Magnetic properties of Ni-Rh alloys near the critical composition for ferromagnetism*

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The magnetizations of Ni-Rh alloys on either side of the critical composition for ferromagnetism ($c_{crit} \approx$ 63-at.% Ni) were measured between 4.2 and 250 °K in fields up to 56 kOe. The initial paramagnetic susceptibility of each alloy is shown to be resolvable into a Curie-Weiss component and a weakly temperature-dependent component of the exchange-enhanced Pauli type. The latter component, which is essentially equal to the high-field differential susceptibility at low temperatures, is maximum near c_{crit}. The onset of ferromagnetism, however, is more directly related to divergence of the Curie-Weiss susceptibility component and is attributed to interactions between superparamagnetic clusters. The average moment per cluster is deduced to be $\sim(20\text{–}24)\mu_B$ over the composition range investigated (65–51-at.% Ni). The concentration of the magnetic clusters, though remaining dilute, increases rapidly as c_{crit} is approached from the paramagnetic side; moreover, it correlates closely with the statistical occurrence of extremely Ni-rich local regions in these atomically disordered alloys. Comparisons are made with the Ni-Cu and Ni-V systems near c_{crit}, for which recent magnetization studies have revealed the analogous existence of dilute concentrations of giant magnetic clusters.

INTRODUCTION

The alloys of nickel pose a special challenge to our understanding of magnetism in metallic systems. Historically, their spontaneous magnetizations and related properties were once thought to have found a simple explanation in the collective-electron rigid-band model, particularly in the case of nickel alloys with non-transition-group metals (Cu, Zn, etc.).[1] This theoretical rationale was applied fairly successfully to ferromagnetic alloys all the way out to the critical composition (c_{crit}, where the Curie point $T_C \to 0$ °K) and even beyond into the paramagnetic composition range, where the exchange forces are expected to cause an enhancement of the Pauli susceptibility. However, it has been known that certain detailed properties of the prototypal Ni-Cu system near c_{crit} (e.g., pronounced susceptibility increases at low temperatures,[2,3] an essentially temperature-independent component of the specific heat[4,5]) are distinctly anomalous from a rigid-band model viewpoint and are more readily attributable to superparamagnetic clusters acting as giant local moments.[6] Generally, these anomalies have been dismissed as extraneous manifestations of gross chemical inhomogeneities in the alloy samples.[7] The fact that Ni, as a dilute impurity in Cu or any other nonmagnetic metal host, shows no signs of having a stable local moment (unlike Fe or Mn)[8] seemed to argue against the possibility of local moments as intrinsic phenomena in concentrated Ni alloys.

Many recent developments have changed this picture profoundly, especially with regard to Ni-Cu. Most recently, neutron scattering experiments on ferromagnetic Ni-Cu alloys by Aldred *et al.*[9] (and earlier on one alloy by Cable *et al.*[10]) have demonstrated that the 3*d*-electron moment of a Ni atom depends sensitively on its local environment, decreasing in magnitude with increasing number of neighboring Cu atoms. Indeed, these results[9] indicate a gradual evolution with increasing Cu concentration toward the highly inhomogeneous spatial distributions of magnetization in the weakly ferromagnetic alloys near c_{crit}. These distributions, as deduced previously by Hicks *et al.* from similar experiments,[11] consist of discrete regions of magnetization, called "magnetic polarization clouds," each extending over many atoms and having an average total moment of about $10\mu_B$. Later susceptibility measurements on Ni-Cu revealed that the giant polarization clouds persist, as superparamagnetic entities, well into the paramagnetic composition range.[12] Moreover, the very dilute concentrations of these superparamagnetic clouds indicated that they nucleate at local regions that are extremely Ni rich. These local Ni-rich regions, the cores of the polarization clouds, may occur statistically even in perfectly random solid solutions, as has been discussed by Perrier *et al.*[13]; the short-range atomic clustering that normally exists in Ni-Cu alloys[9,14] simply increases the concentration of these regions. This statistical picture, with the magnitude of each Ni atomic moment having a simple prescribed dependence on its local chemical environment, gives a good quantitative fit to the saturation magnetizations of Ni-Cu alloys near c_{crit}.[13] An analogous but more complicated prescription adopted by Robbins *et al.*[15] gives excellent agreement over the entire Ni-Cu composition range. This viewpoint of a spatially inhomogeneous magnetic state has been incorporated, often with some allowance for the magnetic aspects of the local environments, into several theoretical analyses of the magnetization and related properties of Ni-Cu.[16–23]

It has now begun to appear that the magnetism of other Ni-based alloy systems, not only of Ni-Cu, can be characterized near c_{crit} in terms of polarization clouds (often referred to as "magnetic clusters"). Among the evidence accumulating to this effect, probably the most convincing have again been the results of elastic neutron scattering experiments on weakly ferromagnetic alloys. These results have been reported for Ni-Pd,[24] Ni-Cr,[25] and Ni-Rh,[26] and in each case a marked upswing of the diffuse magnetic cross section at very small scattering vectors has signified the existence of moment-density distributions extending over many atomic volumes, each distribution constituting a magnetic cluster. The case of Ni-Rh is particularly interesting since Bucher *et al.*[27] have claimed, on the basis of a specific-heat and susceptibility study of this system, that the anomalies observed near c_{crit} (~63-at.% Ni) arise from a critical exchange enhancement accompanied by large spin fluctuations. Their low-temperature specific-heat results were later reanalyzed by Hahn and Wohlfarth[28] and shown to be equally consistent with the existence of superparamagnetic clusters, for which there also was some qualitative susceptibility evidence in alloys near c_{crit}.[29] More recently, Triplett and Phillips[30] found that the anomalous low-temperature component of the heat capacity of the alloy $Ni_{62}Rh_{38}$ is largely suppressed (probably shifted to higher temperatures) by the application of a 38-kOe field. They concluded that this observed effect is in good quantitative agreement with the predicted behavior of superparamagnetic clusters and, furthermore, that it would have required much larger fields if the anomalous heat capacity contribution were associated with spin fluctuations.

The existence of magnetic clusters in Ni-Rh clearly calls for more complete documentation in order to establish whether or not it is a significant intrinsic phenomenon. In view of this experimental need and the continued interest in Ni-Rh, we have carried out a systematic magnetization study of this alloy system near the critical composition for ferromagnetism (c_{crit}). Our preliminary results for a few paramagnetic Ni-Rh alloys, recently reported in brief,[31] appear to indicate that magnetic clusters do indeed play an intrinsic role, qualitatively akin to their role in Ni-Cu. In the present paper, our complete results for a much broader range of Ni-Rh compositions, extending through c_{crit}, are described and discussed in full detail. Pertinent aspects of the magnetic critical-point behavior of the weakly ferromagnetic alloy $Ni_{65}Rh_{35}$, which we have reported separately,[32] are also included in the present discussion.

EXPERIMENTAL TECHNIQUES

Nickel-rhodium alloy buttons were prepared by arc-melting weighed mixtures of the two metals (both specified as 99.999% pure) in a helium atmosphere and were cold worked by hammering. The samples machined from each button were cylinders (2.5 mm diam, 5 mm long) whose edges were beveled down in order to approximate ellipsoids (for reasonably uniform demagnetization). After their surfaces were acid etched, the samples were sealed in helium-filled quartz tubes, annealed for 3 days at 1200 °C, then quenched into water. The nominal compositions of the alloy samples are indicated in Table I. As later inferred from our susceptibility data, the ferromagnetic (presumably Fe) impurity level in the samples was probably well under 50 ppm.

Magnetization measurements on these samples were carried out with a temperature-controlled vibrating-sample magnetometer system (Princeton Applied Research Corp., Model No. 150-A). The Nb-Ti superconducting solenoid (Westinghouse Corp., Model No. H-5794) in this system was typically operated at currents up to 50 A corresponding to a maximum field at the sample of 56 kOe. The current-field specifications of the magnet were tested by means of a calibrated field-sensitive Bi-film resistor (American Aerospace Controls Corp., Model No. MRA-12); the field homogeneity over the sample length was found to be within 1%. The sample-zone temperature was monitored by a GaAs-diode thermometer (Lake Shore Cryotronics, Inc.), which we calibrated in zero field against a precalibrated Ge resistor (Cryocal, Inc., Model No. CR-1000) and a precalibrated Pt resistor (Minco Products, Inc., Model No. 51061-2) over the ranges (4.2–40)°K and (40–260)°K, respectively. Each temperature calibration point corresponded to a recorded setting of the temperature-control unit that governed the current to an electric heat exchanger (temperature monitored by another GaAs diode), through which vapor from the liquid-He reservoir flowed into the sample chamber at a controlled rate. Since the GaAs diodes were not entirely field insensitive, calibration corrections were determined as a function of field at various fixed temperatures set by means of a He-gas thermometer bulb inserted in the sample chamber. The maximum uncertainty in the sample temperature over the ranges of operation [(4.2–250)°K, 0–56 kOe] was about 0.1 °K; the uncertainty was much less at 4.2 °K, when the sample chamber was generally flooded with liquid He. The magnetic-moment calibration of the magnetometer output voltage was tested at different sensitivity levels against the saturation moments of various sized samples of pure Ni and Fe at 4.2 °K.[33] The mag-

TABLE I. Magnetic properties of Ni-Rh alloys.

Alloy (at.% Ni)	$\sigma_0(0)$ (emu/g)	$\sigma_s(0)$ (emu/g)	T_C (°K)	Θ (°K)	$\chi'(0)$[a]	C_{CW}[a]	$\bar{\mu}^*$ (μ_B)	$\bar{c}^*$ (%)	P_{12} (%)
65.0	7.40	11.70	44	62	0.77	5.15	19.7	0.790	0.370
64.4	5.00	10.20	27	43	0.91	4.59	20.1	0.676	0.328
64.0	3.95	9.10	19	34	1.05	4.16	20.4	0.595	0.302
63.2	1.70	7.40	5	11	1.43	3.76	22.7	0.438	0.257
62.6	~0	5.80	~0	4	2.00	2.78	21.4	0.365	0.227
62.0	...	4.95	...	−2	2.22	2.29	20.7	0.324	0.200
60.0	...	3.40	...	−21	2.00	1.61	21.1	0.220	0.131
58.0	...	1.55	...	−43	1.76	0.826	23.8	0.090	0.084
56.0	...	0.96	...	−47	1.63	0.501	23.3	0.058	0.053
53.0	...	0.52	...	−43	1.33	0.263	22.6	0.033	0.026
51.0	...	0.33	...	−50	1.04	0.160	21.7	0.022	0.016
				estimated uncertainties (±)					
0.1	2%	5%	0.5	2	5%	3%	8%	13%	...

[a] $\chi'(0)$ and C_{CW} in units of 10^{-5} emu/Oe g and 10^{-3} emu °K/Oe g, respectively.

netization values for these samples checked within 1%, which represents the absolute error in all our magnetization measurements. The relative error in the magnetizations measured at different temperatures and fields for a given mounting of a sample was about an order of magnitude smaller.

EXPERIMENTAL RESULTS AND DISCUSSION

The magnetization (σ) of each Ni-Rh alloy sample was measured as a function of magnetic field (H, as corrected for demagnetization) at temperatures (T) from 4.2 °K up to about 250 °K. Our results for σ vs H at 4.2 °K are illustrated in Fig. 1. The curves for the alloys of 63.2-at.% or more Ni show a spontaneous magnetization (σ_0) clearly indicative of ferromagnetism. The Curie points (T_C) of these alloys were determined from isothermal σ^2 vs H/σ plots of the data; their values, together with those of σ_0 extrapolated to 0 °K, are listed in Table I. A similar plot for the 62.6-at.% Ni sample gives $T_C \approx 0$ °K, indicating that this alloy is essentially at the critical composition for ferromagnetism (c_{crit}). Moreover, the σ^2 vs H/σ plots near T_C, which are extremely sensitive to any macroscopic inhomogeneities, revealed no metallurigical complications in our alloy samples.

For each alloy over its paramagnetic range of temperatures, the initial susceptibility (χ_0) was determined from $d\sigma/dH$ extrapolated to $H=0$; its reciprocal values (χ_0^{-1}) are shown plotted versus T in Fig. 2. In their concave-downward shape (particularly evident at low temperatures), these curves agree with the earlier Ni-Rh results of Cottet *et al.*[29] They differ qualitatively from the flat concave-upward curves normally predicted from simple itinerant-electron band theory.[1] In this sense, the χ_0^{-1} vs T curves for Ni-Rh resemble those for paramagnetic Ni-Cu near c_{crit}, where the latter were each found to be analyzable into a Curie-Weiss component and a weak temperature-independent component, attributed respectively to superparamagnetic clusters and itinerant-electron band polarization.[12]

Following an analogous procedure for Ni-Rh alloys near c_{crit}, we assume that the measured magnetization may be expressed as

$$\sigma(H,T)=\sigma_{cl}(H,T)+H\chi'(T) \quad , \tag{1}$$

where σ_{cl} represents the magnetization arising from the spin alignment of clusters and χ' is a band-polarization susceptibility, which is expected to be

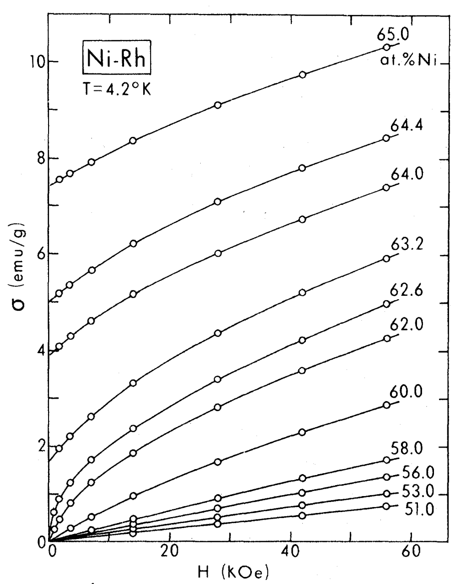

FIG. 1. Magnetization vs field at 4.2 °K for Ni-Rh alloys (compositions in at.% Ni).

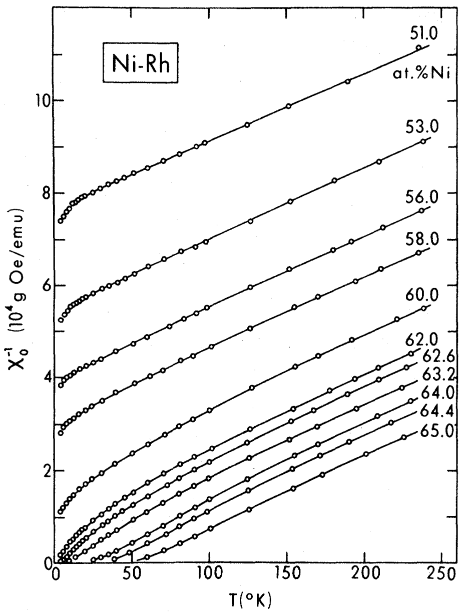

FIG. 2. Inverse initial susceptibility vs temperature for Ni-Rh alloys (compositions in at.% Ni).

only mildly temperature dependent and negligibly dependent on laboratory fields. Consequently, the spontaneous magnetization of a ferromagnetic alloy below T_C will derive entirely from the zero-field alignment of cluster moments, i.e., $\sigma_0(T) = \sigma_{cl}(0, T)$, whereas the initial susceptibility above T_C (and at all temperatures for a paramagnetic alloy) will include a band polarization term as well as a superparamagnetic cluster term, the latter of which is taken to be Curie-Weiss-like, i.e.,

$$\chi_0(T) = \chi_{cl}(T) + \chi'(T) = C_{CW}/(T-\Theta) + \chi'(T) \ . \qquad (2)$$

In the high-field limit, achieved by extrapolation from laboratory fields, it follows that

$$\sigma_s(T) \equiv \sigma_{cl}(\infty, T) = \lim_{H\to\infty}[\sigma(H,T) - H\chi'(T)] \qquad (3)$$

and

$$\chi'(T) = \lim_{H\to\infty}(d\sigma/dH)_T \ , \qquad (4)$$

where σ_s represents the saturation limit of the cluster spin alignment.

In order to separate our $\chi_0(T)$ data for Ni-Rh into the components indicated in Eq. (2), we took as an approximate low-temperature value for χ' the differential susceptibility at 4.2 °K extrapolated to infinite field, as suggested by Eq. (4). The value thus estimated was ~ 2×10^{-5} emu/Oe g for all the paramagnetic alloys. This starting value for χ' was then adjusted for each of these alloys until $(\chi_0 - \chi')^{-1}$ vs T became linear over a range of low temperatures (i.e., up to ~ 20 °K). In order that this linearity continue up to moderate temperatures (~ 100 °K), it was found necessary in each case to let χ' have a weak temperature dependence of the form

$$\chi'(T) = [\chi'^{-1}(0) + \alpha T^2]^{-1} \qquad (5)$$

with α positive. At still higher temperatures, where the latter procedure began to overestimate the variation in χ', we simply extended the linear dependence of $(\chi_0 - \chi')^{-1}$ on T and computed backwards to obtain the χ' values corresponding to the measured values of χ_0. Our results for the temperature dependence of $\chi_{cl}^{-1} \equiv (\chi_0 - \chi')^{-1}$ and of χ'^{-1} are plotted in Figs. 3(a) and (b), respectively; the values of $\chi'(0)$ are listed in Table I. For the ferromagnetic alloys, since the above analysis cannot be used below T_C, we adjusted $\chi'(0)$ to the values listed in Table I and assumed that $\chi'^{-1}(T)$ is displaced upward uniformly from the curve determined for the critical-composition (62.6-at.% Ni) alloy, as illustrated in Fig. 3(b). The linear plots thus obtained for $\chi_{cl}^{-1}(T)$ above T_C, which are presented in Fig. 3(a), are rather insensitive to the exact shape assumed for $\chi'^{-1}(T)$ owing to the fact that χ' is only a small part of χ_0 over this temperature range.

For each Ni-Rh alloy, the values of C_{CW} and Θ obtained from the Curie-Weiss behavior of $\chi_{cl}(T)$, as expressed in Eq. (2), are listed in Table I. Allowing that χ_{cl} is associated with superparamagnetic clusters, we can write for the Curie-Weiss constant

$$C_{CW} = Nc^*\langle\mu^{*2}\rangle/3k \ , \qquad (6)$$

where N is the number of atoms per gram, c^* the cluster concentration (number per atom), $\langle\mu^{*2}\rangle$ the mean squared value of the magnetic moment per cluster, and k the Boltzmann constant. We further allow that the saturation magnetization $\sigma_s(0)$ obtained by high-field extrapolation of $\sigma(H,0) - H\chi'(0)$, as indicated in Eq. (3), represents tha parallel spin alignment of all the magnetic clusters in the alloy and can therefore be expressed as

$$\sigma_s(0) = Nc^*\langle\mu^*\rangle . \qquad (7)$$

Typical high-field extrapolations for the Ni-Rh alloys, involving our low-temperature magnetization data (extended trivially to 0 °K) and the values deduced for $\chi'(0)$, are illustrated in Fig. 4. The values thus determined for $\sigma_s(0)$ are listed in Table I. From a combination of Eqs. (6) and (7), we obtain the "average" quantities defined as

$$\overline{\mu}^* \equiv \langle\mu^{*2}\rangle/\langle\mu^*\rangle = 3kC_{CW}/\sigma_s(0) \ , \qquad (8a)$$

$$\overline{c}^* \equiv c^*\langle\mu^*\rangle/\overline{\mu}^* = \sigma_s^2(0)/3NkC_{CW} \ . \qquad (8b)$$

(Note that a distribution in cluster moment size will

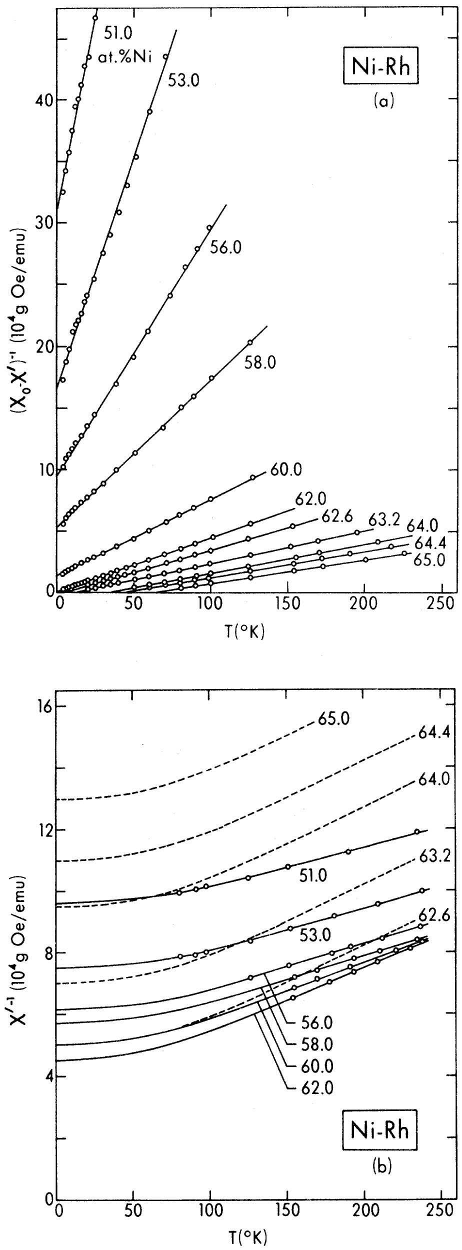

FIG. 3. (a) Inverse of $\chi_{el} \equiv \chi_0 - \chi'$ and (b) inverse of χ', as functions of temperature, for Ni-Rh alloys (compositions in at.% Ni).

affect the meaning of $\overline{\mu}^*$ and $\overline{c}^*$, rendering them respectively higher and lower than the true average values, $\langle\mu^*\rangle$ and c^*.) Substituting our experimentally derived values for C_{CW} and $\sigma_s(0)$ into these expressions, we calculated the $\overline{\mu}^*$ and $\overline{c}^*$ values listed in Table I. From these results we see that the cluster moment $\overline{\mu}^*$ has enormous values [~ (20–24)μ_B] that are remarkably constant over the composition range of study. Contrastingly, the cluster concentrations $\overline{c}^*$ are very small and decrease rapidly with decreasing at.% Ni, passing smoothly through the critical composition. Qualitatively, these results for the average moment and concentration of magnetic clusters in Ni-Rh are very similar to those previously obtained for[12] Ni-Cu and[34] Ni-V by the same method of data analysis, which yielded giant $\overline{\mu}^*$ values of ~ $10\mu_B$ and ~ $40\mu_B$, respectively, near c_{crit}.

Evidence for giant magnetic clusters in the weakly ferromagnetic $Ni_{65}Rh_{35}$ alloy was also extracted from detailed $\sigma(H, T)$ data in the vicinity of T_C, as we have already reported separately.[32] From these data, the spontaneous magnetization just below T_C and the initial susceptibility just above T_C were determined and their temperature dependences described respectively as $\sigma_0(T) = m_0|1 - T/T_C|^{\beta}$ and $\chi_0(T) = (m_0/h_0)|1 - T/T_C|^{-\gamma}$, where β and γ are critical exponents, and m_0 and m_0/h_0 may be regarded as critical coefficients. Our results for the latter were compared with the theoretical predictions that $m_0/\sigma_0(0) = n_1$ and $\mu_0 h_0/kT_C = n_2$, in which the elementary moment μ_0 is normally taken to be the average moment per atom and the numbers n_1 and n_2 are both slightly larger than unity for various classic models (mean field, Ising). While our experimental value for n_1 obeyed these predictions quite well, the value for n_2 was only ~ 0.007 when μ_0 was equated to $\sigma_0(0)/N \approx 0.1\mu_B$. Indeed, agreement with the near-unity values predicted for n_2 required that we set $\mu_0 \approx 22\mu_B$, which is essentially the $\overline{\mu}^*$ value for this alloy given in Table I. Thus, it was concluded that the elementary dynamical moments involved in the Curie-point transition are those of giant magnetic clusters rather than of in-

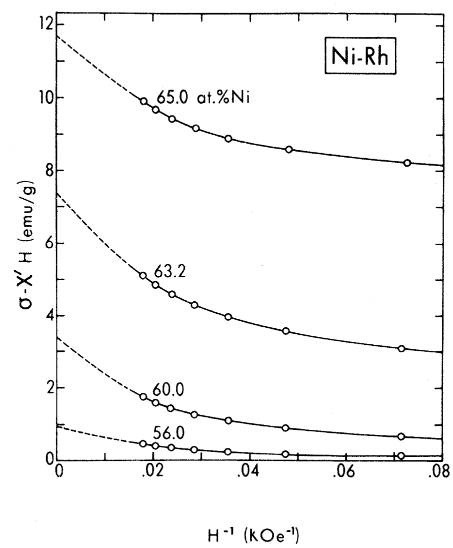

FIG. 4. Variation of $\sigma - \chi'H$ (for $T \to 0$°K) with H^{-1} for Ni-Rh alloys (compositions in at.% Ni).

dividual atoms. We further showed that the same conclusion can be reached analogously from the Curie-point properties of dilute *Pd*Fe, the classic giant-moment alloys—whose critical exponent values, incidentally, also are anomalous and similar to those of the Ni-Rh alloy.

As was done in the case of Ni-Cu,[12] let us examine the possibility that the magnetic cluster concentrations in Ni-Rh correlate with the statistical occurrences of a particular local chemical composition. For simplicity, consider the probability P_n that any lattice site of a fcc Ni_cRh_{1-c} alloy (assumed to be a random solid solution) is occupied by a Ni atom whose 12 nearest neighbors include n or more Ni atoms. In the extreme case of $n = 12$, we have $P_{12} = c^{13}$, whose values for the alloys studied are listed in Table I. The variation of P_{12} with alloy composition is clearly quite similar to that of $\bar{c}^*$, although at the Ni-rich end of this range the values of $\bar{c}^*$ appear to be rising towards $P_{11} = c^{13} + 12c^{12}(1-c)$, whose values exceed those of P_{12} by nearly an order of magnitude. Despite the oversimplicity of these considerations, they are probably correct in indicating that the giant magnetic clusters in Ni-Rh are nucleated in local regions that are extremely Ni rich. In the case of Ni-Cu, $\bar{c}^*$ was originally found[12] to lie between P_{10} and P_{11}, but it was recently shown[20] in a more rigorous statistical treatment of the atomic clustering that $\bar{c}^*$ follows closely the values of P_{12}. Thus, the local conditions for magnetic cluster nucleation in Ni-Rh and Ni-Cu appear to be fairly alike. In either case, according to the neutron diffraction work on Ni-Rh,[26] and Ni-Cu,[11] the average moment density inside a magnetic cluster decreases gradually over a radial distance of many lattice spacings and thus extends considerably outside the central core of nearest-neighbor Ni atoms.

Whatever may be the underlying mechanisms for magnetic cluster formation in Ni-Rh (which will be commented upon later), it is clear from our results that giant magnetic clusters persist, as superparamagnetic entities in extremely dilute concentrations, well into the paramagnetic composition range. The negative values of the Curie-Weiss temperature (Θ) for the weakly paramagnetic Ni-Rh alloys, similar to (though larger in magnitude than) the negative Θ values for paramagnetic Ni-Cu,[12] and Ni-V,[34] may reflect local anisotropy effects[35] as much as a predominance of antiferromagnetic over ferromagnetic exchange interactions, presumably of the indirect Ruderman-Kittel-Kasuya-Yosida type.[18] As c_{crit} is approached from the paramagnetic side and the cluster concentration in Ni-Rh increases, Θ rises sharply to large positive values, probably as the outcome of increased ferromagnetic (direct overlap) interactions between adjacent clusters. Beyond c_{crit}, in the weakly ferromagnetic Ni-Rh alloys, the spontaneous magnetization $\sigma_0(0)$ is much smaller than the corresponding saturation value $\sigma_s(0)$, although their ratio does approach unity with increasing at.% Ni, as seen in Table I. This behavior shows that in zero external field many of the magnetic clusters in these alloys are not ferromagnetically aligned even at very low temperatures—which is consistent with the fact that specific-heat anomalies of the type ascribable to superparamagnetic clusters[28] are observed in the weakly ferromagnetic as well as the strongly paramagnetic Ni-Rh alloys.[27]

According to our interpretation, the onset of ferromagnetism in Ni-Rh arises from the exchange coupling between magnetic clusters and is manifested at T_C in the divergence of χ_{cl}, the Curie-Weiss-like cluster component of the initial susceptibility, as indicated in Fig. 3(a). The other component of the initial susceptibility (i.e., χ') exhibits only a mild T^2 deviation from its zero-temperature value and, as further seen from Fig. 3(b), the variation of χ'^{-1} at higher temperatures seems to approach a weak linear dependence. A very similar behavior was recently deduced for χ' vs T in Ni-V alloys close to the critical composition,[34] for which $\chi'(0)$ is comparable in size to our Ni-Rh results. Values obtained for χ' in Ni-Cu alloys near c_{crit} are about an order of magnitude smaller and were assumed to have negligible temperature dependence[12]; they were attributed, as was mentioned earlier, to weak band polarization effects. In the case of Ni-Rh, the χ'^{-1} vs T curves in Fig. 3(b) have exactly the shape predicted for such effects when the exchange enhancement is large and accompanied by spin fluctuations.[36] An analogous explanation was given for the similar χ'^{-1} vs T curves for Ni-V.[37]

Our $\chi'(0)$ results for Ni-Rh are plotted versus alloy composition in Fig. 5, together with $d\sigma/dH$ values taken at various fields from the σ vs H curves for 4.2 °K in Fig. 1. The peaks in the $d\sigma/dH$ curves at the critical composition (~63-at.% Ni), which are most prominent at the lowest fields, are clearly the vestiges of the divergence of $d\sigma/dH$ at zero field. Hence, they relate more closely to the magnetic cluster component of the initial susceptibility (i.e., χ_{cl}) than to the band polarization component (χ'), and this appears to be true even at the highest fields where, according to Eq. (4), $d\sigma/dH$ should be extrapolating to χ'. It is evident from Fig. 5 that the high-field approach of $d\sigma/dH$ to χ' is extremely slow in all these alloys, not only in those very close to c_{crit}. This behavior quite possibly derives from a broad distribution in the magnetic moments of the individual clusters, in which case the clusters with relatively small moments continue to contribute to the low-temperature differential susceptibility up to very high fields.

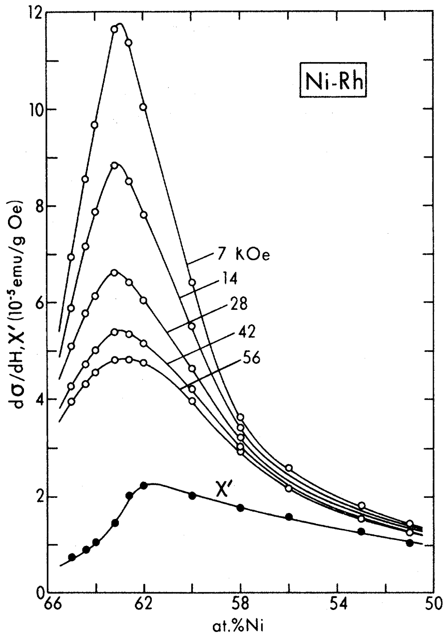

FIG. 5. Open circles: $d\sigma/dH$ at various fields (in kOe) at 4.2 °K; closed circles: $\chi'(0)$, as functions of Ni-Rh alloy composition.

Cluster moment distributions in[38] Ni-Cu and[34] Ni-V have been proposed on the basis of similar experimental evidence.

The variation of our results for $\chi'(0)$ is shown in Fig. 6 with respect to the entire Ni-Rh composition range. On this scale, the values for $\chi'(0)$ define a sharp but finite peak centered near the critical composition. Their rapid decrease on the Rh-rich side of the peak extends smoothly through the points representing some previous high-field susceptibility data,[27] which at these compositions can be safely assumed to contain virtually no magnetic cluster contribution. The descent of $\chi'(0)$ on the Ni-rich side of the peak appears to be even faster, probably reaching very low values for most of the ferromagnetic range, as indicated in the figure. This behavior suggests that the band polarization effects that produce χ', though not responsible for the onset of ferromagnetism, become highly exchange enhanced in the vicinity of c_{crit}. It seems reasonable to associate this strong exchange enhancement with local (presumably fairly Ni rich) regions that are nearly unstable towards the formation of local (cluster) moments. As implied by our χ' results in Fig. 6, these local regions grow and approach critical enhancement as the alloy composition increases in at.% Ni, but at c_{crit} they suddenly become exchange-polarized by the spontaneously aligned moments of the stable magnetic clusters and, as a result, their contribution to the susceptibility (χ') drops precipitously. Spin fluctuations can be expected to occur in these regions of strong local enhancement and in fact, according to a recent resistivity study,[39] they appear to provide the dominant scattering mechanism in Ni-Rh alloys near c_{crit}. In the case of Ni-V, where χ' attains values comparable to those in Ni-Rh, the observed temperature dependence of the resistivity was also found to be characteristic of spin-fluctuation scattering[34]; as in Ni-Rh, any scattering from the superparamagnetic clusters seems to be completely obscured. However, in the case of Ni-Cu, where χ' is very much weaker, the measured resistivity versus temperature curves indicate that the predominant scattering is from superparamagnetic clusters,[40] as was recently elucidated theoretically.[21]

Having attributed the $\chi'(0)$ peak in Fig. 6 to regions of near-critical exchange enhancement, we would be consistent in regarding each statistical Ni-rich local region that constitutes the core of a magnetic cluster in Ni-Rh as having reached and exceeded critical enhancement. There would consequently exist stable local moments on all the Ni atoms (and perhaps the Rh atoms as well)[41] within each cluster core. From this point of departure in the case of Ni-Cu, Roth has proceeded to show analytically that the Ni atoms outside the core of a magnetic cluster will become magnetically polarized but to a decreasing extent with increasing distance from the core.[17] This picture of a giant magnetic cluster or polarization cloud in Ni-Cu agrees with the spin density distribution deduced from

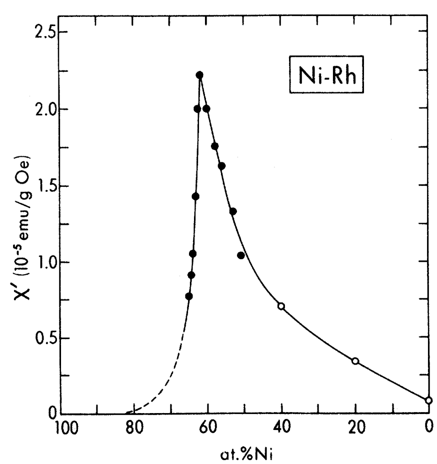

FIG. 6. Closed circles: $\chi'(0)$ vs Ni-Rh alloy composition; open circles: high-field susceptibility data from Ref. 27.

neutron diffraction data and has subsequently been described by Garland and Gonis in a self-consistent theoretical treatment of the entire magnetic cluster (core and all).[20] The cooperative magnetic processes invoked in the latter work are presumably also responsible for the giant magnetic clusters in Ni-Rh, but this remains to be investigated.

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