron spin susceptibility caused by the saturation of the magnetization. This argument has not been developed quantitatively. The formulation of $\langle\sigma_z(q)\rangle$ and $d\sigma/d\Omega$ in this Letter, Eqs. (1) and (6), accounts for the nonlinearities of the conduction-electron spin polarization due to impurities in a self-consistent and quantitative manner. To reproduce the experimental data from Eqs. (1) and (6) for PdFe dilute alloys we adopt these additional simplifications: We assume three equally occupied parabolic d bands (corresponding to the three nonequivalent X points), and $g(q)$ is set equal to zero, since the magnitudes of the Coulomb and exchange integrals between electrons of the same spin are approximately equal. Thus $\chi_{0\sigma}{}^*(q)$ in Eq. (1) is simply $\chi_{0\sigma}(q)$. $U(q)$ is approximated by a Lorentzian $U(q) = U(0)(1+Bq^2/12k_F{}^2)^{-1}$, where B is chosen to fit the observed q dependence of the neutron scattering in the very low-impurity limit and remains constant for the more concentrated calculations. The g factors of the impurity and host-metal electrons are set equal to 2, $g_H = g_I = 2$. In the small q region in which we are interested, the q dependence of $F_H(q)$, $F_I(q)$, and $J(q)$ are neglected compared with that of $\chi_{0\sigma}(q)$ and $U(q)$ in the denominator of Eq. (1). If we are interested in the large q region, $F_H(q)$, $F_I(q)$, and $J(q)$ are most important and must be included.

All the parameters appearing in Eqs. (1) and (3) can be obtained from experimental data and are estimated as follows[9]: $J(0) = 0.15$ eV, $S = \frac{3}{2}$, and $N(0)U(0) = 0.9$, where $N(0) = m^*k_F/2\pi^2\hbar^2$ is the density of states of each of the three d bands of Pd at the Fermi surface, the total moment per Fe atom in the low-impurity limit is approximated by $10\mu_B$, and $k_F = 0.62\times10^8$ cm^{-1}, yielding the total number of holes/atom equal to 0.36.[7] The only parameter remaining to be determined is B which characterizes the range of the electron interaction $U(q)$. We choose B so as to reproduce the lowest concentration data (see the bottom curve in Fig. 1). We expect that the changes observed in $d\sigma/d\Omega$ for higher concentration data should be reproduced automatically simply by changing the impurity concentration in the calculation. With $k_F = 0.62\times10^8$ cm^{-1} and $B = 12$, the lowest concentration data can be satisfactorily reproduced; however, the sharp change in the scattering cross section as the Fe concentration increases from 0.26 to 4.0% is not realized. Namely, the theoretical relationship obtained for the impurity concentration c versus the variation of the neutron scattering cross section from Eq. (1) is about half as rapid as the experimental data. This difficulty can be resolved by considering the actual band structure of Pd more properly. Recent band calculations[8] indicate that the slope of the density of states at the Fermi surface is about twice as rapid as that which would be obtained in our parabolic approximation normalized to 0.36 holes/atom. Therefore, as far as the small impurity concentration region is concerned where the band structure near the Fermi surface is important, the actual bands would be better represented by a parabolic band with a smaller $k_F{}' = 0.62$ Å$^{-1}/2^{1/3} \sim 0.5$ Å^{-1}, and accordingly, a larger effective mass $m'^* = 2^{1/3}m^*$. These values for $k_F{}'$ and m'^* give the same density of states at the Fermi surface but have a slope twice as big as that with $k_F = 0.62$ Å. With this set of parameters and with $B = 7$, the observed relation between $(d\sigma/d\Omega)/N_0$ vs c is satisfactorily reproduced as is shown in Fig. 1.

One final point to be discussed is that to fit the data a rather large value for B was required. Since the magnitude of the interatomic electron interaction has been estimated to be about $\frac{1}{10}$ of the intra-atomic electron interaction,[10] B should be on the order of 1. The usual range parameter

κ which characterizes the large distance behavior of the spin polarization of the form $\exp(-\kappa r)/r$ is obtained by expanding Eq. (1) for small q. In the very low-impurity concentration limit $\kappa = \{12k_F^2[1-N(0)U(0)]/[1+N(0)U(0)B]\}^{1/2}$. A best fit to $d\sigma/d\Omega$ leads to $\kappa \sim 0.2$ Å^{-1}. (Note that this value is smaller than $\kappa = 0.3$ Å^{-1} obtained by Low and Holden in their analysis of their neutron scattering data.) The above expression for κ relies on the fact that for a parabolic band $\chi_0(q) \sim N(0)[1-\frac{1}{12}(q^2/k_F^2)]$ for small q. As indicated above, the actual bands in Pd are different from parabolic and more generally the small q expansion of $\chi_0(q)$ is $N(0)[1-(A/12)(q^2/k_F^2)]$ with $A \neq 1$. The expression for κ then becomes $\{12k_F^2[1-N(0)U(0)]/[A+N(0)U(0)B]\}^{1/2}$. In our analysis, as well as in others', a rather large value of B was required. The magnitude of B, however, can be reduced if $A > 1$, and we feel that this is the case in Pd.

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[1]G. G. Low and M. F. Collins, J. Appl. Phys. 34, 1195 (1963); G. G. Low, J. Appl. Phys. 39, 1174 (1968).

[2]W. Marshall, J. Phys. C: Phys. Soc. (London) Proc. 1, 88 (1968).

[3]G. G. Low and T. M. Holden, Proc. Phys. Soc. 89, 119 (1966); T. J. Hicks, T. M. Holden, and G. G. Low, J. Phys. Soc. C: Phys. Soc. (London) Proc. 1, 528 (1968).

[4]B. Giovannini, M. Peter, and J. R. Schrieffer, Phys. Rev. Letters 12, 736 (1964); T. Moriya, Progr. Theoret. Phys. (Kyoto) 34, 329 (1965).

[5]A. M. Clogston, Phys. Rev. Letters 19, 583 (1967).

[6]D. J. Kim and B. B. Schwartz, Phys. Rev. Letters 20, 201 (1968).

[7]J. J. Vuillemin and M. G. Priestley, Phys. Rev. Letters 14, 307 (1965).

[8]A. J. Freeman, A. M. Furdyna, and J. O. Dimmock, J. Appl. Phys. 37, 1256 (1966); A. Misetich and R. E. Watson, private communication; F. Mueller and A. J. Freeman, private communication.

[9]See, for instance, T. Takahashi and M. Shimizu, J. Phys. Soc. Japan 20, 26 (1965); D. J. Kim, Phys. Rev. 149, 434 (1966); S. Doniach and E. P. Wohlfarth, Proc. Roy. Soc. (London), Ser. A 269, 442 (1967).

[10]See, for instance, J. Hubbard, Proc. Roy. Soc. (London), Ser. A 276, 238 (1963); C. Herring, in Magnetism, edited by G. T. Rado and H. Suhl (Academic Press, Inc., New York, 1966), Vol. IV.