Magnetic form factor and spin-density asphericity of Ni-Cu

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Magnetic form-factor measurements were made on single crystals of a Ni-Cu alloy containing 47.6 at.% Cu. The results give a magnetic moment of 0.088 μ_B/atom at $T=4.2$ K and $H=40$ kOe in excellent agreement with bulk magnetization data. Despite this order of magnitude decrease in moment relative to Ni, the form factor is the same in both the radial distribution and the asphericity as that previously observed for pure Ni. This result is in strong contrast with an early coherent potential approximation calculation which yields a continuous variation of γ, the fractional E_g population, with Cu content. It is suggested that some of this discrepancy may be attributed to the neglect of local environment effects, which are known to be important for the magnetism of Ni-Cu alloys.

INTRODUCTION

The best experimental information available regarding d-electron wave functions in transition metals is obtained from magnetic form-factor data. The information gained from these data includes both the radial distribution and the symmetry character of the spin density. Furthermore, the data obtained by use of polarized neutron methods are sufficiently precise to provide a sensitive test for current band-structure calculations. For example, the excellent energy-band calculation by Wang and Callaway[1] for ferromagnetic Ni yields reasonable agreement with the experimental results for the magnetic moment, the Fermi surface, and the radial distributions of the charge and spin densities. However, this calculation fails to reproduce the observed asphericity of the spin density and therefore needs additional refinement.

In an alternative approach, the observed asphericity can be used as an input parameter for these calculations. Several years ago Cooke[2] formulated an energy-band treatment that included a band and wave-vector dependence for the interaction matrix elements. In this treatment, which has been highly successful in the calculation of the dynamic susceptibilities of Fe and Ni,[3] the interaction matrix is adjusted to reproduce the observed asphericity and magnetic moment.

The asphericity of the spin density is clearly an important parameter that is directly dependent on the details of the band structure. Any changes in the band structure of a metal caused by alloying should therefore be observable in the asphericity. The largest changes in this symmetry character should occur for alloy systems with sharp density-of-states features near the Fermi energy, such as those associated with Ni-based alloys. Of these, the most unambiguous results are obtained with nonmagnetic diluents for which the Ni form factor can be isolated and studied as a function of dilution. Some of the alloy systems that fall within this category, and for which form-factor data are available, are the Ni-based alloys with Ti, V, Cr, and Cu diluents. Polarized-neutron, diffuse-scattering data[4-7] for these alloy systems show that all of the ferromagnetic moment is located at the Ni sites. The experimental observations[8-13] and theoretical calculations[5,11,14] of the asphericity for these alloy systems are summarized in Fig. 1. Here, the data points are the experimental form factor results, and the curves represent the results of coherent-potential approximation (CPA) calculations.

Figure 1(b) shows the asphericity results for the Ni-Ti, Ni-V, and Ni-Cr alloys. The average ferromagnetic mo-

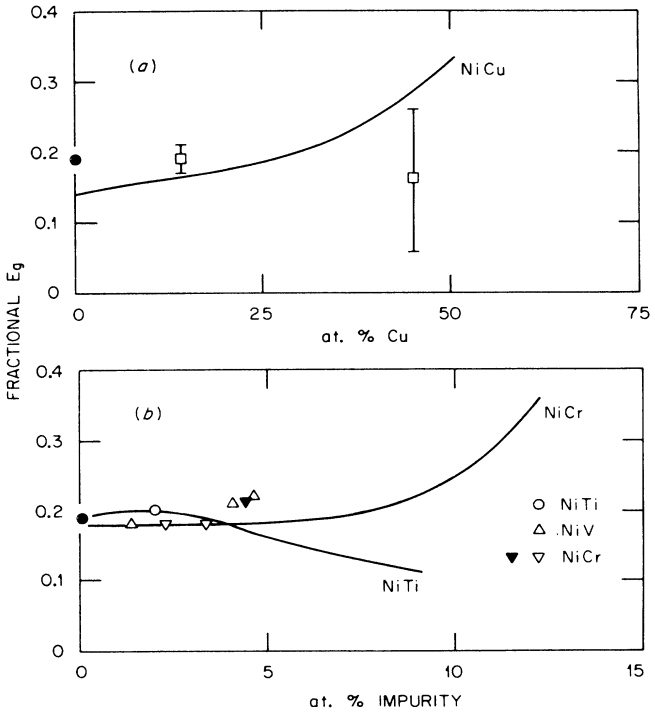

FIG. 1. Concentration dependence of the fractional E_g population for Ni-based alloys. The data points are taken from ●: Ni (Ref. 15), ⊡: Ni-Cu (Ref. 13), ⊙: Ni-Ti (Ref. 8), △: Ni-V (Ref. 8), ▼: Ni-Cr (Ref. 10), and ▽: Ni-Cr (Ref. 11). The solid curves are results of CPA calculations for Ni-Cu (Ref. 14), Ni-Ti (Ref. 15) and Ni-Cr (Ref. 11).

ment (and the precision of the magnetic form-factor measurement) decreases very rapidly with dilution for these alloys and, as a result, form-factor data are available for a limited concentration range. Even though the observations are consistent with the calculated concentration dependence, they do not extend to high enough concentrations to provide an adequate test for the theory. The Ni-Cu results shown in Fig. 1(a) are also consistent with the calculation, but the data at high Cu content are not sufficiently precise to test the theory. In the present experiment, we attempted to provide a better test by performing a precise measurement of the magnetic form factor and asphericity for a Ni-Cu alloy in the high Cu concentration region.

EXPERIMENTAL PROCEDURE

The magnetic form factor measurements were made on the HB-1 polarized-neutron spectrometer located at the High Flux Isotope Reactor in Oak Ridge. The data were taken with a vertical magnetic field of 40 kOe applied to the sample, which was held at 4.2 K in a cryomagnet. A neutron wavelength of $\lambda = 0.826$ Å was used to obtain the flipping ratios for the first 13 Bragg reflections with scattering vectors perpendicular to a [110] direction. Standard corrections were applied for sample depolarization and for the polarizing and flipping efficiencies of the spectrometer.

Extinction corrections were made by assuming a linear dependence on intensity. Specifically, we used $R = R_0[1 - a(R-1)I]$, where R and R_0 are the observed and corrected flipping ratios, and I is the intensity. The extinction parameter a was determined from flipping ratio measurements on the first 5 Bragg reflections from 3 different size crystals cut from the same region of the original single-crystal ingot. Crystals I and II were pillar shaped with dimensions of $0.5 \times 1 \times 10$ mm^3 and $1 \times 2 \times 10$ mm^3, respectively, while crystal III was in the shape of a disk 1 mm thick and 10 mm in diameter. Consistent values for the extinction parameter were obtained for these five reflections and the average was used to correct the remaining flipping ratios for the outer reflections. The maximum extinction corrections were 4%, 15%, and 50% in R-1 for crystals I, II, and III, respectively. The data for crystal III were rejected from further consideration because of the large extinction correction. Fully corrected flipping ratio data for the other two crystals are presented in Table I. Here, the numbers enclosed by parentheses indicate the errors in counting statistics only. These are consistent with the observed variations in R except for the case of the (111) reflection. Simultaneous reflection effects may be responsible for the additional uncertainty in this data point.

TABLE I. Corrected flipping ratios for crystals I and II and the average $\mu f(K)$ values.

hkl	$\frac{\sin\theta}{\lambda}$	R_{I}	R_{II}	$\mu f(K)$
111	0.243	1.0791(6)	1.0861(5)	0.0668
200	0.281	1.0760(8)	1.0732(5)	0.0605(4)
220	0.397	1.0477(7)	1.0484(5)	0.0395(4)
311	0.466	1.0330(6)	1.0347(5)	0.0280(4)
222	0.487	1.0332(7)	1.0342(5)	0.0279(4)
400	0.562		1.0168(5)	0.0140(4)
331	0.612		1.0190(5)	0.0158(4)
422	0.688		1.0119(5)	0.0100(4)
511	0.730		1.0046(5)	0.0039(4)
333	0.730		1.0111(5)	0.0093(4)
440	0.795		1.0062(7)	0.0052(6)
442	0.843		1.0054(7)	0.0045(6)
600	0.843		0.9972(8)	−0.0024(7)

RESULTS AND DISCUSSION

The magnetic scattering amplitudes, $p = 0.2695 \mu f(K)$, were determined from the averaged flipping ratios, $R_0 = (b+p)^2/(b-p)^2$, using nuclear amplitudes of 1.03×10^{-12} cm for Ni and 0.772×10^{-12} cm for Cu. Here, μ is the average ferromagnetic moment per alloy atom and $f(K)$ is the magnetic form factor. The $\mu f(K)$ are given in column 4 of Table I in units of μ_B/atom. Here, the error in μf for the (111) reflection has not been included because of the additional, nonstatistical uncertainty in this data point. The angular variation in $\mu f(K)$ is quite similar to that observed by Mook[15] for pure Ni, except that the alloy results are smaller in magnitude. Division of the $\mu f(K)$ by the Ni form-factor values [corrected to $0.579 \mu_B$/atom at $T = 295$ K (Ref. 16)] yields an average moment of $0.088(1) \mu_B$/atom for the alloy. This agrees perfectly with the magnetization obtained by interpolation of the spontaneous magnetization data of Kouvel and co-workers[17,18] with a subsequent 5% correction for an applied field of 40 kOe.[19]

The magnetic form factor of the Ni–47.6 at.% Cu alloy is compared with that of pure Ni in Fig. 2. The two are identical within experimental error. This form factor can be written as

$$f = \frac{2}{g} f_{\mathrm{spin}} + \frac{g-2}{g} f_{\mathrm{orbit}} , \tag{1}$$

with

$$f_{\mathrm{spin}} = \langle j_0 \rangle + (\tfrac{5}{2}\gamma - 1) A_{hkl} \langle j_4 \rangle , \tag{2}$$

and

$$f_{\mathrm{orbit}} = \langle j_0 \rangle + \langle j_2 \rangle . \tag{3}$$

Here g is the gyromagnetic ratio, γ is the fractional E_g population, A_{hkl} is a directionally dependent factor, and the $\langle j_n \rangle$ are integral functions related to different components of the spin density. The $\mu f(K)$ data in Table I were fitted to Eq. (1) using $g = 2.27$ as obtained from magnetomechanical data[20] and with $\langle j_n \rangle$ functions extracted from the Ni form-factor data[15] by use of the projection operator method.[21] The fitting parameters obtained are $\mu = 0.088(1) \mu_B$/atom and $\gamma = 0.18(1)$. The moment is the same as that obtained using the experimental Ni $f(K)$ values directly, and γ is the same, within experimental error, as that obtained for pure Ni.

We find, then, that the magnetic form factor of the alloy remains unchanged from that of pure Ni, even though enough Cu has been added to approach the critical composition, and the average moment has decreased by an or-

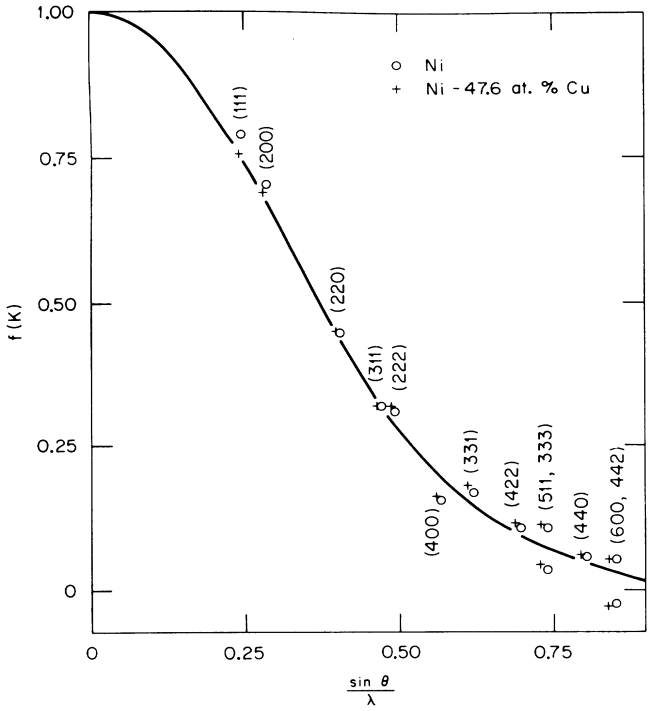

FIG. 2. Comparison of the magnetic form factors of pure Ni and Ni–47.6 at.% Cu. The solid curve is the spherical form factor, $f_{\text{spher}}=\langle j_0\rangle+(g-2)/g\langle j_2\rangle$, with $g=2.27$ and with $\langle j_0\rangle$ and $\langle j_2\rangle$ taken from the Ni data (Ref. 15) using the projection operator method (Ref. 21).

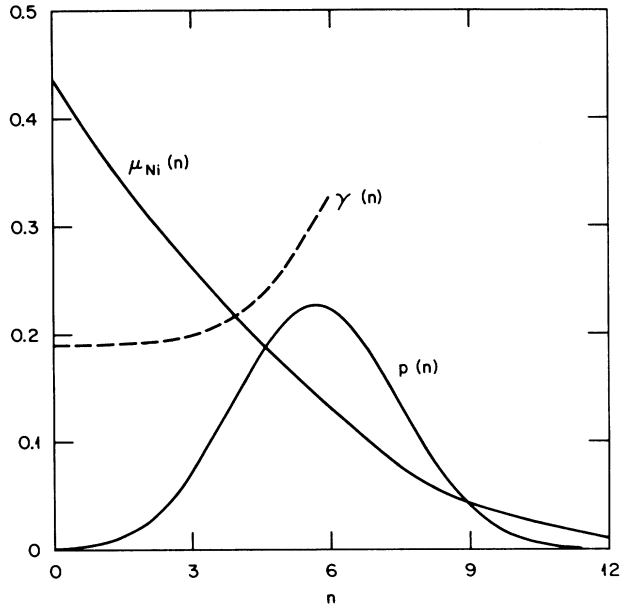

FIG. 3. The effect of local environment on γ. $P(n)$ is the probability that a Ni atom has n Cu nearest neighbors for $c=0.476$, $\gamma(n)$ is a schematic representation based on both theory and experiment, and $\mu_{\text{Ni}}(n)$ is the calculated (Ref. 22) Ni moment for each local environment. Weighting of $\gamma(n)$ by $\mu_{\text{Ni}}(n)P(n)$, as in Eq. (4), shifts $\langle\gamma\rangle$ toward lower n.

der of magnitude. This contrasts strongly with the results[14] of the CPA calculation, which are shown in Fig. 1(a), and for which γ varies continuously in the zero to 50% Cu region. Some of this lack of agreement may arise from the neglect of local environment effects in this single-site CPA calculation which uses the average chemical environment for each site. Magnetic disorder scattering measurements[7] as well as cluster-CPA calculations[22] show the importance of local environment for the magnetism of Ni-Cu alloys. The effect of local environment on the asphericity of the spin density is illustrated by Fig. 3. Here, $\mu_{\text{Ni}}(n)$ is the magnetic moment at a Ni site with n Cu nearest neighbors at a fixed Cu concentration of 48%. This is taken from Hamada's cluster-CPA calculation[22] and represents the effect of environment on the moment. $P(n)$ is the probability that a Ni atom has n Cu nearest neighbors which, for this fcc lattice, is given by

$$P(n)=[12!/n!(12-n)!]c^n(1-c)^{12-n}\ ,$$

where c is the Cu concentration. $P(n)$ for $c=0.476$ is shown in Fig. 3. Hamada also obtained the local density of states for Ni atoms in different local environments and at different concentrations. The shapes of these density-of-states curves and the positions of the Fermi energy for different n suggest that the asphericity parameter should also be a function of n. The dashed curve in Fig. 3 represents a schematic $\gamma(n)$ dependence based on $\gamma(0)\simeq 0.19$ for pure Ni and $\gamma(6)\simeq 0.32$ from Sacchetti's calculation[14] for Ni–50% Cu. The average asphericity is

$$\langle\gamma\rangle=\frac{\sum_n P(n)\mu_{\text{Ni}}(n)\gamma(n)}{\sum_n P(n)\mu_{\text{Ni}}(n)}\ , \tag{4}$$

and, of course, the single-site and cluster calculations take different averages. The single-site calculation considers only the average environment which at 50% Cu corresponds to $n=6$ for $\gamma(n)$. For the cluster or local environment model, the strong n dependence of $\mu_{\text{Ni}}(n)$ shifts the weighting toward smaller n and a lower value for $\langle\gamma\rangle$. However, a less pronounced $\gamma(n)$ dependence than that sketched in Fig. 3 is required to obtain agreement with the present results.

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