A NEW ANALYSIS OF NEUTRON DIFFRACTION IN FERROMAGNETIC METALS

D. Kim, B. Schwartz, H. Praddaude

A NEW ANALYSIS OF NEUTRON DIFFRACTION IN FERROMAGNETIC METALS

D. J. KIM, B. B. SCHWARTZ (*) and H. C. PRADDAUDE
Francis Bitter National Magnet Laboratory (**)
Massachusetts Institute of Technology Cambridge, Massachusetts 02139

Résumé.— En se basant sur le modèle d'échange s-d, on obtient une expression de la diffusion des neutrons dans les alliages ferromagnétiques dilués en fonction de la matrice des susceptibilités. En évaluant la section efficace, on étudie deux cas :

1° La levée de dégénérescence des bandes de spin de la matrice sous l'influence des impuretés et son effet sur la susceptibilité magnétique $\chi^{mm}(q)$.

2° La réponse des spins du potentiel de charge $\chi^{me}(q)$ à l'intérieur d'une sous-bande de spin.

Abstract. — Based on the s-d exchange model we derive a convenient expression for the magnetic elastic diffuse scattering of neutrons in ferromagnetic dilute alloys in terms of the host metal susceptibilities. In evaluating the cross-section we consider the following two points : (1) The spin splitting of the host metal band due to the impurities and its effect on the magnetic susceptibility $\chi^{mm}(q)$. (2) The spin response to charge potential $\chi^{me}(q)$ in a spin-split band.

I. Elastic diffuse scattering cross section. — An original and extensive theoretical discussion on the magnetic elastic diffuse scattering cross section of neutron due to the random distribution of magnetic impurities in a metal was given by Marshall [1, 2]. His model assumed a localized spin for both the host metal and the impurity. The s-d exchange model in which the host metal is represented by itinerant electrons whereas the impurity is represented by a localized spin is known to be convenient in discussing dilute magnetic alloys. In this paper we present an expression for the elastic diffuse scattering based on the s-d exchange model. Our expression is very useful since the scattering cross section is directly related to the host metal susceptibilities and band structure.

According to Marshall [1] the elastic diffuse scattering cross section is given by subtracting the Bragg scattering part from the total elastic scattering. The total elastic scattering cross section including the Bragg scattering is given by

$$\frac{d\sigma}{d\Omega} = \left(\frac{\gamma e^2}{2 m_0 c^2}\right)^2 (1 - k_z^2) < M^z(q) M^z(-q) > \qquad (1)$$

where $M^z(q)$ is the expectation value of the Fourier component of the magnetization density due to a given distribution of magnetic impurities and is obtained from the sum of the impurity magnetization and the conduction electron magnetization around each impurity in the unit of μ_β

$$M^z(q) = g_I F_I(q) \sum e^{iqR_i} S_i^z + \sigma_{mm}^z(q) + \sigma_{me}^z(q) \qquad (2)$$

where

$$\sigma_{mm}^z(q) = F_H(q) \frac{J(q)}{N} \frac{1}{g_H \mu_B^2} \chi^{mm}(q) \sum_i e^{iqR_i} S_i^z \qquad (3)$$

and

$$\sigma_{me}^z(q) = F_H(q) \frac{1}{\mu_B} \chi^{me}(q) [- V_0(q)] \frac{1}{S} \sum e^{iqR_i} S_i^z \qquad (4)$$

(*) Also Physics Department, M. I. T.

(**) Supported by the U. S. Air Force Office of Scientific Research.

and other notations are the same as ref. [1]. In the above we assumed the system is ferromagnetic and the magnetization is along the z axis. S_i^z is the spin of the i^{th} impurity and therefore in the ferromagnetic state $S_i^z = S$ on the impurity site and $S_i^z = 0$ elsewhere. We associate with the impurity not only a spin S, which produces a conduction electron spin magnetization $\sigma_{mm}^z(q)$ through the s-d exchange interaction, but also a charge potential $V_0(q)$ at each impurity site. In the ferromagnetic state the charge potential can also produce a spin polarization $\sigma_{me}^z(q)$ in the host metal through the non-diagonal susceptibility $\chi^{me}(q)$ [3, 4, 5]. $F_I(q)$ and $F_H(q)$ are respectively the form factors of the impurity and the host metal band, N is the total number of lattice points, $J(q)$ is the s-d exchange interaction constant, $\chi^{mm}(q)$ is the longitudinal magnetic susceptibility of the host metal and g_H and g_I are respectively the g-factors of the impurity and the host metal. In eq. (4) we used the convenient relation that $S_i^z/S = 1$ on the impurity site and 0 otherwise. The average $< \quad >$ is over the random distribution of the impurities.

From eq. (2), (3), and (4) we notice the Fourier component of the magnetization density due to the presence of the impurity can be written in a compact form

$$M^z(q) = A(q) \sum_i e^{iqR_i} S_i^z$$

where

$$A(q) = g_I F_I(q) + F_H(q) \frac{J(q)}{N} \frac{1}{g_H \mu_B^2} \chi^{mm}(q) + F_H(q) \frac{1}{\mu_B} \chi^{me}(q) \frac{- V_0(q)}{S}. \qquad (5)$$

With eq. (5), eq. (1) is rewritten in a simple form

$$\frac{d\sigma}{d\Omega_{elastic}} = \left(\frac{\gamma e^2}{2 m_0 c^2}\right)^2 (1 - k_z^2) A(q) A(-q) \times \left\langle \sum_{i,j} e^{iq(R_i - R_j)} S_i^z S_j^z \right\rangle. \qquad (6)$$

In eq. (6) the part of the scattering related to the distribution of the impurities is separated out in a very simple form, and therefore the Bragg reflection part can be unambiguously subtracted. If the concentration of the magnetic impurity is c, the Bragg scattering in eq. (6) comes from the average impurity magnetic moment cS at each lattice point. Subtracting the Bragg scattering from the total elastic scattering cross-section we obtain the cross-section of the elastic diffuse scattering,

$$\frac{d\sigma}{d\Omega_{\text{diffuse}}} = \left(\frac{\gamma e^2}{2 m_0 c^2}\right)^2 (1 - k_z^2)\, A(q)\, A(-q) \times \times \left\langle \sum_i \sum_j e^{iq(R_i - R_j)} (S_i^z - cS)(S_j^z - cS) \right\rangle . \quad (7)$$

The origin of the elastic diffuse scattering is very clear from eq. (7). The diffuse scattering is caused by the deviation of the magnetization from the uniform average magnetization cS at each lattice point. Approximations are introduced through the estimation of the quantity $A(q)$ and

$$T(q)_{\text{diffuse}} \equiv \left\langle \sum_i \sum_j e^{iq(R_i - R_j)} (S_i^z - cS)(S_j^z - cS) \right\rangle .$$

If the impurities form some specific type of clusters this will be reflected in $T(q)$ since it depends on the distribution of impurities. Here, however, we assume that the concentration of the impurities, c, is very small and the distribution is random. In that case $T(q)_{\text{diffuse}}$ is estimated as

$$T(q)_{\text{diffuse}} = c(1 - c)\, NS^2 . \quad (8)$$

In the calculation of eq. (8) the correlation of two impurities at different sites is approximated by assuming average uniform magnetization cS at each lattice site whereas the correlation at the same site is taken into account exactly. By inserting eq. (8) into eq. (7) we obtain

$$\frac{d\sigma}{d\Omega_{\text{diffuse}}} = \left(\frac{\gamma e^2}{2 m_0 c^2}\right)^2 \times (1 - k_z^2)\, Nc(1 - c)\, S^2\, A(q)\, A(-q) \quad (9)$$

where $A(q)$ is given by eq. (5).

II. **The spin and charge magnetic susceptibilities, $\chi^{mm}(q)$ and $\chi^{me}(q)$.** — To obtain $\sigma^z_{mm}(q)$, eq. (3), we usually calculate the host spin polarization for a single impurity and then sum it up independently. In this approximation $\chi^{mm}(q)$ appearing in eq. (3) is the magnetic susceptibility of the pure host, and therefore the scattering cross-section, eq. (9), normalized by the impurity concentration $c(1 - c)$ (.. c for $c \ll 1$) becomes independent of c. Low and Holden [6], however, showed that for Fe in Pd even in the low impurity concentration region of $c < 0.04$, the neutron scattering per impurity changes with impurity concentration. In order to explain this discrepancy, we made a new calculation of $\sigma^z_{mm}(q)$ using the Green's function method [7]. Our calculation self consistently accounts for the fact that there are many impurities and that the impurities are spin ordered. Therefore $\chi^{mm}(q)$ in eq. (3) is no longer the simple susceptibility of the pure host metal but the susceptibility of the host metal where the bands are spin split by the uniform part of the exchange field of the impurities. This leads to a c-dependence of $\chi^{mm}(q)$ and explains the concentration dependence of the neutron diffraction data of Fe in Pd. As we increase the Fe concentration the spin splitting of the Pd band increases and this results in a sharp decrease in the range of the conduction electron spin polarization.

A recent neutron scattering experiment by Cable and Hicks [8] on ferromagnetic Co with 5 at % of V, Cr, Mn or Ni can also be interpreted from the above concepts. The Ni impurity whose moment is parallel to that of host Co did not produce any disturbance on the surrounding Co atoms whereas Mn, whose moment is antiparallel to the magnetization of Co produced a long range disturbance on the surrounding Co. We believe the reason Mn can produce long range spin polarization is that Mn reduces the spin splitting of the host Co band.

Since Friedel's work [3] it has been known that in a spin split band, a charge potential can cause a spin polarization. It is due to the simple fact that in spin split bands different numbers of electrons for up and down spins are available for the screening of the impurity charge. Actually in a number of ferromagnetic metal based alloys, the spatial behavior of the spin response of the host metal cannot be understood simply from $\sigma^z_{mm}(q)$, and $\sigma^z_{me}(q)$ seems to be playing an important role [2]. In order to estimate the effects of $\sigma^z_{me}(q)$, we extended Friedel's original consideration to the general q case and obtained the magnetic response to a charge, $\chi^{me}(q)$, in the same approximation used to obtain $\chi^{mm}(q)$. On the basis of this result we can interpret some of the previously not understood characteristics of the data [5]. For instance, the anomalous dependence of the spatial dependence of the spin polarization on the relative position of the impurity on the period table either left or right of the ferromagnetic host metal can be explained by accounting for both the spin and charge disturbance associated with the impurity.

References

[1] MARSHALL (W.), *J. Phys. C (Proc. Phys. Soc.)*, 1968, **1**, 88.

[2] For an extensive review of experimental data see LOW (G. G.), *Advances in Physics*, 1969, **18**, 371.

[3] FRIEDEL (J.), *Nuovo Cimento*, 1958, Suppl. **2**, 287.

[4] RAJAGOPAL (A. K.), BROOKS (H.) and RANGANATHAN (N. R.), *Nuovo Cimento*, 1967, Suppl. **5**, 807.

[5] KIM (D. J.), PRADDAUDE (H. C.) and SCHWARTZ (B. B.), *Phys. Rev. Letters*, 1969, **23**, 419.

[6] LOW (G. G.)) and HOLDEN (T. M.), *Proc. Phys. Soc.*, 1966, **89**, 119.

[7] KIM (D. J.) and SCHWARTZ (B. B.), *Phys. Rev. Letters*, 1968, **21**, 1744.

[8] CABLE and (J. W.) HICKS (T. J.), *J. Appl. Phys.*, 1970, **44**, 870.