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THE ANALYSIS OF MAGNETIC NEUTRON SCATTERING DATA

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ABSTRACT

The determination of the proper magnetic form
factors for use in the analysis of diffuse-scattering
data is discussed, and that information which is
immediately available from such data is considered.
Apparent discrepancies between the results of diffuse
scattering and diffraction experiments on ferromag-
netic Ni-Cu alloys are resolved. It is shown that the
data indicate that the negative spin density usually
attributed to conduction electrons instead arises
largely from the overlap of localized wavefunctions,
as was first suggested by Moon.

INTRODUCTION

In principle, elastic neutron scattering offers
by far the most complete experimental information on
the spatial distribution of the magnetization in mag-
netic materials. Yet, despite the pioneering work of
Marshall, et al.,[1] the extraction of theoretically
meaningful information from diffuse elastic neutron-
scattering data has remained a difficult, not com-
pletely solved problem. This is illustrated by the
recent appearance of apparent major inconsistencies
between the results of diffuse neutron-scattering
experiments[2,3] and other experiments[3,4] for ferro-
magnetic Ni-Cu alloys. We discuss here the determina-
tion of the proper magnetic form factors, $f(\vec{K})$, for
use in the analysis of diffuse-scattering data, and
consider that information which is immediately
available from such data in the limits of small and
large momentum transfers. Upon explicit consideration
of the diffuse elastic scattering[2,3,5] and diffrac-
tion[4] results for Ni-Cu alloys, the apparent incon-
sistencies are found to disappear under proper
analysis. Space limitations prevent our giving here
more than a few simple results; derivations and a
complete set of formulas will appear elsewhere.

NEUTRON DIFFRACTION

Neutron diffraction measures the Fourier transform,

$$\tilde{M}(\vec{G}) = N^{-1} \int M(\vec{r}) \; \exp(i\vec{G}\cdot\vec{r}) \; d^3\vec{r}, \qquad (1)$$

of the magnetization density at the reciprocal lattice vectors, $\vec{G}$, where N is the number of unit cells. By the definition of $\vec{G}$, this is just the Fourier transform of the average magnetization density of a unit cell, which is usually assumed to be given by

$$\langle M_n(\vec{x}) \rangle \simeq \langle \mu_n^{loc}(\vec{x}) \rangle + \mu^{cond}. \qquad (2)$$

Here, $\mu_n^{loc}(\vec{x})$ is the localized atomic magnetization density of the atom(s) in the nth cell, and μ^{cond} is the average conduction-electron magnetization density. One can define an average magnetic form factor,

$$f(\vec{G}) = \langle \tilde{M}_n(\vec{G}) \rangle / \langle M_n \rangle, \qquad (3)$$

where $M_n = \tilde{M}_n(0)$ is the total magnetization of the nth cell. Then, from Eq. (2) one finds

$$f(\vec{G}) \simeq (1+\alpha) f^{loc}(\vec{G}) - \alpha \delta_{\vec{G}0}, \qquad (4)$$

where

$$f^{loc}(\vec{G}) = \langle \tilde{\mu}_n^{loc}(\vec{G}) \rangle / \langle \mu_n^{loc} \rangle. \qquad (5)$$

Equation (4) allows one to determine α and $f^{loc}(\vec{G})$ from the experimentally determined $f(\vec{G})$. For elements and for alloys in which only one chemical species is magnetic, theoretical calculations generally are in excellent agreement with the values of $f^{loc}(\vec{G})$ obtained from Eq. (4), thus supporting the use of Eqs. (2) and (4) and allowing one to determine $f^{loc}(\vec{K})$ for all $\vec{K}$.

DIFFUSE ELASTIC SCATTERING

The diffuse elastic scattering of neutrons offers information on the Fourier components, $\tilde{M}(\vec{K})$, of the magnetization of disordered magnetic materials for all wavevectors away from the Bragg peaks. The use of spin-polarized neutrons enables one to measure directly the correlation function $M(\vec{K})$, which for a binary alloy is given by

$$\tilde{M}(\vec{K}) = [Nc(1-c)]^{-1} \sum_{m,n} \langle (p_m-c)\mu_n(\vec{K}) \rangle \exp(i\vec{K}\cdot\vec{R}_{nm}), \qquad (6)$$

where c is the impurity concentration, p_m is the impurity occupation number of site m, and $\vec{R}_{nm} = \vec{R}_n - \vec{R}_m$.

The analysis of $M(\vec{K})$ is especially simple in the limits of small and large momentum transfers. An exact limiting point for $M(\vec{K})$ is provided by the relationship

$$\lim_{K \to 0} \{M(\vec{K})/S(\vec{K})\} = d\langle\mu\rangle/dc, \qquad (7)$$

where $S(\vec{K})$ is the Fourier transform of the Cowley short-range-order parameters. In the opposite limit

$$M(\vec{K}) = \langle\mu(\vec{K})\rangle_{imp} - \langle\mu(\vec{K})\rangle_{host} + \delta M(K), \qquad (8)$$

where the subscripts "imp" and "host" denote averages over impurity atoms and host atoms, respectively, and where $\delta M(\vec{K})$ is an oscillatory, rapidly decreasing function of K.

Unfortunately, until recently diffuse elastic scattering experiments have been performed only with unpolarized neutrons and multicrystalline scatterers. Thus, they have measured directly only the spherical averages of $S(\vec{K})$ and of the function

$$T(\vec{K}) = [Nc(1-c)]^{-1} \sum_{m,n} [\langle\mu_m(\vec{K})\mu_n(\vec{K})\rangle - \langle\mu(\vec{K})\rangle^2] \exp(i\vec{K}\cdot\vec{R}_{nm}), \qquad (9)$$

not $M(\vec{K})$. In general, one may write

$$T(\vec{K}) = M^2(\vec{K})/S(\vec{K}) + \delta T(\vec{K}), \qquad (10)$$

where $\delta T(\vec{K})$ contains small linear terms plus complex nonlinear terms which are very difficult to evaluate numerically. These terms render the accurate determination of $M(\vec{K})$ next to impossible for alloys in which μ_n depends very nonlinearly on its chemical environment. The usual procedure for finding $M(\vec{K})$ is poor for large K, as is discussed in reference 5, and for some systems is poor even for small K.

APPARENT INCONSISTENCIES

In previous analyses of diffuse elastic-scattering data the form factor $f^{loc}(\vec{K})$ has been used, and it has been assumed that some part of the total magnetization arises from a conduction-electron spin polarization not seen by the neutrons. Thus, the extrapolated K=0 limit of $M(K)/S(K)$ should have given $d\langle\mu^{loc}\rangle/dc$ rather than the bulk magnetization values $d\langle\mu\rangle/dc$ typically found. For the case of ferromagnetic Ni-Cu alloys such an analysis[3] gave the surprising result that $\mu^{cond} = \langle\mu\rangle - \langle\mu^{loc}\rangle$ is very nearly equal to -0.10, independent of Cu concentration over the range $0 \leq c \leq 0.4$, assuming $\langle\mu\rangle_{Cu} = 0$. Thus, the data appeared to be internally self-consistent. However, more recent neutron-diffraction studies indicate that μ^{cond} decreases with increasing

Cu concentration even more rapidly than does $\langle\mu\rangle$. This is in substantial disagreement with the result inferred from both the low- and high-K limits of the diffuse-scattering data. The disagreement found is too large to be explained either by possible experimental errors or uncertainties or by nonlinear terms *not included in the Marshall[1] formalism.*

RESOLUTION OF INCONSISTENCIES--THE PROPER FORM FACTOR

Although only the local atomic magnetization, $\langle\mu^{loc}\rangle$, is seen in neutron diffraction, it is clear that the total moment associated with the site n in an alloy must be fairly well localized and should be seen in small-angle diffuse scattering. It is improper to ascribe any part of the bulk magnetization of a magnetically inhomogeneous alloy to a uniform conduction-electron spin polarization not seen by small-angle diffuse scattering or to use a purely local atomic form factor in analyzing diffuse-scattering data. Either a conduction-electron spin polarization induced by the local atomic part of μ_n or, as was first suggested by Moon,[6] overlap bonding or antibonding spin densities can give rise to a "nonlocal" contribution to μ_n having a form factor, $f^{nonloc}(\vec{K})$, which is large for small values of K but is very small at the Bragg peaks. This is confirmed by rough calculations for Ni alloys, which also indicate that induced conduction-electron spin densities and d-band overlap spin densities yield very similar form factors, as is shown[7] in Fig. 1.

The *proper form factor* for use in the analysis of diffuse-scattering data can be written as

$$f(\vec{K}) = (1+\alpha)f^{loc}(\vec{K}) - \alpha f^{nonloc}(\vec{K}), \qquad (11)$$

where α is determined by Eq. (4), as is discussed above in the section on neutron diffraction. Since α typically is small and since $f^{nonloc}(\vec{K})$ is rather insensitive to its method of calculation, the proper $f(\vec{K})$ is easily determined to order one or two per cent. The proper f(K) for Ni-Cu alloys is compared in Fig. 2 with $f^{loc}(K)$ as determined by Mook[8] and with that f(K) which would be produced by a consistent or very long-range conduction-electron spin polarization.

The use of the correct form factor for Ni-Cu alloys should lead to the satisfaction of Eq. (7) with $\langle\mu\rangle$ given by the bulk magnetization value and to the result $(1-c)M(\vec{K})/f(\vec{K}) \rightarrow \langle\mu\rangle$ in the limit of large momentum transfers. Since $f(\vec{K})$ approaches $f^{loc}(\vec{K})$ as K approaches zero, Eq. (7) is properly satisfied. An improved analysis of the old diffuse-scattering data[2,3] gives a satisfactory value of $\langle\mu\rangle$. The results of

recent polarized-scattering experiments[5] give excellent agreement with the bulk magnetization values of both $<\mu>$ and $d<\mu>/dc$; the data is sufficiently good to lend quantitative support to our choice of $f(\vec{K})$. The correct choice of form factor and correct analysis of large-angle scattering data removes all inconsistencies.

In principle, the concentration dependence of α can be used to judge the relative importance of conduction-electron and d-d overlap contributions to the nonlocal part of $<\mu>$. If $<\mu^{nonloc}>$ were due to conduction-electron spin polarization, to first order α should be independent of c. On the other hand, if it were due to d-d overlap it should depend on the number of host-host bonds and hence on c according to the formula

$$\alpha_{eff} \cong \alpha_1 - c(\alpha_1-\alpha_2)[1 + (1-c)g_1/<\mu>_{host}], \qquad (12)$$

where $<\mu>_{imp}$ is assumed to be zero, α_1 and α_2 are independent of c and measure host-host and impurity-host bonding, respectively, and $g_1 \cong -.035$ is defined by Marshall[1] and is known experimentally. For Ni-Cu alloys, one may assume $\alpha_2 \cong 0$; Fig. 3 compares the prediction of Eq. (12) for Ni-Cu alloys with the values of α given by diffraction experiments.[4] The experimental values support the point of view of Moon, but better experimental results clearly are desirable.

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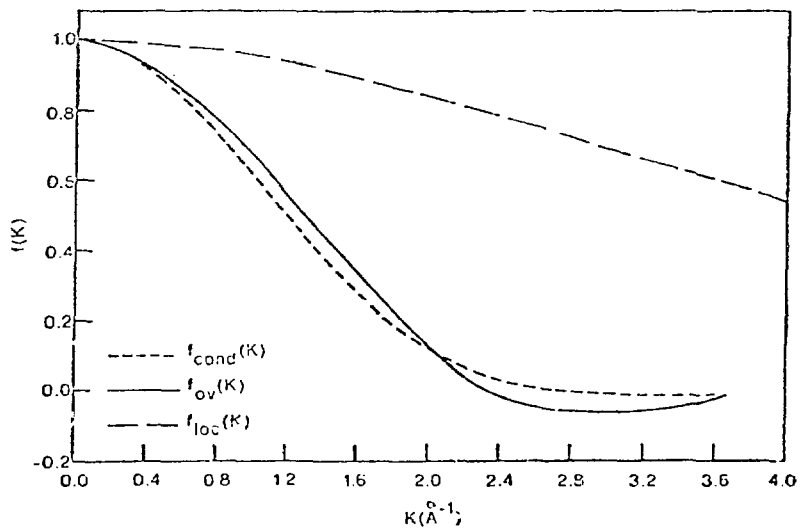8. H. A. Mook, Phys. Rev. 148, 495 (1966).

Figure 1. Partial form factors for Ni-Cu alloys: f(K)
for an induced conduction-electron polarization, for
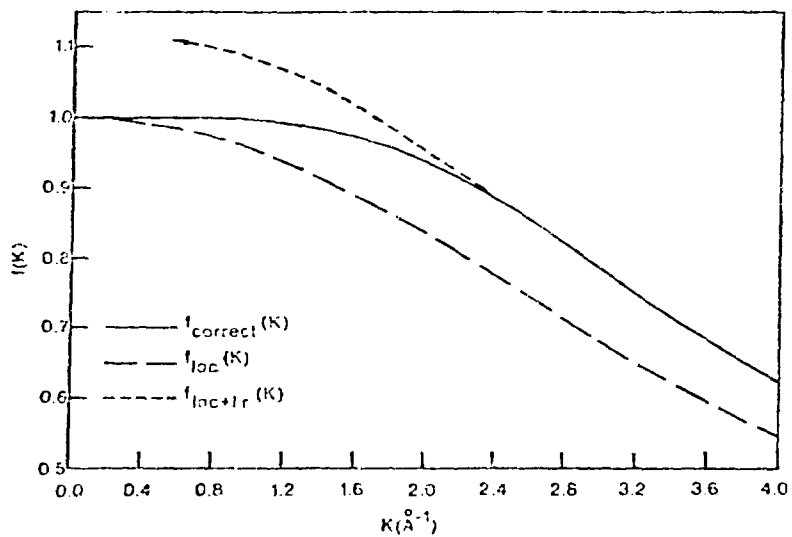the Ni-Ni overlap spin density, and for the local spin,

Figure 2. Form factors for Ni-Cu alloys: the correct f(K), the local or Mook f(K), and the f(K) appropriate to a local plus a long-range conduction-electron spin.
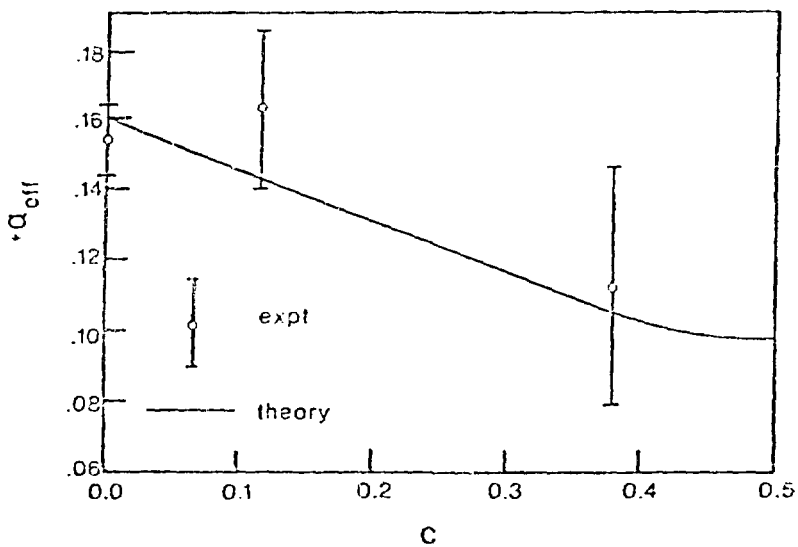
Figure 3. The ratio, α_{eff}, of the delocalized part of $\langle\mu\rangle$ to the total $\langle\mu\rangle$ for Ni-Cu alloys from diffraction experiments and from Eq. (13), based on ref. 6.