Instability toward the Formation of Local Moments in Ni-Cu Alloys

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An examination of the nonlocal exchange-enhanced susceptibility of disordered alloys such as Cu-Ni leads to the development of criteria for the formation of localized extended magnetic moments. A simplified model is developed for Cu-Ni, in which the nonlocal susceptibility without exchange enhancement Γ_{ij} is assumed to be finite only if i and j are nickel sites. Using the random-phase approximation for the exchange enhancement, and assuming that Γ_{ij} extends only to nearest neighbors, the instability criterion is examined by a variational method, and conditions for formation of both localized and extended moments are obtained. The results are consistent with the neutron diffraction findings that, near the critical concentration for ferromagnetism, giant moments or polarization clouds occur. These are nucleated by small nickel clusters which are due to statistical fluctuations in the nickel concentration. The number of these centers is enhanced by short-range-order effects. The disappearance of all the effects above the nickel Curie temperature is discussed.

INTRODUCTION

Recent neutron diffraction results obtained by Hicks *et al.*[1] have given strong evidence that the magnetization of Cu-Ni alloys near the critical concentration is carried by giant polarization clouds. It has been suggested by Kouvel and Comly[2] that the clouds are nucleated by statistical fluctuations in the concentration of nickel atoms. Thus, a localized region which is more nickel rich than the average would form a local moment and then polarize the surrounding medium to form the extended structure deduced from the neutron results.

In this article a rather oversimplified model of the exchange-enhanced susceptibility will be developed and a criterion obtained for instability toward spontaneous magnetic polarization. This polarization can be extended throughout the crystal, or it can be localized in a cloud about a nickel cluster. By means of a variational method, we shall examine several examples of each case. Thus for the extended case, we shall look at a uniform polarization and a nonuniform polarization in which the distribution of nickel moments depends only on the immediate environment. For the localized case, we shall examine the polarization clouds around both spherical and flat clusters.

FORMULATION OF MODEL

Lederer and Mills[3] and Doniach and Wohlfarth[4] have made extensive use of a nonlocal exchange-enhanced susceptibility for a uniform system such as Pd. If we consider a d band in which $n_{i\uparrow} - n_{i\downarrow} = m_i$ is the polarization on the ith Wannier site, we write

$$m_i = \sum_j \Gamma_{ij} (g\beta H_j^{\text{eff}}) , \tag{1}$$

where H_j^{eff} is the effective magnetic field on the jth site. Γ_{ij} is the nonenhanced susceptibility, which is given by

$$\Gamma_{ij} = \sum_{\vec{q}} e^{i\vec{q} \cdot \vec{R}_{ij}} \chi(\vec{q}, 0) , \tag{2}$$

where the wave-number-dependent susceptibility is given by

$$\chi(\vec{q}, 0) = \left(\frac{1}{N}\right) \sum_{\vec{k}} \frac{f(\epsilon_{\vec{k}}) - f(\epsilon_{\vec{k}+\vec{q}})}{\epsilon_{\vec{k}+\vec{q}} - \epsilon_{\vec{k}}} , \tag{3}$$

with $f(\epsilon_{\vec{k}})$ the Fermi function for energy $\epsilon_{\vec{k}}$. The effective magnetic field is the sum of the applied field and a term due to the interaction. Using the constant-I model,[3,4] let us assume that the effective field due to the interaction is

$$g\beta H_j^{\text{int}} = I m_j . \tag{4}$$

We are thus working in the random-phase approximation and neglecting correlation except insofar as it modifies I. Equation (1) therefore becomes

$$m_i = \sum_j \Gamma_{ij} (I m_j + g\beta H_j) . \tag{5}$$

Thus far we have been dealing with a uniform system. Let us now assume that an equation like Eq. (5) holds in unpolarized NiCu, except that it holds only on the nickel. We introduce a quantity N_i which is 0 or 1:

$$N_i = 1 \text{ for Ni}$$
$$= 0 \text{ for Cu} . \tag{6}$$

Then our assumption is

$$m_i N_i = N_i \sum_j \Gamma_{ij} N_j (I m_j + g\beta H_j) . \tag{7}$$

Let us note that had we assumed a direct-exchange interaction in addition to the constant I term, then the effective field on the jth site would include the term $\sum_l J_{jl} N_l m_l/g\beta$. This is of the same general

form as that in Eq. (7), except for the factor N_i which would give an additional concentration dependence.

Equation (7) can be obtained with some approximation by the use of the coherent-potential theory of alloys,[5] together with the constant-I Hartree-Fock interaction term, which led to Eq. (4). Alternatively, Kim[6] has found a similar equation using the Anderson model of magnetic impurities. It would seem that Eq. (7) is more general than either of the derivations, and also it is very likely that both s-d interactions and d-d interactions are important. We expect to publish a derivation of Eq. (7) in the future, but for the present we shall adopt the result phenomenologically.

In general, we expect Γ_{ij} to depend on concentration, and probably local environment, and the same is true for I. We shall neglect the environment dependence of Γ, but bear in mind the possible concentration dependence, although we do not have a detailed theory for it.

If we now suppose I to be variable, we can look for the condition for the instability toward formation of a local or extended moment by seeking solutions of the homogeneous form of Eq. (7), i.e., by setting $H_j = 0$. If I_0 is the critical value of I, then we have

$$\sum_j N_i \, \Gamma_{ij} N_j m_j = I_0^{-1} N_i m_i \ . \tag{8}$$

We see that we have an eigenvalue equation for I_0^{-1}. Since we are seeking the smallest value of I_0, we look for the largest eigenvalue of Eq. (8). This can be approached by a variational method. Multiplying by m_i and summing, we have

$$I_0^{-1} = \left(\sum_{ij,nn} N_i m_i \, \Gamma_{ij} N_j m_j \right) \Big/ \sum_i m_i^2 N_i \ . \tag{9}$$

Treating Γ_{ij} as a parameter, let us consider the case in which we neglect all but the $i = j$ and the nearest-neighbor terms (Γ_0 and Γ_1). Then we have

$$I_0^{-1} = \Gamma_0 + \Gamma_1 \left(\sum_{ij,nn} N_i m_i N_j m_j \right) \Big/ \sum_i m_i^2 N_i \ . \tag{10}$$

We can then find the largest eigenvalue of the coefficient of Γ_1,

$$Z_{\text{eff}} = \left(\sum_{ij,nn} N_i m_i N_j m_j \right) \Big/ \sum_i m_i^2 N_i \ . \tag{11}$$

In the case of the pure crystal, Z_{eff} reduces to the number of nearest neighbors, so that we regard it as an effective neighbor number in the alloy case.

INSTABILITY TOWARD EXTENDED POLARIZATION

Uniform Polarization

We consider first a trial function for Eq. (11) consisting of a uniform polarization so that m_i is independent of i. Since it is known that Ni-Cu

alloys exhibit a chemical clustering[7] on an atomic scale, we include short-range order. Let x be the fraction of Ni, and g be the probability that a neighbor of a nickel atom will be nickel. If we include a nearest-neighbor short-range-order parameter[8] α_1, we have

$$g = x + (1 - x) \alpha_1 \ . \tag{12}$$

Then calling $Z_{\text{eff}} = Z_u$ for this case

$$Z_u = \sum_{ij,nn} N_i N_j \Big/ \sum_i N_i = gz \ , \tag{13}$$

where $z = 12$ is the nearest-neighbor number for the fcc lattice. Thus the criterion for instability toward the formation of a uniform polarization is

$$I_0^{-1} = \Gamma_0 + \Gamma_1 Z_u \ . \tag{14}$$

If the parameters I_0, Γ_1, and Γ_0 were fixed and the concentration were varied, this would give a condition for instability in terms of a critical number of nickel neighbors for a given nickel to have in order for a polarization to appear. We can define

$$Z_c = (1 - \Gamma_0 I_0)/\Gamma_1 I \ , \tag{15}$$

which is the critical neighbor number, in general dependent upon concentration. We then do not need to worry further about the parameters Γ_0, Γ_1, and I_0, but can simply think in terms of the critical neighbor number.

Nonuniform Polarization

We have also made a calculation using a trial solution in which the whole crystal is polarized, but the moment on each Ni depends on its local environment. Such a dependence was studied by Marshall[9] in connection with neutron scattering. In our case, we assume that m_i is a function $m(n)$ of the number n of nickel neighbors surrounding the site i. Then Z_{eff} becomes

$$Z_{\text{eff}} = zg \sum_{nn} p(n, n') m(n) m(n') \Big/ \sum_n p(n) m^2(n) \ , \tag{16}$$

where the sum is now over neighbor number distributions. $p(n)$ is the probability of there being n nickel neighbors

$$p(n) = g^n (1-g)^{12-n} \binom{12}{n} \ , \tag{17}$$

and $p(nn')$ is the joint probability that a pair of nickel neighbors will have n and n' neighboring nickels:

$$p(nn') = \sum_{s=0}^{4} g^{n+n'-2-s} (1-g)^{20+s-n-n'}$$
$$\times \binom{7}{n-1-s} \binom{7}{n'-1-s} \binom{4}{s} \ . \tag{18}$$

The variation of Eq. (16) with respect to $m(n)$

yields a 12×12 system of equations whose secular equation is

$$\det\{gp(nn') - \delta_{nn'}\lambda p(n)\} = 0 \quad . \tag{19}$$

We have found the eigenvalues of this equation, and Z_{eff} is somewhat increased over the uniform polarization case as we expect. The two can be compared in Figs. 1 and 2, which give Z_{eff} for various cases with and without inclusion of short-range order. An interesting feature of the curves in Fig. 1 is that as x approaches 0, the nonuniform Z_{eff} approaches 1. This therefore describes, in this limit, a cluster of two. For the results in Fig. 2, we should ignore the region near $x = 0$ as α_1 should go to 0 with x. We discuss the comparison with localized solutions below.

LOCALIZED TRIAL FUNCTIONS

Spherical Clusters

The extended polarization solutions do not necessarily give the largest value of Z_{eff}. We can ask the question as to whether a localized trial function could give a larger value. That is, we look for a solution in the form of a polarization cloud. We assume that this is based on a small cluster of Ni atoms, and initially we choose a particularly simple kind of cluster, namely, a nickel atom surrounded by N nickel nearest neigh-
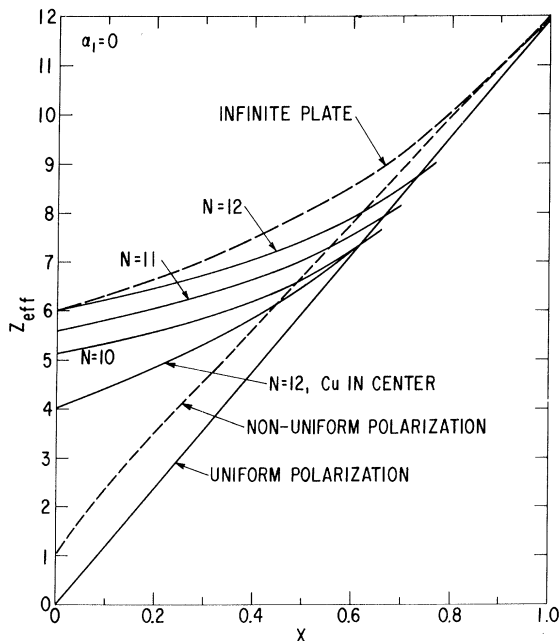
FIG. 1. Effective neighbor number Z_{eff} versus Ni concentration x for various trial polarizations: spherical clusters with N neighbors, infinite plate, uniform and nonuniform extended solutions. No short-range ordering assumed.
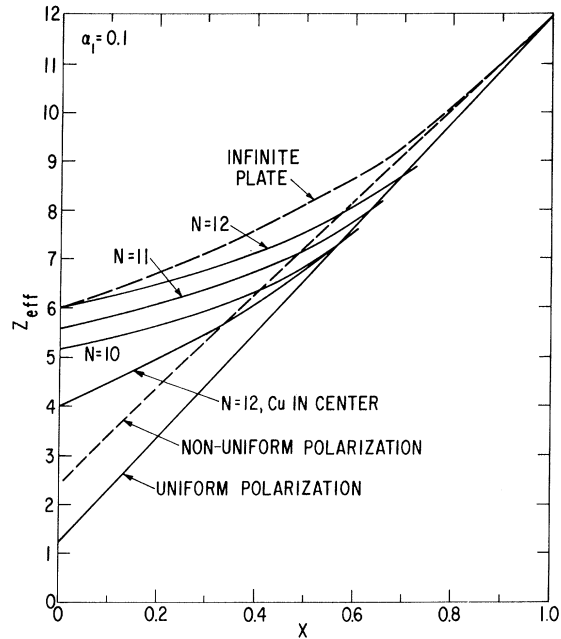
FIG. 2. Same as Fig. 1 but with short-range-order parameter $\alpha_1 = 0.1$.

bors where $N = 10$, 11, or 12. The next-neighbor shell is assumed to have a distribution of nickel atoms characteristic of the crystal. We call this a spherical cluster.

As a trial function we choose the following:

central Ni: $m_i = m_0$,

nearest-neighbor shell: $m_i = m_1$,   (20)

outside cluster: $m_i = m_2 e^{-q_0 R_i}/R_i$.

The moment distribution assumed outside the cluster is of the form we expect to find in an enhanced medium.[4] Using these trial functions we have for the denominator of Eq. (11)

$$\sum_i m_i^2 N_i = m_0^2 + N m_1^2 + {\sum_i}' m_2^2 (e^{-2q_0 R_i}/R_i^2) x \quad , \tag{21}$$

where x is the nickel concentration, and the prime on the summation excludes the nickel cluster. The numerator is

$$\sum_i m_i N_i m_j N_j = 2 m_0 m_1 N + m_1^2 H(N)$$
$$+ 2 m_1 m_2 N x {\sum_j}' e^{-q_0|\vec{R}_{110}+\vec{\rho}_i|}/|\vec{R}_{110}+\vec{\rho}_j|$$
$$+ m_2^2 g x \sum_{ij} e^{-q_0|\vec{R}_i+\vec{\rho}_j|}/|\vec{R}_i||\vec{R}_i+\vec{\rho}_j| \quad , \tag{22}$$

where, in the sums, $\vec{\rho}$ goes over nearest neighbors. Here H is the part of the sum in the nearest-neighbor shell: $H(12) = 48$, $H(11) = 40$, $H(10) = 32$ or 34, the latter if the two Cu atoms are themselves neighbors.

Varying this equation with respect to m_0, m_1, and m_2 yields the determinental equation

$$\det \begin{pmatrix} -\lambda & N & 0 \\ N & H - \lambda N & NxR(q_0) \\ 0 & NxR(q_0) & gxS(q_0) - \lambda xT(q_0) \end{pmatrix} = 0 \quad , \quad (23)$$

where R, S, and T are lattice sums. The largest solution of this equation can be calculated for a range of q_0, and then the maximum found by inspection. This calculation has been carried out by machine, and the resulting values of N_{eff} versus nickel concentration are plotted in Fig. 1 for $\alpha_1 = 0$ and Fig. 2 for $\alpha_1 = 0.1$. These are to be compared with the extended solutions. We have also done the calculation for a cluster with 12 nickels surrounding a copper atom instead of a nickel. The lack of the central nickel reduces Z_{eff} considerably. We see that the localized solutions have larger values of Z_{eff} than the uniform solution for sufficiently small concentration. Thus the model predicts that the instability toward giant localized moments will occur before the instability toward formation of an extended polarization. For x in the vicinity of $\frac{1}{2}$ the two are becoming comparable.

While the magnitude of the moment is not obtained by the present considerations, we can look at the distribution of the moment. In Fig. 3, we show the moments on various neighbors for a typical case, namely, the 11 neighbors $\alpha_1 = 0.1$ case. Also in Fig. 4(a) we plot the parameter q_0 for this case for $\alpha_1 = 0$ and 0.1. It is interesting to note that for slowly varying m_1, Eq. (7) gives a solution of exponential form with $q_0 a = [Z_c/g - z]^{1/2}$ with Z_c defined by Eq. (15). The result in Fig. 4 is close to this result for $Z_{eff} = Z_c$. In Fig. 4(b), we show the distribution of the moment between the atoms on the actual cluster and that of the surrounding region. This is valid, of course, just at the formation point of the moment. We see that for the vicinity of the
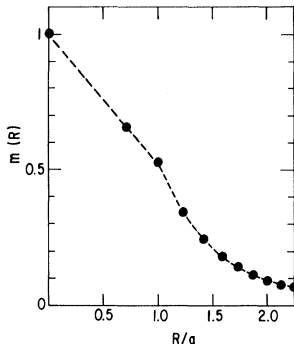

FIG. 4. Total moment and moment on cluster relative to m_0, and (on the right) q_0, versus Ni concentration x for the 11-neighbor case.

50-50 alloy, 75% of the moment is outside the actual cluster.

Platelike Clusters

Recent computer calculations of Cohen[10] show that Cu-Ni alloys should contain flat platelike clusters in $\{111\}$ planes. We have consequently solved also for the case of an infinite platelike cluster. We assume a $\{111\}$ plane of nickel atoms, and assume that the remainder of the crystal has a random composition. A given ion has six neighbors in the plane and three in the neighboring planes on either side. If we assume that the magnetic moment is constant on a given plane, we have

$$Z_{eff} = 6\sum_n (x_n^2 m_n^2 + x_n x_{n+1} m_n m_{n+1}) / \sum_n x_n m_n^2 \quad , \quad (24)$$

where $\delta_{n0}(x_0 - x) + x$ is the nickel concentration on the nth plane. We are assuming $\alpha_1 = 0$ but will later quote the result for α_1 small. Varying with respect to m_n, we have to solve the set of equations

$$(6x_n - Z_{eff})m_n + 3(x_{n+1}m_{n+1} + x_{n-1}m_{n-1}) = 0 \quad , \quad (25)$$

which actually corresponds to a one-dimensional version of Eq. (7). Equation (25) is a linear difference equation which can be solved exactly. We make the ansatz

$$m_n = m_i y^{|n| - 1}, \quad |n| > 0 \quad (26)$$

where y is unknown. Substituting into Eq. (25) for $n > 2$ we have

$$Z_{eff} = 3x(1 + y)^2/y \quad . \quad (27)$$

Then we have from the $n = 0$ and $n = 1$ equations

FIG. 3. Calculated moment distribution in and about a spherical cluster with 11 neighbors, for $\alpha_1 = 0.1$ and Ni concentration $x = 0.5$.
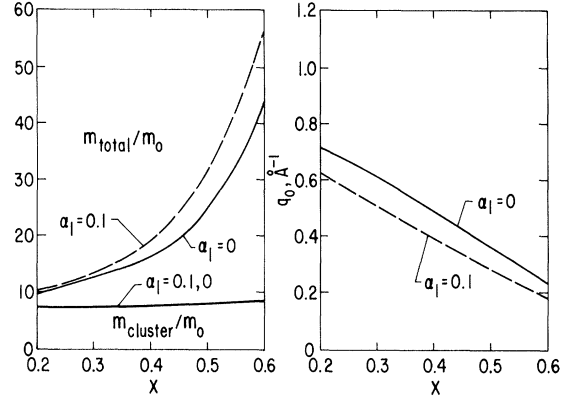
$$(6x_0 - Z_{eff})m_0 + 6xm_1 = 0 \quad ,$$

$$3x_0 m_0 + (6x + 3xy - Z_{eff})m_1 = 0 \quad . \tag{28}$$

From the secular equation together with Eq. (26) we find

$$y = x/(2x_0 - x) \quad , \tag{29}$$

$$Z_{eff} = 2x_0^2/(2x_0 - x) \quad . \tag{30}$$

This is plotted in Fig. 1 for the infinite case $x_0 = 1$. For a finite plate we would approximate by taking x_0 slightly less than 1, say, an average fraction for the cluster of Ni neighbors in the plane. The result plotted is an upper limit to Z_{eff} for finite-sized flat clusters. It appears that the values of Z_{eff} for platelike clusters are comparable to those for spherical clusters.

For the finite α_1 case, the result has been worked out assuming that the concentration x_1 in the first row is x. There is some argument that it should be g, because this plane neighbors the all-nickel plane, but the way we do it is consistent with the assumption we made in the spherical cluster case. The equation corresponding to Eq. (25) can still be solved exactly, and for small α_1, the result is the same as Eq. (30) with

$$x \to \frac{x}{1 - \alpha_1(1 - x)/x_0} \quad . \tag{31}$$

This is plotted in Fig. 2, for $\alpha = 0.1$, and there is very little difference between this and the curve in Fig. 1.

The total moment (for $\alpha_1 = 0$) can be readily calculated:

$$m_{tot} = m_0 x_0/(x_0 - x) \quad , \tag{32}$$

where m_0 is the moment on the plate. For the 50% alloy, half the moment is outside the cluster.

DISCUSSION

Comparison with Experiment

The model we have developed accounts in a general way for the formation of polarization clouds in a Cu-Ni alloy. Using the idea of the effective neighbor number Z_{eff}, we have found that when the concentration of nickel is less than about 50%, a significantly higher number of nickel neighbors can be gathered on the average by localizing an assumed polarization than by spreading it out, at least for the types of clusters considered. Let us consider the cluster consisting of a nickel and eleven nickel neighbors. The occurrence of this cluster can be readily estimated, and it was found[2] to have a concentration roughly equal to that of the polarization clouds observed experimentally. There are certainly other kinds of clusters, but for the sake of argument we shall at this point concentrate

on this typical one. Comparing Z_{eff} for this cluster with that for a uniform polarization (neglecting the "nonuniform polarization" case for the moment) we see that near the critical concentration we can find a value of Z_c such that the cluster is magnetized while the region as a whole is not.

The general character of the moment distribution about this cluster is in fair agreement with neutron diffraction results. Thus from Fig. 4 we have $q_0 = 0.284$ Å when $\alpha_1 = 0.1$ for the 50% alloy. This is to be compared with 0.41 which is the observed value.[1] The total moment for this cluster would agree with experiment (8.5 μ_B) if the central Ni had a moment of 0.27 μ_B which is certainly adequate. One apparent discrepancy is that experimentally q_0 increases with Ni concentration, whereas Fig. 4 shows a decrease. This is probably related to the fact that the exchange enhancement decreases when a medium is polarized.[11] In the paramagnetic region there is no conclusive experimental evidence, and we do expect q_0 to decrease with concentration. It is interesting to note that there should be a change in behavior at the critical percolation concentration [12] of about 20% Ni, below which there are nonoverlapping Ni clusters with copper between. We might expect a rather abrupt decrease in q, and increase in Z_{eff}, as x increases past x_p. This does not occur in the approximation we have used thus far.

As the Ni concentration is increased, the Z_{eff} curve for our cluster crosses that for uniform polarization. We note also that it crosses the nonuniform polarization first. We should point out that the latter represents a better result for the noncluster part of the material than we used in the tail of the local moment solution so that the crossover point is at too low a concentration for this case, and in fact a compromise is probably in order. At any rate, beyond the crossover, the situation is not so clear cut as previously, although it is happily consistent with the fact that the entire system has magnetized by this concentration. To treat this region correctly we should take into account the overlapping of the local moments. It is also desirable to extend the calculation beyond the linear regime when there is finite polarization. Toward much higher nickel concentration we expect that overlap between clusters will be so great that the local moment picture will lose its meaning altogether.

Two more remarks are in order. One is that the short-range-order parameter has more effect on the concentration of the local moments than it does on Z_{eff} and the shape of the moments. The other is to emphasize the fact that the neutron diffraction results for local moments are quite different from what would be observed if the polar-

ization were of the nonuniform extended variety, in which the correlation was with the local environment. The analogous Marshall[9] result was invoked by Cable, Wollan, and Child[13] in interpreting high-Ni-concentration alloys, but is inadequate to describe the lower-concentration alloys.

Let us now consider the fact that our "typical" clusters are only some of many. We expect a variety of different shaped clusters with a distribution of values of Z_{eff}, which goes up to 12. If we sit at a given concentration, say, 50%, and turn on the exchange interaction, this corresponds to lowering Z_c, the critical neighbor number. As soon as Z_c goes below 12, the largest clusters begin to magnetize, and as Z_c is lowered further, more clusters magnetize until they begin to interact, and the polarization clouds begin to overlap. Eventually, the system becomes ferromagnetic.

It is interesting to note that, because the cluster size distribution would fall off rapidly with cluster size and hence with Z_{eff}, most clusters will have Z_{eff} close to Z_c, and the variation in local moment size would not be so great as might be supposed.

Finite Temperature Considerations

It is interesting to speculate on what the results would be for finite temperatures. Clearly as the temperature is raised, it must become more difficult to magnetize a cluster and, in fact, for temperatures approaching the Curie temperature for pure Ni, the actual neighbor number Z_c approaches 12. Above the nickel Curie temperature, no moments can form at all.

It has been observed by van Elst et $al.$,[14] and confirmed recently by Kouvel and Comly,[2] that Cu-Ni alloys in the range of which we speak exhibit a strong paramagnetism outside of the ferromagnetic region. This is understood to be a superparamagnetism due to the local moments. Recently, Griffiths[15] has shown that a disordered Ising model displays a nonanalytic temperature dependence of the susceptibility between the Curie temperature for the pure Ising model and the alloy Curie temperature. While the present system is an itinerant one, we find that the difference is not

very great – in our approximation which amounts to a high-temperature molecular field approximation. For the Ising case, the unenhanced susceptibility goes as $1/T$, whereas we expect the same quantity to go as $\chi_0 + \chi_1 T^2$.

Therefore, the following reasoning would apply roughly to the Ising case also. We have a critical neighbor number which is going through 12 at T_C: Suppose

$$Z_c = 12 - \alpha(T_C^2 - T^2) \ . \tag{33}$$

Near T_C we expect only rather large clusters to magnetize, for which we can take Z_{eff} as

$$Z_{eff} = 12(1 - \beta N^{-1/3}) \ , \tag{34}$$

where N is the number of nickel atoms in the cluster. The reasoning is that a fraction of order $N^{-1/3}$ of the Ni atoms in the cluster are in the surface of the cluster and these have fewer nickel neighbors. Then the size of cluster which is just critical is, upon equating Z_c to Z_{eff},

$$N = [12\beta^3/\alpha(T_C^2 - T^2)] \ . \tag{35}$$

The superparamagnetic contribution to the susceptibility depends on the number of clusters, which goes as X^N. This should, in fact, be the dominant temperature dependence, so that near T_C

$$\chi_{super} \propto e^{-C/(T_C - T)^3} \ , \tag{36}$$

where $C = (\beta/2\alpha T_C)^3 \ln(1/x)$. This is thus the prediction of our model for the nonanalytic behavior of the susceptibility due to the disappearance of the superparamagnetism.

Let us remark, however, that if most of the clusters present are flat plates, then perhaps there will be a temperature T_C' at which these will no longer be able to magnetize. We would then expect a component of χ to go as $e^{-D/(T_C' - T)^2}$, because the "surface-to-volume ratio" is now $N^{-1/2}$.

It would be interesting to observe this effect experimentally.

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Study of Far-Infrared Excitations in Metamagnetic $FeCl_2 \cdot 2H_2O$[†]

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The spin-wave spectra in the antiferromagnetic, ferrimagnetic, and ferromagnetic phases
of $FeCl_2 \cdot 2H_2O$ have been examined at 2 and 6 °K. These spectra are well described by a spin-
wave calculation; it is shown that $g_{\parallel} = 2.23 \pm 0.02$, $S = 2$, and that the exchange interactions are
isotropic. The (large) longitudinal anisotropy is included in the Hamiltonian as a single-ion
anisotropy $(D = +9.58 \pm 0.05$ cm$^{-1})$, which is shown to make an anomolously large contribution
to the spin-wave energies. These results are discussed and interpreted from the point of view
of crystal-field theory. In all three metamagnetic phases, the magnetic resonance modes are
observed to interact with a field-independent excitation with energy 31.5 cm^{-1}, which is pre-
sumably an optical phonon. The measured value of the metamagnetic transition field H_{c2}
$= 45.0 \pm 0.5$ kOe compares quite well with Narath's value of 45.6 kOe, but our value of H_{c1}
$= 35.0 \pm 0.5$ kOe is in poor agreement with Narath's 39.2 kOe. Near H_{c1} (the antiferromagnetic-
to-ferrimagnetic transition field), the far-infrared spectrum appears to indicate that both
antiferromagnetic and ferrimagnetic "domains" coexist over a certain range of field. The
temperature dependence and hysteresis of these domains are also described and compared
with Tinkham's microscopic description of these transitions.

I. INTRODUCTION

Ferrous chloride dihydrate is one of a family of
crystals which exhibit strong exchange interactions
along one crystallographic axis and weak interac-
tions perpendicular to this axis. The initial mag-
netic measurements on $FeCl_2 \cdot 2H_2O$ (or more
briefly FC2) were performed by Narath[1] and are
reviewed in this section. In Sec. II our experimen-
tal techniques and procedures are described. The
results of the far-infrared measurements in all
three metamagnetic regions are shown in Sec. III
to determine both the magnitude *and* anisotropy of
some of the exchange interactions. These results
are interpreted in terms of crystal-field theory
(Sec. IV) and compared to the results of Johnson[2]
and of Inomata and Oguchi[3] (which are in disagree-
ment). In Sec. V the observed coexistence of anti-
ferromagnetic and ferrimagnetic resonance modes
at the same field is described. The results of this
work are summarized and discussed in Sec. VI

and compared to the results on $CoCl_2 \cdot 2H_2O$ (CC2).[4]
One of the most prominent features of the data is
a phonon which is observed to interact with the
magnon states. This interaction is only briefly
described in this paper, as it is discussed in more
detail elsewhere,[5] together with the observation of
a similar level in CC2.

Ferrous chloride dihydrate (FC2) crystallizes
in linear chains of $-FeCl_2-$which run along the c
axis.[6] The exchange interaction J_0 between Fe^{2+}
spins within the same chain is ferromagnetic and
much stronger than the antiferromagnetic interac-
tions between chains. The chemical bonding with-
in a chain is similarly stronger than the weak
forces between chains; hence, the crystals cleave
easily parallel to the c-axis. The crystal sym-
metry is monoclinic with a twofold axis (b) and
a mirror plane (ac). Although the unit cell con-
tains two formula units, these are equivalent.
Narath[1] points out that magnetic canting effects
are not expected because the midpoints between