skii, Dokl. Acad. Nauk SSSR 168, 1283 (1966) [translation: Soviet Phys.-Doklady 11, 522 (1966)].
[9]O. Madelung, Physics of III-V Compounds (John Wiley & Sons, Inc., New York, 1964).
[10]H. Ehrenreich, Phys. Rev. 120, 1951 (1960); also E. Haga and H. Kimura, J. Phys. Soc. Japan 19, 658 (1964).
[11]R. Stratton, Proc. Roy. Soc. (London), Ser. A 246, 406 (1958).
[12]B. V. Paranjape, Rept. Brit. Elec. Allied Ind. Res. Assoc. No. L/T 285 (1953); see also E. M. Conwell, Solid State Phys. Suppl. 9, 1 (1967).

MAGNETISM IN Ni-Cu ALLOYS*

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On the assumption that in Ni-Cu alloys the spin moment on a Ni atom depends on the local atomic environment, it was possible to find moment values for the various atomic configurations so as to give average moments in reasonable quantitative agreement with the values measured in the ferromagnetic composition range. The local environment is specified by the number of Ni nearest neighbors and the number of Ni second-nearest neighbors. This model allows also a consistent qualitative interpretation of the effect on the average moment of low-temperature annealing treatment and of plastic deformation.

The linear decrease in the average atomic moment with Cu content[1,2] of the ferromagnetic Ni-Cu solid solutions has long been interpreted in terms of filling a rigid d band by the extra electron of Cu,[3] with the filling completed at about 53 at.% Cu. However, magnetization measurements[4] showed the presence of permanent moments (and of superparamagnetism) at 70 and even at 80 at.% Cu. The temperature-independent susceptibility[5] and the electronic specific heat[6-8] of Cu-rich alloys increase with the Ni content, suggesting that d states lie at the Fermi surface. Photoelectron energy-spectrum and optical-reflectivity data for Cu-rich alloys[9] show that the Ni solute atoms give rise to a high density of virtual bound d states at and just below the Fermi-energy level. As estimated from the published electronic specific-heat value,[7] the contribution per Ni atom to the density of states at the Fermi surface for alloys with less than 30 at.% Ni is only slightly less than that for pure Ni. Mössbauer isomer-shift measurements with ^{61}Ni in Ni-Cu alloys led to the conclusion[10] that the electronic charge density at the Ni nucleus remains approximately the same over the entire composition range, in agreement with the proposition that the number of d holes per Ni atom stays approximately constant at all concentrations.[11] The interpretation of Cu NMR measurements with alloys containing as little as 1% Ni allows the conclusion that, even at this concentration, d holes are present at the Ni.[12] Thus, various types of evidence indicate that the decrease in ferromagnetic moment with increasing Cu concentration cannot be due to the filling of the d band. A different interpretation is, therefore, needed.

Jaccarino and Walker[13] suggested that the moment associated with an Fe atom in dilute solid solution in Nb-Mo alloys is determined by the number of nearest-neighbor Nb atoms. A similar relationship was found by those authors for the moment on Co atoms dissolved in Rh-Pd alloys. The magnetic moment on Fe in Fe-Al alloys, both disordered and ordered, can be described in terms of the nearest-neighbor atomic environment.[14] The coherent magnetic scattering of neutrons from ordered Fe_3Al showed[15] that Fe atoms, all eight nearest neighbors of which are Fe, have a moment of $2.14\mu_B$ at room temperature, or nearly the same as that of Fe in bcc iron, even though in ordered Fe_3Al all second-nearest neighbors are Al and the moment on the nearest-neighbor Fe atoms is only $1.46\mu_B$. When the number of nearest-neighbor Fe atoms decreases from five to three (or less) the moment at $T=0$ decreases from a value just slightly below the full $2.2\mu_B$ to zero.[14] The near disappearance of Langevin paramagnetism from VAu_4 when the alloy is disordered was also interpreted on the basis of nearest-neighbor atomic environment.[16] In ordered VAu_4 the V atoms, which have a full complement of 12 Au nearest neighbors, possess a permanent moment, and the alloy becomes ferromagnetic at about 45°K.[17] In the disordered alloy nearly all V atoms have at least one V nearest neighbor and, under such conditions, they have no moment.[16] Recent Ag NMR linewidth data taken as a function of the V content in $V_x(Au_{0.8}Ag_{0.2})_{1-x}$ alloy[18] and careful magnetic susceptibility versus temperature mea-

surements for V_xAu_{1-x} alloys[19] seem to be consistent with this interpretation. The moment on an Fe atom with a complete nearest-neighbor shell of Ni[14] or of Co[20] is about $3.1\mu_B$, but the moment on Fe is lower when some of the nearest-neighbor Ni or Co atoms are replaced by Fe (for instance by thermal disordering of $FeNi_3$ or FeCo). In view of these findings, it was undertaken to investigate whether or not it is possible to assign moments to Ni atoms with different local atomic environments in Ni-Cu alloys in such a way as to account for the measured average moment as a function of composition.

A recent neutron nuclear-disorder scattering study[21] established that in a furnace-cooled alloy near the composition NiCu the value of the short-range-order parameter α_1 is 0.121, while $\alpha_2 \cdots \alpha_9$ are all near zero. In the absence of other data it was assumed that α_1 varies with the composition as shown in Fig. 1 and that the other parameters remain near zero in the entire composition range.

By definition

$$\alpha_1 = 1-(1-y)/(1-x), \tag{1}$$

where x is the atomic fraction of Ni in the alloy, and y is the average atomic fraction of Ni in the nearest-neighbor shells around all Ni atoms. From Eq. (1)

$$y = (1-x)\alpha_1 + x. \tag{2}$$

For the second-nearest-neighbor shell the corresponding parameter is equal to x, since $\alpha_2 = 0$. The probability of finding in the alloy a Ni atom with n nearest and m second-nearest Ni neighbors is

$$P_{n,m} = x\frac{12!6!}{(12-n)!(6-m)!n!m!} \times y^n(1-y)^{12-n}x^m(1-x)^{6-m}. \tag{3}$$

The average moment (in Bohr magnetons) per atom in the alloy is

$$\bar{\mu}(x) = \sum_{n=0}^{12}\sum_{m=0}^{6} P_{n,m}\mu_{n,m}, \tag{4}$$

where $\mu_{n,m}$ is the moment assigned to a Ni atom with n nearest and m second-nearest Ni neighbors. It was found that the experimental average ferromagnetic moments can be reproduced with reasonable accuracy, Fig. 1, if the $\mu_{n,m}$ values corresponding to the various local atomic configurations are chosen as shown in Fig. 2. Apart from the weakly ferromagnetic alloys Ni_xCu_{1-x} with $0.42 < x < 0.5$, the largest deviation (1.38%) was obtained for alloy $Ni_{0.85}Cu_{0.15}$. The fit could be undoubtedly further improved by slight changes in the $\mu_{n,m}$ values. Also, these values may have to be altered somewhat when the dependence of the short-range-order parameters on composition becomes known. However, Fig. 1 illustrates the feasibility of interpreting the average atomic moment in Ni-Cu alloys in terms of the nearest and second-nearest neighbor atomic environment of the Ni atoms.

The formation of Ni-rich atomic clusters near the composition $Ni_{0.5}Cu_{0.5}$, that follows[20] from the measured values of the short-range-order parameters,[21] gives rise to magnetic clusters.

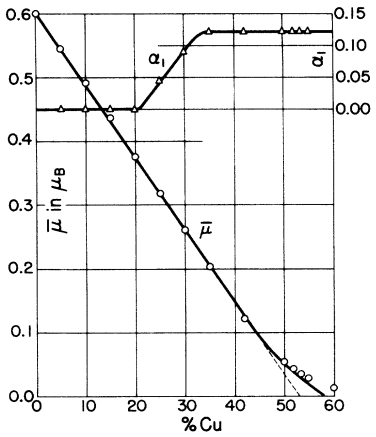

FIG. 1. The average moment per atom, $\bar{\mu}$ (in Bohr magnetons), as a function of composition. Measured values (Refs. 1 and 2 and C. G. Robbins, H. Claus, and P. A. Beck, to be published) are shown by heavy line. Circles represent values calculated from Eq. (4). The assumed variation of short-range-order parameter with composition is shown by curve α_1.

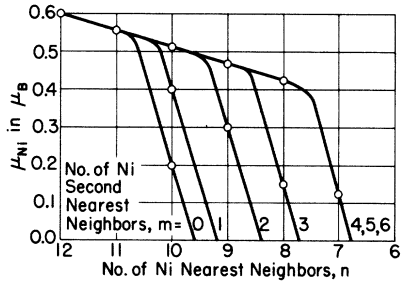

FIG. 2. Moment on Ni atoms as a function of the number n of Ni nearest-neighbors and of the number m of Ni second-nearest neighbors. The only significance of the curves is that they connect the points representing moments at fixed values of m.

The latter were first detected experimentally by virtue of their superparamagnetic effect[5,4] and of the changes in magnetic susceptibility upon low-temperature annealing treatments[5,22,23] which allow the atomic (and hence the magnetic) clusters to increase in size. The magnetic clusters were found to give rise to a low-temperature specific-heat anomaly.[24,25] They were recently studied by means of neutron magnetic diffuse scattering[26] in the weakly ferromagnetic alloys with approximately 50 at.% Cu. This very illuminating study showed that, even in the ferromagnetic state, the alloys in question contain magnetic clusters, which overlap only slightly, so that the ferromagnetic bulk moment can be taken as the sum of the moments of the clusters (approximately $8\mu_B$ per cluster).[27] It was suggested by the neutron work that magnetic clusters of about the same size are present even in the paramagnetic alloy $Ni_{0.4}Cu_{0.6}$. In view of its pronounced superparamagnetism,[4] the alloy $Ni_{0.3}Cu_{0.7}$ undoubtedly also has a very similar magnetic structure, although it is, of course, quite possible that the average size and the moment of the clusters here is somewhat smaller.

It should be pointed out that all the above observations with regard to magnetic clusters, in both the ferromagnetic and the paramagnetic Ni-Cu alloys, are very consistent with the interpretation of the ferromagnetic bulk moment given in this paper. A rough estimate shows that, on the basis of the $\mu_{n,m}$ values given in Fig. 2, a moment of $8\mu_B$ per cluster requires approximately 30 Ni atoms. Nickel clusters of approximately that size may be expected in the required concentration on a statistical basis[28] as a consequence of the measured value of the short-range-order parameters.[21] From the point of view of the present interpretation, one may expect an annealing treatment at 325°C to increase the average atomic moment in alloys near the composition $Ni_{0.5}Cu_{0.5}$. This expectation is based on the following considerations. Such a heat treatment increases the paramagnetic susceptibility,[22,23] without changing the number of magnetic clusters, as inferred from the fact that it does not affect the cluster specific-heat data in Table II of Ref. 25. Thus, one may conclude that, as a result of the low-temperature annealing, the Ni-rich clusters grow in size, while the Ni content of the Cu-rich regions becomes lower. In this way the local atomic environment of a certain fraction of the Ni atoms becomes more Ni rich, so that additional Ni atoms will contribute to the ferromagnetic bulk moment, or they make larger contributions. This is confirmed by the experimental data given in Fig. 3. The magnetization of alloy $Ni_{0.5}Cu_{0.5}$ increased by almost 10% on annealing at 325°C, as compared with the same alloy after annealing at 1100°C and quenching. It was found[25] that plastic deformation increases the magnetic cluster specific heat, i.e., the temperature-independent term of the low-temperature specific heat[24] of alloys near the composition $Ni_{0.5}Cu_{0.5}$. It was suggested[25] that, as a result of slip during deformation, many of the Ni clusters are sliced into two or more smaller ones. This accounts for the observed specific-heat effect, since the magnetic-cluster specific heat is proportional to the number of clusters, regardless of their moment.[24] The subdivision of the Ni clusters into smaller ones may be expected to decrease the average moment of the alloy, since many Ni atoms originally well inside clusters (and, therefore, endowed with near-maximum moment) are located after deformation at newly created cluster-boundary interfaces (thus losing several of their Ni nearest neighbors and much, or even all, of their moment). Figure 3 shows that a considerable decrease in the average moment was in fact observed after deformation in alloy $Ni_{0.5}Cu_{0.5}$.

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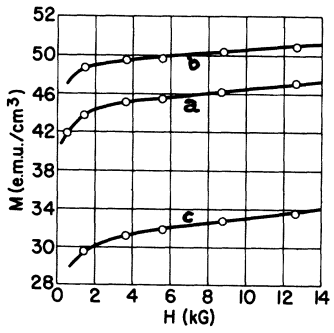

FIG. 3. Effect of heat treatment and of plastic deformation on the magnetization versus field curve at 4.2°K of alloy $Ni_{0.5}Cu_{0.5}$: *a*, after a homogenizing anneal for 72 h at 1100°C and quenching; *b*, after annealing for 24 h at 325°C (following homogenization); and *c*, after heavy plastic deformation (following homogenization).

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[1]S. A. Ahern, M. J. C. Martin and W. Sucksmith, Proc. Roy. Soc. (London), Ser. A 248, 145 (1958).
[2]A. J. P. Meyer and C. Wolff, Compt. Rend. 246, 576 (1958).
[3]N. F. Mott, Proc. Phys. Soc. (London) 47, 571 (1935).
[4]H. C. Van Elst, B. Lubach, and G. J. Van Den Berg, Physica 28, 1297 (1962).
[5]F. M. Ryan, E. W. Pugh, and R. Smoluchowski, Phys. Rev. 116, 1106 (1959).
[6]W. H. Keesom and B. Kurrelmeyer, Physica 7, 1003 (1940).
[7]G. L. Guthrie, S. A. Friedberg, and J. E. Goldman, Phys. Rev. 113, 45 (1959).
[8]K. P. Gupta, C. H. Cheng, and P. A. Beck, Phys. Rev. 133, A203 (1964).
[9]D. H. Seib and W. E. Spicer, Phys. Rev. Letters 20, 1441 (1968).
[10]F. E. Obenshain, J. C. Love, and G. Czjzek, in Proceedings of the Eleventh International Conference on Low-Temperature Physics, St. Andrews, Scotland, 1969, edited by J. F. Allen, D. M. Finlayson, and D. M. McCall (St. Andrews University, St. Andrews, Scotland, 1968), p. 532.
[11]N. D. Lang and H. Ehrenreich, Phys. Rev. 168, 605 (1968).
[12]T. J. Rowland and N. Shiotani, private communication; M. T. Beal-Monod, Phys. Rev. 164, 360 (1968).
[13]V. Jaccarino and L. R. Walker, Phys. Rev. Letters 15, 258 (1965).
[14]T. M. Srinivasan, H. Claus, R. Viswanathan, P. A. Beck, and D. I. Bardos in Phase Stability in Metals and Alloys, edited by P. S. Ruderman, J. Stringer, and R. I. Jaffee, (McGraw-Hill Book Company, Inc., New York, 1967).
[15]R. Nathans, M. T. Pigott, and C. G. Shull, J. Phys. Chem. Solids 6, 38 (1958).
[16]H. Claus, A. K. Sinha, and P. A. Beck, Phys. Letters 26A, 38 (1967).
[17]L. Creveling, Jr., H. L. Luo, and G. S. Knapp, Phys. Rev. Letters 18, 851 (1967).
[18]A. Narath and A. C. Gossard, to be published.
[19]L. Creveling, Jr., and H. L. Luo, Phys. Rev. 176, 614 (1969).
[20]D. I. Bardos, to be published.
[21]B. Mozer, D. T. Keating, and S. C. Moss, Phys. Rev. 175, 868 (1968).
[22]L. E. Hedman and R. D. Mattock, J. Phys. Chem. Solids 23, 955 (1961).
[23]A. Kussmann and H. Wollenberger, Z. Metallk. 54, 521 (1963).
[24]K. Schroder, J. Appl. Phys. 32, 880 (1961).
[25]C. G. Robbins, H. Claus, and P. A. Beck, to be published.
[26]T. J. Hicks, B. Rainford, J. S. Kouvel, G. G. Low, and J. B. Comly, Phys. Rev. Letters 22, 531 (1969).
[27]The magnetic clusters in Ni-Cu alloys appear to be physically quite different from the "polarization clouds" in Pd-rich Pd-Fe alloys. In the latter the "giant moment" is formed around single Fe solute atoms, and its magnitude is determined solely by the strength and the range of the polarization effect of the Fe moment on the surrounding Pd atoms; it cannot be altered by plastic deformation or by heat treatment of the alloy. On the other hand, in Ni-Cu alloys the Cu atoms are most likely not polarized, and the magnitude of the giant moment depends on the number of Ni atoms in the cluster; the latter can be altered by deformation (Robbins, Claus, and Beck, Ref. 25) or by low-temperature annealing (Hedman and Mattock, Ref. 22, and Kussmann and Wollenburger, Ref. 23).
[28]J. B. Cohen, in "Phase Transformations" (American Society for Metals, Cleveland, Ohio, to be published), and private communication. See also J. B. Cohen, Phys. Rev. 139A, 844 (1965).

PROPOSAL FOR OBTAINING ULTRASMALL FERROMAGNETIC-RESONANCE LINEWIDTHS

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It is shown that it should be possible to reduce the dominant two-magnon linewidth well below the smallest value obtained to date by choosing the radius R and thickness S of a thin film of ferromagnetic insulator such that there are no modes degenerate with the main-resonance mode. For a sufficiently thin sample ($S \lesssim 1\ \mu$) the frequencies of the exchange modes are well above that of the main-resonance mode, and for a sufficiently small aspect ratio ($2R/S \lesssim 3\times 10^3$) the low-order magnetostatic modes are also well resolved. In the limit of no two-magnon scattering, the resonance linewidth of yttrium iron garnet at 9.3 GHz approaches 0.2 Oe at 300°K, or a few mOe at 4.2°K.

A method is proposed for substantially reducing the dominant two-magnon contribution[1,2] ΔH_2 to the ferromagnetic-resonance linewidth by making the frequency of the main-resonance mode in a thin film lie well below the frequencies of all other magnetic modes, thus reducing the density of degenerate states to zero. The resulting small linewidths would be important for applications re-