GIANT MOMENTS IN Ni-Cu ALLOYS NEAR THE CRITICAL COMPOSITION

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Neutron scattering experiments reveal that in weakly ferromagnetic Ni-Cu alloys near the critical composition the low-temperature spontaneous magnetization is inhomogeneously distributed in magnetic polarization clouds of large total moment (over $8\mu_B$) extending over many atoms.

Certain alloys near the critical composition for ferromagnetism can be regarded as itinerant electron systems whose exchange enhancement is close to the threshold of critical instability.[1] This viewpoint has been supported by susceptibility and other experimental evidence on Pd-Ni[2] and Ni-Rh[3] solid solutions and, most recently, on near-stoichiometric Ni_3Al and Ni_3Ga.[4] An equally promising candidate is the fcc Ni-Cu system, which we have chosen to study by neutron diffraction as well as bulk magnetic measurement. Our original plan was ultimately aimed at measuring the diffuse neutron scattering from Ni-Cu alloys doped with a localized-moment impurity such as Mn. By analogy to earlier results on Fe-doped Pd,[5] these measurements might be expected to reveal magnetic polarization clouds centered at the Mn impurity atoms and extending a large distance into the exchange-enhanced Ni-Cu matrix. Indeed, our current measurements are confirming that precisely this situation exists in Mn-doped Ni-Cu alloys.

However, this Letter is concerned with what we discovered about the Ni-Cu binaries when we first studied them in the pure, undoped state. As will be described below, the weakly ferromagnetic Ni-Cu alloys, by themselves, develop magnetization clouds of considerable strength and size.

In view of these results, a statement must be made about the metallurgy of Ni-Cu pertinent to our alloy samples. Recent neutron-diffraction work[6] has shown that Ni-Cu alloys develop a significant amount of chemical clustering even when quenched from high temperatures but that the clustering correlation appears to be essentially restricted to that between nearest-neighbor atoms. This clustering effect, now confirmed by our own neutron experiments, is much too short-ranged to be responsible for the extended magnetization clouds that we have observed. Another potential problem is the gross chemical inhomogeneity associated with the dendritic structure of a cast alloy. To break up such structure our Ni-Cu ingots, prepared by induction melting and chill casting under argon, were very severely cold-rolled before being cut into shape. The samples were then annealed for 3 days at 1000°C and water quenched; their compositions were 50-, 48-, 46-, 44-, 42-, and 40-at.% Ni with an impurity content (mostly Fe) of ~40 ppm. For the samples thus prepared, our magnetization data gave no indication of any gross variation in composition.

The Curie points (T_c) and spontaneous moments per atom at 4.2°K (μ_0) deduced from our data for the ferromagnetic alloys are listed in Table I. Their values, consistent with previous results for the Ni-rich alloys,[7] decrease almost linearly to zero at ~44-at.% Ni. In fact, our data for the 44-at.% Ni alloy show that $T_c \approx 0$°K and thus define the critical composition very closely. The 42- and 40-at.% Ni alloys are progressively weaker paramagnets.

The equipment and techniques used in our neu-

Table I. Magnetic properties of Ni-Cu alloys: Curie point T_c and spontaneous moment μ_0 at 4.2°K from magnetization measurements; zero-angle magnetic cross section $(d\sigma/d\Omega)_0$ and range parameter κ_0 from neutron scattering experiments at 4.2°K; concentration C and total moment $\mathfrak{M}$ of magnetization clouds deduced from μ_0 and $(d\sigma/d\Omega)_0$.

Ni-Cu alloy (at.% Ni)	T_c (°K)	μ_0 (μ_B/atom)	$(d\sigma/d\Omega)_0$ (b/sr atom)	κ_0 (A^{-1})	C (%)	$\mathfrak{M}$ (μ_B)
50	40	0.059	0.0240	0.41	0.70	8.4
48	23	0.041	0.0175	0.37	0.47	8.8
46	9	0.022	0.0115	0.34	0.20	10.8

tron experiments have been described previously.[5] Basically, we have measured the elastic diffuse scattering of neutrons from the polycrystalline Ni-Cu samples at 4.2°K. The long neutron wavelength (~4.8 Å) allowed us to operate at very small scattering vectors and avoid any multiple Bragg reflections. At each counter angle, data were taken alternately with the sample either demagnetized in zero field or subjected to a field of ~4 kOe parallel to the scattering vector. The difference between the measured cross sections for these two conditions is plotted against the scattering vector κ in Fig. 1(a). For our three alloys that are ferromagnetic at 4.2°K, this difference corresponds to the magnetic scattering cross section for the demagnetized state.

Consider hypothetically that in these ferromagnetic alloys all the Ni atoms have the same magnetic moment and all the Cu atoms have zero moment. The magnetic scattering cross section may then be expressed as

$$d\sigma/d\Omega = \tfrac{2}{3}\times 0.0729C(1-C)[M(\kappa)]^2, \qquad (1)$$

where the factor $\frac{2}{3}$ arises from the assumed random orientation of the domain magnetizations in the demagnetized state, C is the Ni or Cu concentration, and $M(\kappa)$ is the Fourier transform of the moment density $\rho(r)$ at distance r from the center of a Ni atom, i.e.,

$$M(\kappa) = \int_v \rho(\vec{r})e^{i\vec{\kappa}\cdot\vec{r}}dv_{\vec{r}}. \qquad (2)$$

Thus, $M(\kappa)$ is the magnetic form factor (times the magnetic moment in μ_B) for a Ni atom, which is essentially constant over our entire experimental range of κ. It follows that $d\sigma/d\Omega$ would also be constant. Moreover, if the moment per Ni atom in the 50-at.% Ni alloy is taken to be $0.118\mu_B$ (i.e., $2\mu_0$), Eq. (1) gives $d\sigma/d\Omega \approx 0.2 \times 10^{-3}$ b/sr atom. The scattering cross sections calculated for the 48- and 46-at.% Ni alloys would be even smaller.

In contrast to the small and constant scattering cross sections predicted for this quasiuniform distribution of magnetic moment, our experimental scattering curves in Fig. 1(a) for the three ferromagnetic alloys show a very rapid increase to large cross-section values at small κ. These curves are in fact very similar in shape to those previously obtained for dilute Fe (or Co) in Pd.[5] We will therefore assume that they can be interpreted analogously in terms of magnetic polarization clouds extending over many atoms. Specifically, we will compare our ferromagnetic

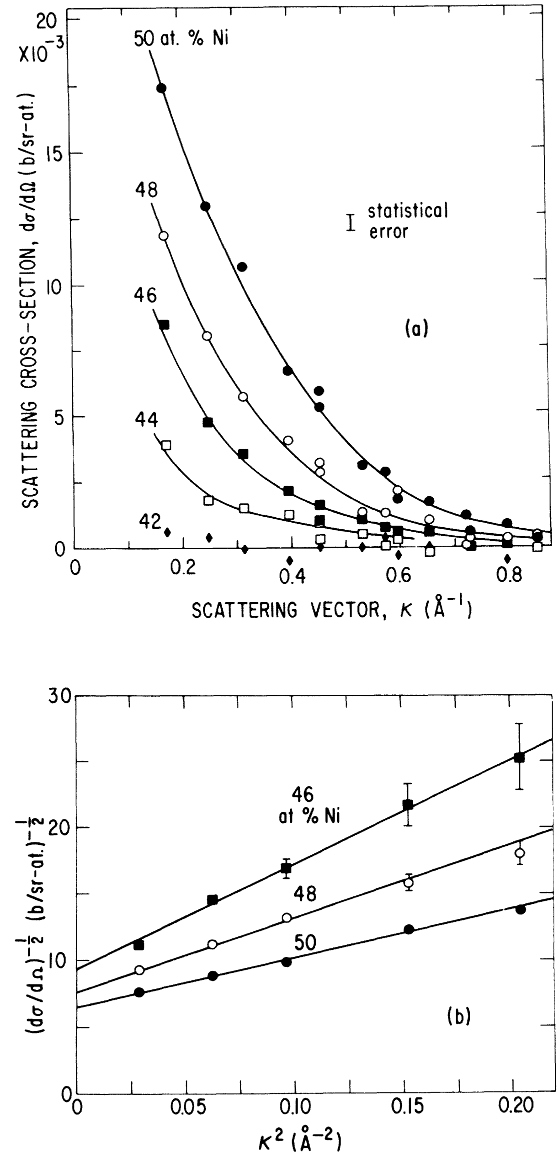

FIG. 1. (a) Magnetic neutron scattering data for various Ni-Cu alloys at 4.2°K. (b) Same data for the ferromagnetic alloys plotted according to assumed Lorentzian variation.

Ni-Cu results with Eqs. (1) and (2), where C will now represent the concentration of the magnetization clouds and $\rho(r)$ the average moment density at distance r from a cloud center. At $\kappa = 0$, Eq. (2) gives

$$M(0) = \int_v \rho(\vec{r})dv_{\vec{r}} \equiv \mathfrak{M}, \qquad (3)$$

the average total integrated moment per cloud (in μ_B), and Eq. (1) becomes

$$(d\sigma/d\Omega)_0 = 0.0486C(1-C)\mathfrak{M}^2. \qquad (4)$$

To determine $(d\sigma/d\Omega)_0$ from our scattering data, we apply the assumption[5] that $M(\kappa)$ is Lorentzian in form [which implies that $\rho(r)$ is a Yukawa function], i.e.,

$$M(\kappa) \propto (\kappa^2 + \kappa_0^2)^{-1}, \quad \rho(r) \propto r^{-1} e^{-\kappa_0 r}, \qquad (5)$$

where κ_0 is a range parameter. Hence, the data at low κ (whose relative error is small) are plotted as $(d\sigma/d\Omega)^{-1/2}$ vs κ^2 in Fig. 1(b), where they clearly give excellent straight lines. From the extrapolations of these lines to $\kappa = 0$ and from their slopes we obtain the values for $(d\sigma/d\Omega)_0$ and κ_0 listed in Table I. Furthermore, the moments of all the magnetization clouds will be considered to contribute additively to the spontaneous moments of the ferromagnetic alloys, so that

$$\mu_0 = C\mathfrak{M}. \qquad (6)$$

Substituting our experimental results for $(d\sigma/d\Omega)_0$ and μ_0 into Eqs. (4) and (6), we deduce the values for C and $\mathfrak{M}$ listed in Table I. Clearly, $\mathfrak{M}$ is large and fairly constant (comparable with the moments of the magnetization clouds around Fe or Co impurity atoms in Pd[5]), whereas the cloud concentration C is very dilute and varies almost linearly with the deviation of the alloy composition from the critical value of 44-at.% Ni (like the variation of the spontaneous moment itself).

The values for C do not correlate with the much lower impurity concentration in our samples nor with the short-range chemical clustering which, according to our neutron data, is relatively independent of alloy composition. Hence, the magnetic polarization clouds appear to be an intrinsic phenomenon in these weakly ferromagnetic Ni-Cu alloys.

Since our scattering data reveal no evidence for any spatial ordering, the magnetization clouds are presumably located randomly throughout the alloys. However, from the concentration C we can estimate a mean distance between the centers of adjacent clouds. For the 50-at.% Ni alloy, this separation distance is ~11.8 Å, which is used together with the transform of our Lorentzian-like scattering curve in obtaining the schematic picture of the moment density distribution shown in Fig. 2. The overlap between adjacent clouds is evidently fairly small; it is even smaller in the alloys nearer the critical composition. Although the moment of the central atom is ambiguous since it depends strongly on the relatively inaccurate data at large κ, the bars representing the moments of successive neighbors

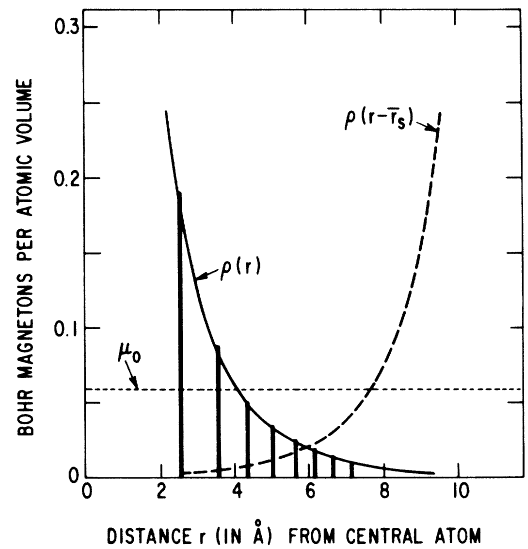

FIG. 2. Schematic distribution of moment density in $Ni_{50}Cu_{50}$ alloy at 4.2°K (spontaneous moment μ_0 also shown uniformly distributed). Two adjacent magnetization clouds are shown at mean separation distance $\bar{r}_S$ of 11.8 Å, the lattice parameter of this fcc alloy being ~3.56 Å. Bars represent average atomic moments of successive neighbors of central atom.

are quite reliable. The moments shown are per average atom, and if the moment of every Cu atom is taken to be zero, the moment per Ni atom will have twice these values and thus be remarkably close to $0.6\mu_B$ (the value in pure Ni) near the center of a magnetization cloud.

Regarding the paramagnetic alloys, Fig. 1(a) shows that essentially no magnetic diffuse scattering difference was observed for the 42-at.% Ni alloy; this was also the case for the 40-at.% Ni alloy. It is clear, however, that the 44-at.% Ni alloy has a significant scattering cross section, whose variation with κ resembles that for the ferromagnetic alloys. Since at 4.2°K this alloy is very close to criticality, the scattering from the slow critical fluctuations will be nearly elastic, which makes it possible for it to be detected in our type of diffraction experiment. In such a case, the nuetrons give an instantaneous picture of the slowly time-varying spatial distribution of moment, which is evidently not very different from the static cloudlike distribution in the ferromagnetic alloys.

We now have similar evidence for giant magnetization clouds in a ferromagnetic Ni-Rh alloy near the critical composition, which suggests that this peculiar and unexpected phenomenon may well pertain to all highly exchange-enhanced alloy systems. A more comprehensive study is currently in progress.

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MAGNETOSTRICTION DUE TO SURFACE CURRENTS IN TYPE-II SUPERCONDUCTORS

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The mixed state behavior of type-II superconductors is determined by both the flux-line density and the superconductive surface currents. These currents give rise to a new magnetostrictive term that depends on the surface currents and the flux-line density. The new term is found to be very large in the indium alloys investigated. The experimental results are explained theoretically and it is shown that the elastic constants can be calculated from measurements of the new magnetostrictive term.

Large diamagnetic or paramagnetic surface currents can be induced in superconductors of the second kind when they are in the mixed state. We have found that in indium alloys these currents give rise to dimensional changes that are comparable with the total change between the normal and superconducting states and much larger than those resulting from magnetostriction in the Meissner phase. This new and unexpectedly large effect can be explained, if the sample is regarded as composed of two phases. One, the volume phase, defines the magnetic field and the field direction just inside the surface by the value of its magnetization. The second phase is the current-carrying surface layer that is exposed to field pressures from both sides. The magnetostriction arising from the surface currents is an elastic deformation under these pressures and it yields a novel method for the determination of the elastic constants. In this note we summarize our experimental results and their theoretical explanation.

When an ellipsoidal specimen is taken into the mixed state by increasing the field H from zero or reducing it from $H > H_{c2}$ and the change in field is then reversed, then screening currents in the surface are induced reversibly. This is shown in the magnetization curves of Figs. 1(c) and 1(d). The short lines indicated by pairs of arrows in the figure are such reversible changes in magnetization. Their slope shows that the whole change in external field is screened from the interior of the sample by the induced currents. That the screening is really complete has

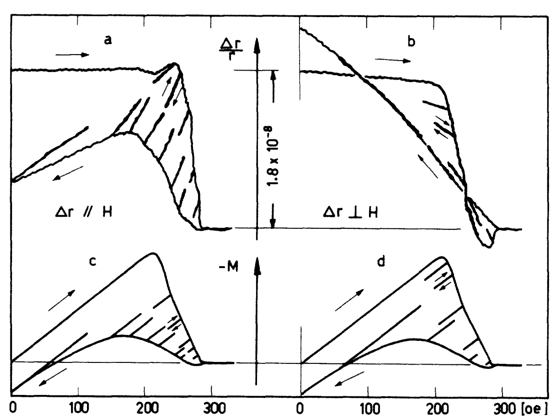

FIG. 1. Magnetostriction and magnetization curves for a flat ellipsoid made from polycrystalline In–14-at.% Tl. The length change is measured at a radius of the equator. The magnetic field was applied in the equatorial plane in directions parallel (a) and (c) and perpendicular (b) and (d) to the measured radius.