the Symposium on Semiconductor Effects in Amorphous Solids, Picatinny Arsenal, Dover, N. J., 14-17 May 1969 (to be published).

⁶The optical density of states will differ from the true density of states only if matrix elements vary; however, such variations should be $h\nu$ dependent. No $h\nu$ dependence has been observed for $h\nu \leq 11.6$ eV despite the fact that the oscillator sum rule is largely exhausted by 11.6 eV (Refs. 4 and 5). Therefore, we assume that the peak in the optical density of states corresponds to a peak in the density of states.

⁷W. E. Spicer and R. C. Eden, in <u>Proceedings of Ninth</u> <u>International Conference on the Physics of Semiconduc-</u> <u>tors, Moscow, U. S. S. R., 1968</u> (Nauka, Leningrad, U.S.S.R., 1968), Vol. 1, p. 65.

⁸This band structure was taken from D. Brust and E. O. Kane, Phys. Rev. <u>176</u>, 894 (1968). Since it is based on the same pseudopotential as Brust's calculation for disordered Ge, it will differ only because of the lower density used in the latter calculation. However, Brust states that this causes energy shifts of $\sim \frac{1}{3}$ eV at the most. Such shifts are inconsequential for the arguments which are presented here. ⁹Brust and Kane, Ref. 8.

¹⁰T. M. Donovan and W. E. Spicer, to be published. ¹¹Samples intermediate between amorphous and crystalline can also be made by evaporation onto a heated substrate.

¹²The strong peak at -2 eV [labeled (2) in Fig. 3] has been identified with transitions from a wide region of $\langle 100 \rangle$ plane of the Brillouin zone. The peak labeled (3) in Fig. 3 is the precursor of a transition from states near the bottom of the highest valence band at X. Details will be given in Ref. 10.

¹³The electron escape function gives the probability of escape for an electron which reaches the surface with a certain energy.

¹⁴The differences between the EDC calculated by Brust on the direct and nondirect model would have been larger if a more realistic escape function had been used.

¹⁵For $h\nu$ of 8.0 and 9.0, the escape function cuts off the EDC before the peak is fully exposed; however, for $h\nu$ of 10.8 and 11.6 eV the position of the peak is not significantly distorted by the escape function.

¹⁶J. Stuke, private communication.

GIANT MOMENTS IN PARAMAGNETIC Ni-Cu ALLOYS

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Bulk magnetic measurements indicate that polarization clouds with giant moments ($\sim 10\mu_B$) persist well into the paramagnetic composition range of Ni-Cu alloys.

Recent neutron-scattering experiments by Hicks et al.¹ have revealed that in weakly ferromagnetic Ni-Cu alloys (46-50 at.% Ni) the low-temperature spontaneous magnetization is inhomogeneously distributed in magnetic polarization clouds each extending over many atoms. Moreover, it was deduced that the average total moment per cloud (M) is large $[\sim (8-11)\mu_{\rm B}]$ and fairly constant over this composition range, whereas the concentration or number per atom of the clouds (C_F) is small (<1%) and is decreasing almost linearly towards zero at the critical composition for ferromagnetism (~44 at.% Ni). However, since $\mathfrak M$ and C_F were evaluated under the assumption that all the polarization clouds contributing to the neutron scattering are aligned ferromagnetically, the disappearance of C_F at the critical composition was a predetermined result. In fact, neutron measurements¹ on the 44 at.% Ni alloy at 4.2° K (just above its Curie point) showed considerable magnetic diffuse scattering with a wave-vector dependence closely resembling the results for the ferromagnetic alloys, thus indicating that magnetic polarization clouds of similar dimensions

(but fluctuating with time) persist into the paramagnetic phase. Unfortunately, no magnetic scattering was observed for alloys with even less Ni, presumably because their susceptibilities are so small that the 4-kOe polarizing field¹ at the sample has very little effect.

In this Letter, we report the results of bulk magnetization measurements on Ni-Cu which demonstrate the existence of giant magnetic polarization clouds well into the paramagnetic composition range. These results also suggest that the formation of the clouds occurs at local, highly Ni-rich regions and that the onset of ferromagnetism is caused by the aligning forces between neighboring clouds.

Our Ni-Cu alloy samples were cut from coldworked ingots, given a homogenization anneal, and water quenched, as described earlier.¹ Their compositions ranged from 44 to 32 at.% Ni in steps of 2 at.%. A Foner-type vibrating-sample magnetometer was used, and the bulk magnetization was recorded continuously against field (0-25 kOe) or temperature (4.2-300°K).

At 4.2°K the magnetization of each alloy was ob-

served to increase linearly with field from zero and then exhibit a gradual approach to saturation; this saturation effect diminishes at higher temperatures, as noted previously.² As a function of temperature, the magnetization was measured in various low fields and converted to an initial susceptibility (χ_0). Our results plotted as χ_0^{-1} vs T are the solid curves in Fig. 1. It is clear that with decreasing Ni concentration, χ_0 not only gets smaller but becomes more nearly constant at high temperatures. Indeed, these results suggest that a temperature-independent (and fairly composition-independent) contribution to χ_0 is beginning to predominate over a progressively weaker Curie-Weiss-like contribution, or specifically that we may write³

$$\chi_0 = \chi' + C_{\rm CW} / (T - \theta). \tag{1}$$

We have tested our data for each alloy against this simple equation and found that for a particular trial value for the temperature-independent term χ' the plot of $(\chi_0 - \chi')^{-1}$ vs *T* becomes perfectly linear within the experimental error for χ_0 (~2%), as shown by the dashed curves in Fig. 1. This linearity extends over the entire temperature range, except for the alloys close to the



FIG. 1. Temperature dependence of measured χ_0^{-1} (solid curves) and of $(\chi_0 - \chi')^{-1}$ (dashed curves) for various Ni-Cu alloy compositions indicated in units of at.% Ni. Dotted lines show linear extrapolations of latter curves to the temperature axis.

Table I. Experimental quantities (defined in text) deduced from paramagnetic susceptibility data on Ni-Cu alloys.

Alloy comp. (at.% Ni)	$\binom{\chi'}{\left(\frac{10^{-6} \text{ emu}}{\text{g}}\right)}$	<i>θ</i> (°К)	$\left(\frac{10^{-4}\mathrm{emu}^{\circ}\mathrm{K}}{\mathrm{g}}\right)$	C (%)
44	1.7	+42	6.88	0.338
42	1.7	+24	4.80	0.236
40	1.7	+4	3.43	0.169
38	1.8	-2	1.86	0.092
36	1.6	-5	0.91	0.045
34	1.4	-9	0.47	0.023
32	1.3	-7	0.25	0.012

critical composition, which at low temperatures show a curvature in $(\chi_0 - \chi')^{-1}$ vs *T* similar to that exhibited by most ferromagnets just above their Curie points. The values for C_{CW} and θ determined from the linear portions of these curves, as well as for χ' , are listed in Table I; the latter two quantities are also plotted in Fig. 2.

Thus, the initial susceptibility of each alloy consists of a temperature-independent component, presumably arising from a field-induced band



FIG. 2. Closed circles: experimental values for (a) concentration of magnetic polarization clouds C, (b) Curie-Weiss interaction temperature θ , and (c) temperature-independent part of initial susceptibility χ' vs Ni-Cu alloy composition. Open circles: values for ferromagnetic cloud concentration C_F and Curie point T_c reported in Ref. 1. Dotted curves represent atomic probability functions defined in text.

splitting, and a Curie-Weiss-type component, which we attribute to the superparamagnetism of large polarization clouds. In terms of the average total moment (\mathfrak{M}) and concentration (C) of these clouds, the classical Curie-Weiss coefficient is expressible as

$$C_{\rm CW} = NC \mathfrak{M}^2/3k, \qquad (2)$$

where N is the number of atoms per gram and k the Boltzmann constant. Very recently, Foner and McNiff⁴ have observed that the low-temperature magnetizations of these same Ni-Cu samples achieve saturation (plus a small residual differential susceptibility) in fields of 100-150 kOe. When these saturation values⁵ are equated to NCM (and thus to $3kC_{CW}/\mathfrak{M}$)⁶ and combined with our C_{CW} values, it is found in each case that \mathfrak{M} = $(10-12)\mu_{\rm B}$, which is quite consistent with the \mathfrak{M} values deduced for the weakly ferromagnetic alloys.¹ We therefore substituted $\mathfrak{M} = 10\mu_{\rm B}$ and our C_{CW} values into Eq. (2) and calculated the values for the cloud concentration C shown in Table I and Fig. 2(a).

In this figure we also show some dotted curves labeled P_{11-12} and P_{10-12} , which represent the probabilities that a lattice site of these fcc alloys is occupied by a Ni atom surrounded by 11 to 12 or by 10 to 12 Ni nearest neighbors. In computing these probabilities, we assumed that the nearest-neighbor atomic clustering parameter α_1 is 0.1, an approximate value consistent with recent neutron diffraction data on Ni-Cu.^{1, 7, 8} A smaller assumed value for α_1 (including 0 for a perfectly disordered alloy) would lower these probability curves but have little effect on their relative shapes. The two calculated curves in Fig. 2(a) clearly resemble and enclose the experimental results for C versus alloy composition, whereas the analogous curves for P_{n-12} with $n \leq 9$ would lie considerably above. From this correlation we infer that each local, highly Ni-rich region in these alloys is the nucleating site for the spontaneous formation of a magnetic polarization cloud.9 Moreover, the exchange-induced moments in the atomically disordered matrix around each site are probably situated only at the Ni atoms¹⁰ and vary in magnitude with their proximity to the nucleating sites as well as with their local atomic environment.¹¹

values remaining fairly low (lower on a molar basis than those of Pd or Pt). A similar nonsingular behavior has been observed for the lowtemperature differential susceptibility at very high fields.⁴ Hence, critical exchange enhancement is not about to occur but, rather, has already occurred in these paramagnetic Ni-Cu alloys, as manifested in the formation of the giant polarization clouds. The onset of ferromagnetism must therefore arise from a different mechanism, which we believe is suggested by the variation of our experimental values for θ with alloy composition. If θ represents an algebraic average over all the exchange interactions between polarization clouds, we may conclude from Fig. 2(b) that negative (antiferromagnetic) interactions, which predominate when the cloud concentration is very dilute, are overwhelmed by a rapid increase in positive (ferromagnetic) interactions that accompanies the rise in cloud concentration near the critical composition.

Finally, we note in Fig. 2(a) that the curve for C, when extended into the ferromagnetic composition region, appears to be merging with the curve for C_F , the concentration of ferromagnetically aligned clouds reported earlier.¹ Thus, the concentration of clouds that are free to be superparamagnetic, represented schematically by $C - C_F$, is highest at the critical composition and drops quickly but continuously on either side, in qualitative agreement with the interpretation of recent specific heat data on Ni-Cu.¹²

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In Fig. 2(c) we see that χ' , the temperature-independent part of the initial susceptibility, changes very gradually as the critical composition for ferromagnetism (44 at.% Ni) is approached, its

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¹T. J. Hicks, B. Rainford, J. S. Kouvel, G. G. Low, and J. B. Comly, Phys. Rev. Letters <u>22</u>, 531 (1969). ²H. C. Van Elst, B. Lubach, and G. J. Van den Berg, Physica 28, 1297 (1962).

³Expressions similar to Eq. (1) were used by E. W. Pugh and F. M. Ryan [Phys. Rev. <u>111</u>, 1038 (1958)] in describing their χ_0 vs *T* data on some paramagnetic Ni-Cu alloys.

⁴S. Foner and E. J. McNiff, Jr., to be published. ⁵In extrapolating the high-field magnetizations (at 4.2°K) linearly back to zero field, we have used our values for χ' rather than the high-field differential susceptibilities, which are consistently several times larger.

⁶More generally, if there is an appreciable spread in the magnitude of the moments of the different clouds, as recent experiments (see Ref. 4) indicate, the saturation magnetization should be equated to $3kC_{\rm CW}/\mathfrak{M}^*$ with $\mathfrak{M}^* = [\mathfrak{M}^2]/[\mathfrak{M}]$, where the brackets signify averages over the moment distribution.

⁷B. Mozer, D. T. Keating, and S. C. Moss, Phys. Rev. 1<u>75</u>, 868 (1968).

⁸J. W. Cable, E. O. Wollan, and H. R. Child, Phys. Rev. Letters <u>22</u>, 1256 (1969).

 9 The possibility that many of the polarization clouds are nucleated at Fe or other magnetic impurity sites is precluded by the low impurity level of ~30-40 ppm in our samples.

¹⁰N. D. Lang and H. Ehrenreich, Phys. Rev. <u>168</u>, 605 (1968).

¹¹By assuming that the magnitude of each Ni-atom moment in Ni-Cu depends only on its local chemical environment, C. G. Robbins, H. Claus, and P. A. Beck [Phys. Rev. Letters <u>22</u>, 1307 (1969)] obtained an empirical fit to their data for bulk ferromagnetic moment versus alloy composition. However, this assumption ignores purely magnetic correlations, i.e., factors in the local environment that are explicitly magnetic and not uniquely determined by the chemical surroundings. The distance from a Ni-rich nucleating site must be such a factor. Thus, the polarization clouds in Ni-Cu are basically analogous in origin to those in Pd-Fe [see G. G. Low and T. M. Holden, Proc. Phys. Soc. (London) 89, 119 (1966)].

¹²C. G. Robbins, H. Claus, and P. A. Beck, J. Appl. Phys. 40, 2269 (1969).

ASYMPTOTICALLY CORRECT SHELL MODEL FOR NUCLEAR FISSION*

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A two-center shell model with oscillator potentials, $\vec{1} \cdot \vec{s}$ forces, and \vec{l}^2 terms is developed. The shell structures of the original spherical nucleus and those of the final fragments are reproduced. For small separation of the two centers the level structure resembles the Nilsson scheme. This two-center shell model might be of importance in problems of nuclear fission.

From physical intuition it is evident that it is not possible to describe the process of nuclear fission all the way from the ground state of the fissioning nucleus up to the final stage of two separated fragments by means of a one-center shell model like, e.g., the Nilsson model. It is, instead, essential to allow for a preformation of both final fragments in the deformed shell model. This is in fact an additional degree of freedom. In a recent paper this type of single-particle potential was proposed.¹ This potential consists of two connected oscillator potentials, including a spin-orbit force and $\overline{1}^2$ term.² Only the angular-momentum-independent terms have been treated in Ref. 1.

In this paper we report the results of a more realistic calculation including all the *I*-dependent terms and obtain consequently the correct asymptotic single-particle levels for a symmetric two-center shell model.

The Hamiltonian for this potential is

$$H = -\frac{\hbar^2}{2m} \Delta + \frac{m}{2} \omega_0^2 [\rho^2 + (|z| - z_0)^2] - \kappa \hbar \omega_0^0 \{ 2\vec{s} \cdot (\nabla V \times \vec{p}) + \mu [(\nabla V \times \vec{p})^2 - \frac{1}{2} N(N+3)] \},$$
(1)

where \vec{p} is the single-particle momentum and V is the momentum-independent part of the potential. It is noticed that V describes two connected oscillators:

$$V = (m/2)\omega_0^2[(z-z_0)^2 + \rho^2], \quad z > 0;$$

= $(m/2)\omega_0^2[(z+z_0)^2 + \rho^2], \quad z < 0;$ (2)

while the momentum-dependent terms become

$$V(\vec{p}) = -\kappa \hbar \omega_0^{0} \{ 2\vec{\mathbf{1}}_1 \cdot \vec{\mathbf{s}} + \mu [\vec{\mathbf{1}}_1^2 - \frac{1}{2}N(N+3)] \},\$$

$$= -\kappa \hbar \omega_0^{0} \{ 2\vec{\mathbf{1}}_2 \cdot \vec{\mathbf{s}} + \mu [\vec{\mathbf{1}}_2^2 - \frac{1}{2}N(N+3)] \},\qquad(3)$$

where \overline{I}_1 and \overline{I}_2 describe the angular momenta

with respect to the two centers at $z = -z_0$ and $z = +z_0$, respectively. It follows from (2) and (3) that the Hamiltonian (1) indeed contains for $z_0 \approx 0$ the case of a spherical Nilsson potential and for $z_0 \approx R$ (*R* being the nuclear radius) the case of two identical and well-separated potentials of the same type. This behavior is due to the special Ansatz for *V* and is, therefore, automatically present also for the p-dependent terms in (1). The structure of those latter terms is determined by quite general invariance requirements.

The shape of the two connected (amalgamated) nuclei described by (1) is spherical. It is also