

Fock itinerant electron theory of magnetism.

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Interaction Effects between Nearly Magnetic Cobalt Impurities in Gold

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The magnetization of Au-Co alloys was studied over a large range of concentrations ($0.05 < c < 4$ -at. % Co), fields ($0 < H < 70$ kOe), and temperatures ($0.05 \text{ K} < T < 200 \text{ K}$). Magnetization and specific-heat results are well represented by a model with the following characteristics: (a) The isolated Co atoms are not magnetic. (b) The isolated pairs are also not magnetic, but have a Kondo temperature nine times smaller than that of isolated Co atoms. (c) The groups of three or more atoms are magnetic and lead to magnetic ordering at very low temperatures.

I. INTRODUCTION

A. General Survey

Blandin and Friedel¹ have classified the behavior of the transition-metal atoms in noble metals into two types. For the first type, the Stoner condition² for magnetism, $U_{\text{eff}} \rho_d(E_F) > 1$, is verified; here $\rho_d(E_F)$ is the d density of states, and U_{eff} the effective intra-atomic interaction in the Anderson Hamiltonian.³ Thus Cr, Mn, and Fe atoms in Cu and Au hosts carry a magnetic moment. For the second type, the previous condition is not satisfied; thus V, Ti, and Ni atoms in the same hosts are not magnetic. The Co impurity in Cu and Au is between the two kinds of behavior, just at the limit of magnetism.

In the theories of the Kondo effect, it is assumed

that the condition for magnetism is satisfied.⁴⁻⁹ The localized spin S interacts with the conduction electrons, via an antiferromagnetic interaction. This s - d interaction, $-2J\vec{S} \cdot \vec{s}$, leads, in the second Born approximation, to a logarithmic divergence in the scattering cross section as the temperature decreases.⁴ This explains the resistivity minimum.¹⁰ More sophisticated techniques⁸ are necessary to remove this divergence at low temperatures: A new many-body state builds up below the Kondo temperature T_K ; this quasibound state is usually represented by a negative polarization of the electrons around the impurity, leading to a compensation of the local magnetic moment. The relevance of this problem was increased by the experimental work of Daybell and Steyert on the Cu-Fe system,¹¹ and by the suggestion of Schrieffer⁵

that the nonmagnetic impurities in the Blandin-Friedel-Anderson scheme could have very high Kondo temperatures. The magnetic moment on Fe was shown by Mössbauer effect,¹² nuclear magnetic resonance,¹³ and magnetization measurements¹⁴ to be nonexistent. It was believed that the negative polarization of conduction electrons around each impurity was observed in the *Cu-Fe* alloys.¹³

For the impurities of the second type, the theories assume that the condition for magnetism is not realized. But U_{eff} is generally large, so that the impurity is nearly magnetic: Spin fluctuations exist on the impurity site which connect the nonmagnetic to the magnetic state of the impurities. The concept of these *local* spin fluctuations (LSF) has been developed by Lederer and Mills¹⁵ for iso-electronic alloys such as the *Pd-Ni* system. Rivier and Zuckermann¹⁶ have used this concept for all transition impurities in noble metals. These LSF have a lifetime

$$\tau_0 = \pi \rho_d(E_F) [1 - U_{\text{eff}} \rho_d(E_F)]^{-1} = \hbar/kT_K.$$

At low temperatures, below the characteristic temperature T_K , the Pauli susceptibility is enhanced by the same factor

$$\eta = [1 - U_{\text{eff}} \rho_d(E_F)]^{-1}. \quad (1)$$

For $T \ll T_K$, the resistivity varies as $1 - \alpha(T/T_K)^2$. At temperatures above T_K , the frequency of the LSF is lower than the thermal fluctuations of the magnetic moment; the thermal fluctuations then govern the decrease of the susceptibility. A Curie-Weiss law is expected and the LSF behavior will become indiscernable from the behavior of a localized magnetic moment. Levine and Suhl¹⁷ have quantitatively shown this Curie-Weiss behavior. The logarithmic behavior of the resistivity at high temperatures has been obtained by Levine *et al.*¹⁸

Experimentally, the existence of the LSF in iso-electronic alloys has been shown by resistivity,¹⁹ susceptibility, magnetization,²⁰ and specific-heat measurements.²⁰⁻²² At low temperatures, the resistivity of *Pd-Ni* alloys contains a positive T^2 term proportional to the concentration c .¹⁹ The enhanced Pauli susceptibility is nearly constant at low temperatures,²⁰ and also proportional to c . The electronic specific heat, proportional to c , contains a T term, and also a corrective, negative T^3 or $T^3 \log T$ term.²² The study of *Pd-Ni Fe*²³ alloys has confirmed the local character of the enhanced susceptibility.

The LSF properties have also been sought and found in different alloys (nonisoelectronic alloys) of transition impurities in noble metals. Caplin and Rizzuto²⁴ have studied the T^2 resistivity behavior of *Al-Cr* and *Al-Mn* alloys at low temperatures. Recently, Star and Nieuwenhuys²⁵ have also shown the T^2 behavior of the *Cu-Fe* resistivity. The nu-

clear magnetic resonance of Mn in Al has shown the high susceptibility on the Mn atoms.²⁶ Caroli *et al.*,²⁷ taking into account the orbital degeneracy and intra-atomic Hund's rule, have shown that the ratio

$$\xi = \left(\chi \frac{d\gamma}{dc} \right) \left(\gamma \frac{d\chi}{dc} \right)^{-1} \quad (2)$$

of the specific heat to the susceptibility per impurity, is a constant for transition impurities in noble metals. Experimentally, ξ is observed to vary from 0.06 for²⁸ *Cu-Co* to 0.3 for²⁷ *Al-Mn*.

Now, there is experimental evidence that the isolated transition impurities are not magnetic in noble metals at very low temperatures.²⁹⁻³¹ Mn, which is the most magnetic impurity in Cu, has susceptibility behavior, indicating that the moment will disappear at very low concentrations and very low temperatures.³² But the definitive experiment which could decide between the Kondo and the LSF theory remains to be done. In the *Cu-Fe* system, it has been shown recently³³ that interaction effects were in fact responsible for low-temperature deviations from the susceptibility which had been previously taken as experimental evidence⁷ for the existence of a polarization cloud around each Fe atom. In addition, the LSF model can explain the T^2 term of the resistivity²⁵ and the linear temperature dependence of the specific heat^{34,35} below T_K .

In the following part of this paper, we call Kondo temperature T_K *that temperature at which the apparent magnetic behavior of the impurity ceases (the end of the log T term resistivity) when the temperature decreases, even when we are considering the LSF model.*

B. Influence of Local Environment around the Impurity on the Existence of a Magnetic Moment

The problem of the effect of the local environment on the existence of a magnetic moment has arisen in different situations. It had been suggested³⁶ that in *Cu-Ni* alloys, the $1/T$ term in the low-temperature susceptibility was due to the existence of a magnetic moment, carried by small groups of three Ni atoms. But these properties were finally attributed to an Fe contamination.³⁷ Blandin and Friedel¹ have suggested that such a phenomenon may exist in the case of virtual bound states. Because the environment changes the d -state density of impurities at the Fermi level, the condition of magnetism may be realized, and a magnetic moment will appear.

Later, the interaction energy and the change of the density of states were calculated for distant impurities by Caroli.³⁸ The role of the d - d interaction on the appearance of a magnetic moment has been studied by Inoue and Moriya.³⁹ The effect of the environment has been discussed by Kim.⁴⁰

An experimental study of these phenomena has been made on *Cu-Co* alloys.⁴¹ The variation of the magnetization as the cube of the concentration had led to the idea that Co atoms included in groups of three atoms carry a magnetic moment, and that isolated atoms and pairs are not magnetic. Recently, it has been suggested²⁸ that the Kondo temperature is much lower for the pairs than for the isolated impurities.

New considerations on the local-environment effect were introduced by Jaccarino and Walker.⁴² The existence of a magnetic moment on a Fe atom in a *Nb-Mo* matrix, and on a Co atom in a *Rh-Pd* matrix, as in *Mo-Nb* or *Mo-Ti*,⁴³ depends on the number of Mo atoms (Pd, Nb, or Ti) which are first neighbors of Fe (or Co) atoms. A similar model may justify the appearance of magnetism and the presence of giant moments in *Cu-Ni* alloys.⁴⁴⁻⁴⁸

Recently, Claus *et al.*⁴⁹ and Creveling and Luo⁵⁰ have tried to explain the decrease of the susceptibility with *c* in *Au-V* by assuming that a V atom carries a magnetic moment, provided there are no V neighbors within a certain critical radius. Narath and Gossard⁵¹ have modified this point of view because their NMR results suggest that the two types of V sites have different spin fluctuation frequencies or, equivalently, different enhancement factors. Then it seems that the *Au-V* alloys should be an example of a system where short-range interactions favor nonmagnetism. On the contrary, the *Cu-Co* alloys^{28, 41} are a good example of a system where short-range interactions favor magnetism.

In this paper, the *Au-Co* alloys are studied because we expect that the conditions for magnetism are favorable, as in the *Cu-Co* system; but here, we hope that the higher limit of solubility will permit a more exhaustive study of the properties of an alloy where a transition from the nonmagnetic state to the magnetic state is observed.

II. EXPERIMENTAL DETAILS

The samples have been prepared by the metallurgy section of our laboratory from 99.99%-pure cobalt and 99.999%-pure gold provided by Johnson-Matthey. The magnetization of the pure gold is a straight line at 4 and 1 K in fields up to 70 kOe, so that the magnetic impurity level is very small. The alloys were melted in a purified hydrogen atmosphere, inside alumina crucibles, for 4 h at 1150 °C. All the samples were annealed at 950 °C for 24 h, and then quenched in a flow of gaseous hydrogen. The specimens were stored in liquid nitrogen to minimize aging effects. They were only warmed at room temperature when it was necessary to place them in the different measurement holders. The specimens were analyzed by atomic-absorption

spectroscopy.

The standard dimensions of the samples (7-mm diam; *l* = 20 mm) allowed measurements of both the specific heat and the magnetization to be made. The hyperfine specific heat⁵² was measured just after quenching. The magnetization was studied several months later so that small aging effects could be observed for the highest concentrations.

For the magnetization measurements, three apparatus have been used. They are described elsewhere.^{41, 53}

The first one was used to measure the magnetization of alloys of concentrations larger than 1% from 0.05 to 4.2 K in fields up to 25 kOe.

The second device of the same type measures the magnetization from 0.05 to 4.2 K in fields up to 75 kOe. The epibond cell containing the paramagnetic salt, silver wires, carbon thermometer, sample holder, and sample, is moved along the axis of the field for a distance of 3.5 cm. A small coil connected to a ballistic galvanometer is used to detect the change of flux, and hence, the magnetization of the sample. The field on the specimen is produced by a Nb-Ti superconducting coil with $\phi_{int} = 3$ cm, which gives a maximum field of 80 kOe. It is necessary (to eliminate the effects of eddy currents when the sample is moved), to have an homogeneity equal to 10^{-3} on a 5-cm distance. The cold source (iron-ammonium-alum) is adiabatically demagnetized when an external field from a Nb-Zr coil is decreased. The field on the sample is proportional to the current intensity in the coil for high fields. For low fields, a badly known remanent field is added; thus, below 5 kOe, the field values are not well determined.

A third apparatus has been used to measure the magnetization from 1 to 200 K in the field produced by a water-cooled copper coil; an extraction method is also used.

III. EXPERIMENTAL RESULTS

For each alloy listed here, the analyzed concentration is followed in parenthesis by the concentration obtained from the weighed constituents. We used the analyzed concentrations for the calculations and figures.

The alloys of 0.044(0.053)-, 0.088(0.102)-, 0.19(0.20)-, 0.47(0.50)-, 0.74(0.75)-, and 0.94(1.00)-at.% Co concentration have been studied in fields up to 70 kOe, at temperatures between 0.05 and 4.2 K.

The alloys of 0.47(0.50)-, 0.74(0.75)-, 0.94(1.00)-, 1.50(1.50)-, 1.92(2.00)-, 3.15(3.00)-, and 3.86(4.00)-at.% Co concentration have been studied from 0.05 to 150 K.

A. Initial Susceptibility

In *low fields*, a great number of measurements

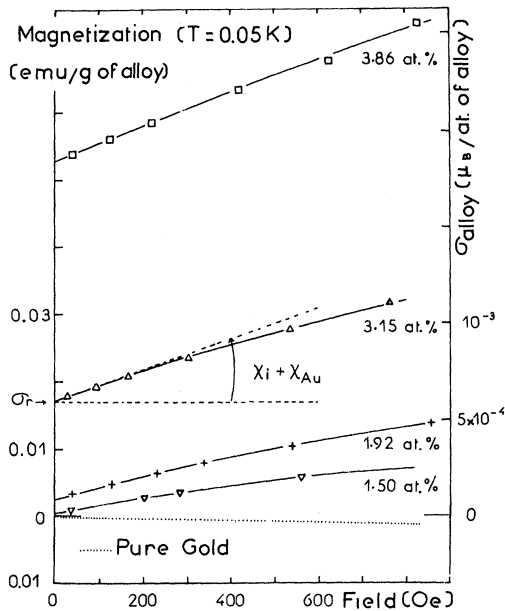


FIG. 1. Magnetization of concentrated alloys in low fields, at $T = 0.05$ K. The linear part permits an evaluation of the initial susceptibility χ_i . The remanent magnetization σ_r rapidly increases with c .

were made in order to *determine the region where the magnetization is proportional to the external field*. The slope of this straight line is called the initial susceptibility χ_i (see, for example, Fig. 1). A plot of $1/\chi_i$ vs temperature is given in Fig. 15 showing the occurrence of magnetic ordering at temperatures below T_N , where χ_i goes through a maximum, and a remanent magnetization appears⁵⁴ (Fig. 1).

In Au-Co, these ordering temperatures T_N are an order of magnitude smaller than those observed, for example, in Cu-Mn alloys of same concentration.⁵⁵ Above the ordering temperature, the initial susceptibility does not follow a Curie-Weiss law (Figs. 2 and 15). We assume that the susceptibility contains contributions from nonmagnetic impurities, and also contributions from a small number of magnetic impurities. The nonmagnetic impurities should contribute as³³

$$\chi_0(T) = C'/(T + T_K).$$

(The characteristic temperature T_K which we have here, should be related to the true Kondo temperature by some proportionality coefficient, which from Ref. 56 is $T_K = 4.5 T_{\text{Kondo}}$. Such a result has never been clearly established experimentally, so T_K will just represent for us the paramagnetic Curie temperature.) For $T \ll T_K$, $\chi_0(T) \approx \chi_0(0) = \chi_0 = \text{const.}$ Then we expect the initial susceptibility may be written for $T_K > T > T_N$:

$$\chi_i = C/(T + \theta) + \chi_0(0) \approx C/T + \chi_0. \quad (3)$$

(The temperature θ , of order T_N , is generally small for the alloys studied,⁵⁴ $T_N < 1$ K for $c < 1.92$ at.%, so that we expect a Curie law to be a good approximation for the contribution of magnetic impurities to the susceptibility.) It can be seen (Fig. 3) that the relation $\chi_i T = C + \chi_0 T$ represents the observed variation of the susceptibility below 10 K very well, providing a good method of separating the magnetic (C) and nonmagnetic (χ_0) contributions. Divergences observed for the 3.15- and 3.86-at.% alloys indicate that for these concentrations T_N is probably already large, and θ cannot be neglected in Eq. (3).

Deviations observed above 10 K for all concentrations, suggest that temperatures of this magnitude may not become negligible in regard to T_K (Fig. 4). To study the behavior $\chi_0(T)$ of the nonmagnetic susceptibilities, the quantity $(\chi_i - C/T)$ is plotted vs temperature (Fig. 5). The net decrease of $\chi_0(T)$, which is observed above 10 K, diminishes around 100 K, and $\chi_0(T)$ becomes approximately proportional to the concentration.

B. Magnetization Curves in High Fields at Low Temperatures

At high fields and *high concentrations* ($c > 0.47$ at.%), the magnetization curves are parallel with each other for the different temperatures. The magnetization at low temperatures (see Figs. 6–8) is composed of two parts: a saturated magnetization which corresponds to the magnetic moments aligned in the field, and another part the slope of which increases with the concentration. In Figs. 6 and 7, the straight line $\sigma = (\chi_0 + \chi_{\text{Au}}) H$ (where χ_0 has been determined as above) is plotted and is to

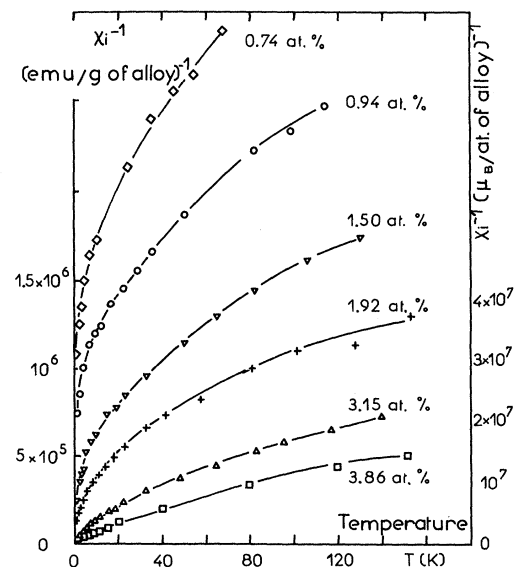


FIG. 2. Temperature dependence of the inverse of the initial susceptibility χ_i due to Co impurities. The Curie-Weiss law is not obeyed.

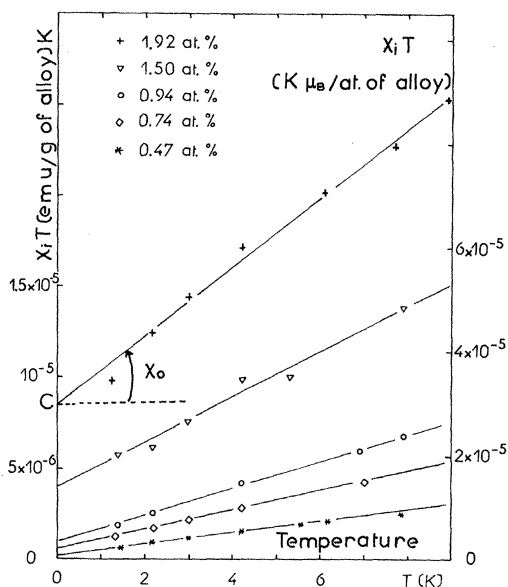


FIG. 3. Plot of $\chi_i T$ vs T . For $T < 10$ K, the law $\chi_i = \chi_0 + C/T$ is obeyed. The alloys of 3.15- and 3.86-at. % Co do not follow such a simple law (see Fig. 4) because their ordering temperature is too high.

be compared to the differential susceptibility $\chi_{20} = (\Delta\sigma/\Delta H)_{H=20\text{ kOe}}$. These two quantities, χ_{20} and χ_0 , are seen to be much the same, considering that an error in the susceptibility determination from low-field data is introduced by the contribution of some Fe impurities (see below). This allows two important observations: The first is that the part σ_{0f} of the magnetization due to the magnetic atoms is *easily saturated*; the second is that the susceptibil-

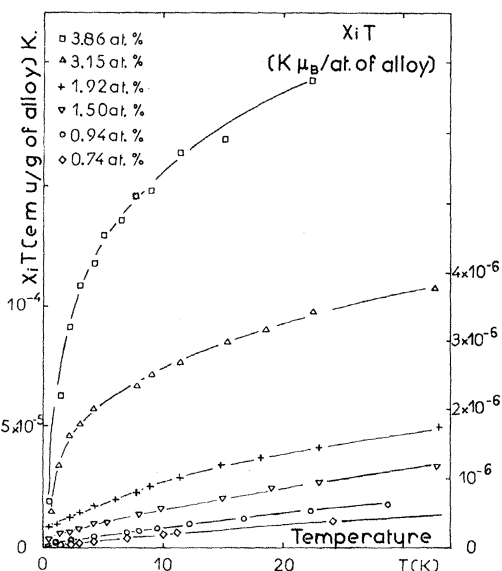


FIG. 4. The initial susceptibility does not follow the law: $\chi_i T = C + \chi_0 T$ above 10 K.

ity of the nonmagnetic Co atoms does not change between 0 and 20 kOe. [For fields much higher than 20 kOe, however, this nonmagnetic part is not exactly proportional to the field for the high concentrations, as may be seen from the slight curvature of the magnetization curves (Fig. 6).]

Since the magnetic contribution is easily saturated, it is possible to obtain σ_{0f} by a linear extrapolation in zero field from the slope of the magnetization in a 20-kOe field (Fig. 7). In Table I, the values of C , χ_0 , χ_{20} , and σ_{0f} are given for the different concentrations.

Contamination by Fe impurities (confirmed by the absorption analysis) make the values of σ_{0f} for the *lower concentrations* of Co (0.044, 0.088, and 0.19 at.%) unreliable. The magnetization of a Au sample prepared in the same manner as our alloys is shown in Fig. 9. In high fields the diamagnetic susceptibility has the correct value⁵⁷ $\chi_{Au} = -1.41 \times 10^{-7}$ emu/g, but the saturation magnetization, which is observed, is of the same order of magnitude as σ_{0f} of the less concentrated alloys. This would correspond to a contamination by about 25 ± 10 ppm of iron impurities (assuming $2.2\mu_B$ per atom of iron⁵⁸). This average iron impurity concentration has been used to estimate correction terms σ_{Fe} and C_{Fe} for σ_{0f} and C for all the alloys, yielding $\sigma_3 = \sigma_{0f} - \sigma_{Fe}$ and $C_3 = C - C_{Fe}$ as the best values for the saturation magnetization and the Curie constant of the magnetic Co atoms. The

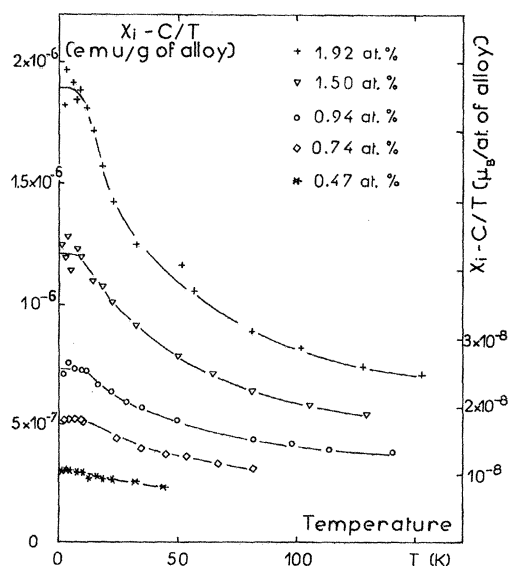


FIG. 5. Temperature dependence of the susceptibility $\chi_0(T)$ attributed to the nonmagnetic Co impurities: $\chi_0(T) = \chi_i - C/T$. A rapid decrease of χ_0 is observed above 10 K; at high temperatures, χ_0 is approximately proportional to the concentration. The one-impurity effects are dominant above 100 K; below 10 K, χ_0 varies more rapidly than c .

values of σ_3 and C_3 are shown in Table I. For the lowest concentrations, χ_0 has been estimated from the slope at high fields, where the Fe impurities are already saturated.⁵⁸

C. Specific-Heat Results

The specific heat has been measured from 0.02 to 0.5 K by Costa-Ribeiro, Souletie, and Thoulouze,⁵² and from 1.2 to 6 K by Crane.⁵⁹ In Ref. 52, both results were analyzed in the following way:

(a) At very low temperatures, below the ordering temperature T_N , a nuclear hyperfine specific heat, due to *magnetic* Co atoms, is observed. The hyperfine term A/T^2 varies as the cube of the concentration. An additional term γT , which increases with the concentration, is observed. It contains an electronic contribution, due to nonmagnetic impurities, which depends on c , and a magnetic term independent of c , analogous to the specific heat of the Cu-Mn system below the ordering temperature.⁶⁰

(b) Around 5 K, the specific heat is proportional to the temperature. The part due to magnetic impurities is negligible because the ordering temperatures are very low. Then Crane's measurements represent the electronic contribution of the nonmagnetic impurities only, and hence give the most accurate values for this nonmagnetic part—which, however, are in good agreement with the very low-temperature values.

In Table II, we give the electronic specific heat for different concentrations, deduced from Crane's work.

IV. INTERPRETATION

A. Model

Co impurities carry no magnetic moment at very

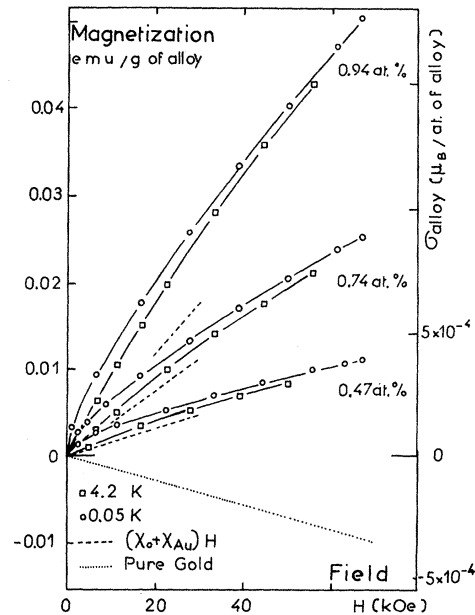


FIG. 6. Magnetization curves of some Au-Co alloys at two temperatures 4.2 and 0.05 K. The small variation of the magnetization with the temperature indicates that the number of magnetic impurities is very small. The dashed straight lines $\sigma = (\chi_0 + \chi_{Au})H$ are compared to the magnetization curves, and the quantity $\chi_0 + \chi_{Au}$ is seen to be equal to $(d\sigma_{alloy}/dH)_{H=20 \text{ kOe}}$. Consequently, all magnetic impurities are saturated in this field and the saturation value is the difference at 20 kOe between the curve and the dashed line.

low concentrations. When the concentration increases, some Co atoms have a moment under some conditions of environment. The following assumptions permit an approximate calculation of all the properties of the alloys.

TABLE I. This table gives for each concentration the following data in emu/g of alloy: the Curie constant C of the magnetic impurities; the contribution χ_0 to the susceptibility of the nonmagnetic impurities; the magnetization slope χ_2 in 20 kOe at 0.05 K; the saturation magnetization σ_{0f} ; the concentration of iron impurities deduced from the average value of the saturation magnetization of the pure gold and the 0.05, 0.1, and 0.2% alloys; the corresponding values of σ_{Fe} and C_{Fe} ; $\sigma_3 = \sigma_{0f} - \sigma_{Fe}$, $C_3 = C - C_{Fe}$.

c at. % Co	0.044	0.088	0.19	0.47	0.74	0.94	1.50	1.92	3.15	3.86
c emu/g				2.3 ± 0.4 $\times 10^{-7}$	6.2 ± 0.2 $\times 10^{-7}$	1.0 ± 0.1 $\times 10^{-6}$	4.0 ± 0.8 $\times 10^{-6}$	8.5 ± 1.0 $\times 10^{-6}$		
χ_0 emu/g	2.8 ± 0.3 $\times 10^{-8}$	4.5 ± 0.5 $\times 10^{-8}$	9.7 ± 0.5 $\times 10^{-8}$	3.0 ± 0.1 $\times 10^{-7}$	5.18 ± 0.1 $\times 10^{-7}$	7.22 ± 0.2 $\times 10^{-7}$	1.22 ± 0.15 $\times 10^{-6}$	1.89 ± 0.1 $\times 10^{-6}$		
χ_2 emu/g				2.93 ± 0.04 $\times 10^{-7}$	5.15 ± 0.04 $\times 10^{-7}$	7.4 ± 0.15 $\times 10^{-7}$	1.34 ± 0.03 $\times 10^{-6}$	2.2 ± 0.15 $\times 10^{-6}$	4.3 ± 0.3 $\times 10^{-6}$	8.3 ± 0.3 $\times 10^{-6}$
σ_{0f} emu/g	1.07 ± 0.05 $\times 10^{-3}$	1.15 ± 0.06 $\times 10^{-3}$	2.7 ± 0.1 $\times 10^{-3}$	2.5 ± 0.1 $\times 10^{-3}$	2.88 ± 0.2 $\times 10^{-3}$	5.15 ± 0.2 $\times 10^{-3}$	1.65 ± 0.1 $\times 10^{-2}$	3.5 ± 0.1 $\times 10^{-2}$	9.0 ± 0.5 $\times 10^{-2}$	1.65 ± 0.1 $\times 10^{-1}$
$c_{Fe} = 25 \pm 10$ ppm Fe emu/g					$\sigma_{Fe} = 1.6 \pm 0.6 \times 10^{-3}$ emu/g			$C_{Fe} = 2.0 \pm 0.8 \times 10^{-7}$ emu/g		
σ_3 emu/g			1.1 ± 0.7 $\times 10^{-3}$	0.9 ± 0.7 $\times 10^{-3}$	1.3 ± 0.8 $\times 10^{-3}$	3.6 ± 0.8 $\times 10^{-3}$	1.50 ± 0.15 $\times 10^{-2}$	3.35 ± 0.15 $\times 10^{-2}$	8.85 ± 0.55 $\times 10^{-2}$	1.64 ± 0.10 $\times 10^{-1}$
c_3 emu/g				0.3 ± 1.2 $\times 10^{-7}$	4.2 ± 1.0 $\times 10^{-7}$	0.8 ± 0.2 $\times 10^{-6}$	3.8 ± 0.8 $\times 10^{-6}$	8.3 ± 1.0 $\times 10^{-6}$		

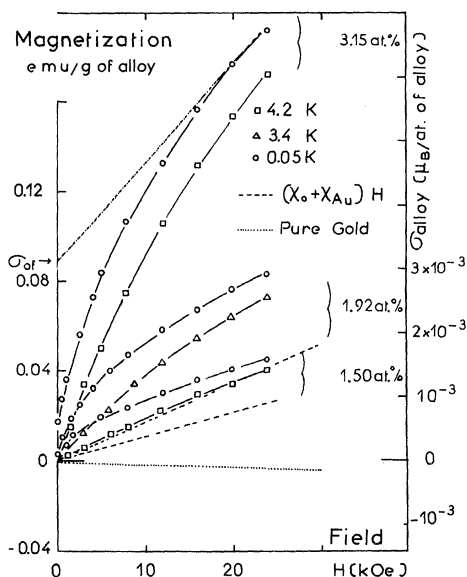


FIG. 7. Magnetization curves of the concentrated alloys. σ_{0f} is determined from the extrapolation to zero field of the magnetization slope at 20 kOe.

A Co atom with no Co first neighbor, and a Co pair, also without a Co first neighbor in the 18 sites around it, carry no magnetic moment; in all other situations, the Co atoms are magnetic.

Let the number of isolated Co atoms in a mole of alloy be N_1 , the number of Co atoms included into isolated pairs be N_2 , and the Co atoms included into isolated groups of three atoms be N_3 . The concentrations being small, it is considered that all other Co atoms in number N_4 belong to groups of four atoms:

$$N_4 = Nc - N_1 - N_2 - N_3, \quad (4)$$

the N being the Avogadro number. In a perfectly disordered alloy, N_1 , N_2 , and N_3 are given by the following formulas^{61, 62}:

$$N_1 = Nc(1 - c)^{12}, \quad (5)$$

$$N_2 = 12Nc^2(1 - c)^{18}, \quad (6)$$

TABLE II. The electronic term $\Delta\gamma$ of the specific heat deduced from Crane's work (Ref. 59) is given with the respective concentration c of Co, of isolated Co atoms N_1/N , and of Co atoms included in pairs N_2/N .

c at. % Co	$\Delta\gamma$ mJ/mole K ²	N_1/N	N_2/N	N_2/N_1
0.57	0.39	5.3×10^{-3}	3.5×10^{-4}	6.6×10^{-2}
0.98	0.85	8.7×10^{-3}	9.6×10^{-4}	1.11×10^{-1}
1.8	1.95	1.45×10^{-2}	2.8×10^{-3}	1.94×10^{-1}
2.0	2.45	1.57×10^{-2}	3.34×10^{-3}	2.13×10^{-1}
2.6	2.96	1.9×10^{-2}	5.05×10^{-3}	2.66×10^{-1}
3.5	5.5	2.28×10^{-2}	7.74×10^{-3}	3.39×10^{-1}

$$N_3 = Nc^3 [24(1 - c)^{22} + 36(1 - c)^{23} + 72(1 - c)^{24} + 18(1 - c)^{24}]. \quad (7)$$

The N_3 is composed of four terms. The first one is the contribution of groups of three atoms which are ordered on an equilateral triangle; the second term is the contribution of right triangles; the third term corresponds to triangles with a vertex angle equal to 120° ; for the fourth term, the three Co atoms are aligned.

In Table III, we give the different values of N_1 , N_2 , N_3 , and N_4 for the different concentrations of our alloys.

B. Saturation Magnetization, Hyperfine Field, and Curie Constant at Low Temperatures

On Fig. 10, the saturation magnetization σ_3 is plotted against concentration on a logarithmic scale. The quantities $(N_3 + N_4)$ and $(N_2 + N_3 + N_4)$ are also plotted on the same diagram to compare with the variation of σ_3 with c . A good agreement is observed, between the $N_3 + N_4$ curve and the experimental points, if it is assumed that each magnetic Co atom, belonging to a group of three or more atoms, carries a mean moment equal to $(1.2 \pm 0.2)\mu_B$.

In Ref. 52, Costa-Ribeiro *et al.* found the coefficient A of the T^{-2} term of the hyperfine specific heat of the same samples to be also proportional to $N_3 + N_4$. From the proportionality factor, they

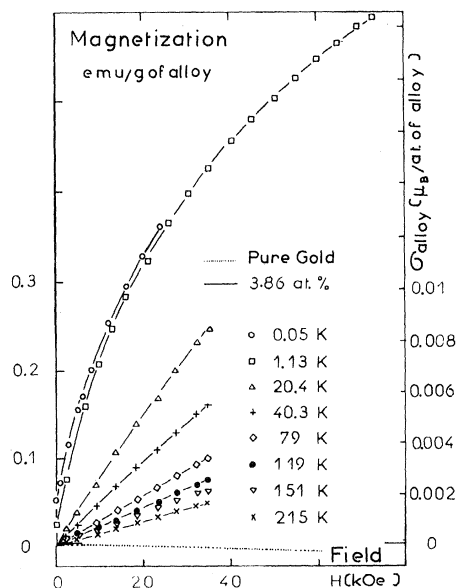


FIG. 8. Magnetization curves of the 3.86-at. % Co at different temperatures. The linear dependence of the magnetization on the external field above 20 K indicates the absence of precipitated Co, even for the most concentrated alloy studied.

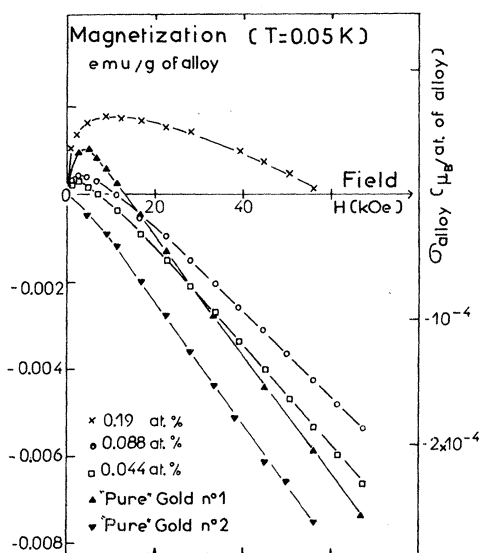


FIG. 9. Magnetization curves of the very dilute alloys at $T=0.05$ K. The magnetization in high fields, $H > 20$ kOe, is a linear function of H . The existence of a saturation magnetization even for two "pure" gold samples, indicates the presence of magnetic impurities. The gold sample No. 1 was melted in a hydrogen atmosphere, exactly like all the Au-Co specimens. The gold sample No. 2 was melted under vacuum. Thus it appears that the magnetic impurities of Fe were mainly introduced by the preparation procedure.

estimated a value of 200 kOe for the hyperfine field H_{eff} on the Co nucleus. H_{eff} is 225 kOe in metallic Co.³³ This suggests that in Au-Co, the value of the moment should not be too different from the value $1.7\mu_B$ which is observed in hexagonal Co.

The Curie constant C_3 also varies with the third power of the concentration (Fig. 11). We may write an expression for C_3 by attributing a spin $3S$ (a ferromagnetic coupling is assumed) to the group of three impurities in number $\frac{1}{3}N_3$, and a spin $4S$ to the group of more than three impurities in number $\frac{1}{4}N_4$ (in our concentration range, the probability associated with groups of more than four atoms is negligible). Then,

$$C_3 = [3S(3S+1)\frac{1}{3}N_3 + 4S(4S+1)\frac{1}{4}N_4] 4\mu_B^2 / 3k. \quad (8)$$

The best agreement between the calculated and experimental values is obtained for $2S=1.7 \pm 0.2$.

The dependence of σ_3 , A , and C_3 on the cube of the concentration, makes it clear that magnetism is associated with groups of three atoms. The quantitative fit can be considered satisfactory for the approximations used. The spin value deduced from the Curie constant is a little larger than that obtained from the saturation magnetization. (Such a difficulty in relating the saturation magnetization to the Curie constant is well known for Co and Ni.³⁴) The value of H_{eff} deduced from specific-heat results

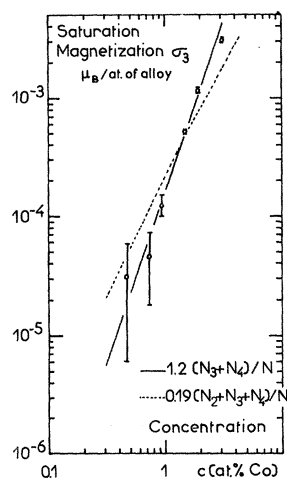


FIG. 10. Concentration dependence of the saturation magnetization σ_3 due to the magnetic Co impurities plotted in a logarithmic diagram; $\sigma_3 = \sigma_0 - \sigma_{Fe}$. The correction σ_{Fe} due to iron impurities, the average concentration being 20 ppm, has been evaluated assuming a magnetic moment of $2.2\mu_B$ per atom of Fe (Ref. 58). σ_3 is proportional to $N_3 + N_4$. The solid line shows the best fit of our data, assuming that each magnetic Co atom, belonging to a group of three or more atoms, carries a mean moment equal to $1.2\mu_B$. The dashed line is obtained by trying to fit the results with the assumption that the pairs are also magnetic, which should lead to a mean moment per magnetic Co atom of $0.19\mu_B$.

however, suggests a value of $2S$ larger than that obtained from saturation magnetization ($1.2\mu_B$). Undoubtedly, a better fit could be obtained by a more precise description of the exact environment which determines the appearance of magnetism on an impurity. A direct microscopic determination of H_{eff} should be useful for this.

C. Susceptibility and Electronic Specific Heat of Nonmagnetic Impurities

The nonmagnetic impurities at very low tempera-

TABLE III. For each concentration c of our samples, we give the concentrations, respectively, of isolated Co atoms, N_1/N ; of Co atoms included in pairs, N_2/N ; and of Co atoms included in groups of three, N_3/N . Here $N_4/N = c - N_1/N - N_2/N - N_3/N$.

c	N_1/N	N_2/N	N_3/N	N_4/N
0.044	4.42×10^{-4}	2.35×10^{-6}	1.30×10^{-8}	6.53×10^{-11}
0.088	8.69×10^{-4}	9.11×10^{-6}	9.96×10^{-8}	1.10×10^{-9}
0.19	1.88×10^{-3}	4.27×10^{-5}	1.01×10^{-6}	2.49×10^{-8}
0.47	4.44×10^{-3}	2.43×10^{-4}	1.39×10^{-5}	5.58×10^{-7}
0.74	6.75×10^{-3}	5.72×10^{-4}	5.07×10^{-5}	5.02×10^{-6}
0.94	8.44×10^{-3}	9.05×10^{-4}	1.02×10^{-4}	1.31×10^{-5}
1.50	1.25×10^{-2}	2.07×10^{-3}	3.58×10^{-4}	7.67×10^{-5}
1.92	1.52×10^{-2}	3.14×10^{-3}	6.79×10^{-4}	1.93×10^{-4}
3.15	2.15×10^{-2}	6.69×10^{-3}	2.22×10^{-3}	1.14×10^{-3}
3.86	2.41×10^{-2}	8.80×10^{-3}	3.43×10^{-3}	2.30×10^{-3}

tures are the isolated atoms and the pairs. The values of their total susceptibility χ_0 are given in Table I. Let χ_1 and $2\chi_2$ be, respectively, the susceptibility of a mole of isolated atoms, and of a mole of isolated pairs. χ_2 is assumed different from χ_1 as has been already shown by Lederer.⁶⁵ Here, they are very different because of the rapid change of χ_0 with the concentration. Then,

$$\chi_0 = \chi_1 N_1/N + \chi_2 N_2/N,$$

so that

$$\frac{\chi_0}{N_1/N} = \chi_1 + \chi_2 \frac{N_2}{N_1},$$

where N_1 and N_2 are defined in Eqs. (5) and (6).

In Fig. 12, we see that the values of $\chi_0/(N_1/N)$ (open circles) are a linear function of N_2/N_1 , for $c < 1.92$ -at. % Co. This fit justifies our assumption and yields

$$\chi_1 = (9 \pm 0.5) \times 10^{-3} \text{ emu/mole of isolated impurities,}$$

$$\chi_2 = (7.6 \pm 0.5) \times 10^{-2} \text{ emu/mole of impurities in pairs,}$$

$$\chi_2/\chi_1 = 8.5.$$

The accuracy of χ_1 and χ_2 is good because results are available for a very large range of concentra-

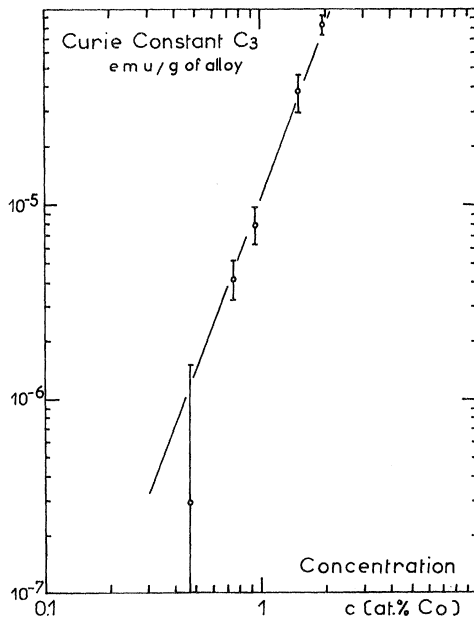


FIG. 11. Concentration dependence of the Curie constant of the magnetic Co impurities C_3 , plotted in a logarithmic diagram; $C_3 = C - C_{Fe}$, C being determined in Fig. 3. The correction C_{Fe} due to iron impurities has been evaluated assuming an effective moment of $3.25\mu_B$ per atom of iron (Ref. 58), the average concentration being 20 ppm. C_3 is proportional to $N_3 + N_4$. The solid line shows the best fit to our data, in a superparamagnetic model, with a spin value of $S = 0.855$ per atom belonging to a group of three or more atoms.

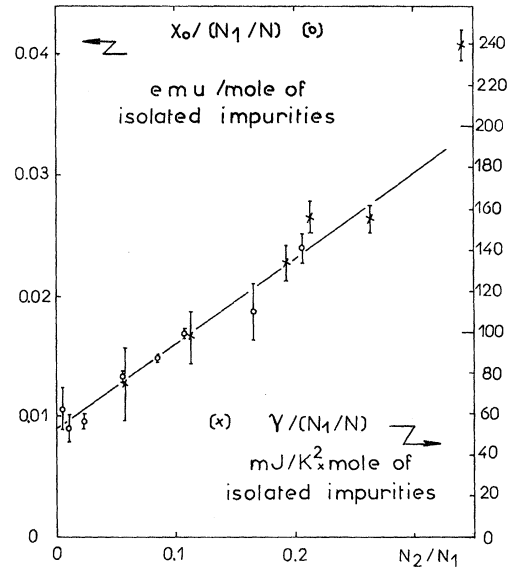


FIG. 12. The nonmagnetic susceptibility χ_0 in emu/mole of alloy (determined in Fig. 3) divided by the concentration N_1/N of isolated Co impurities is plotted vs N_2/N_1 (open circles). The quantity $\gamma_0(N_1/N)^{-1}$, deduced from Crane's work (Ref. 59), is also plotted vs N_2/N_1 (crosses). For $c < 2\%$, these two quantities appear to be linear functions of N_2/N_1 . χ_1 , χ_2 , γ_1 , and γ_2 are deduced from this diagram, and $\gamma_2/\chi_2 \approx \gamma_1/\chi_1$ within experimental error.

tions. If γ_0 is the electronic specific-heat coefficient (Table II), we may write

$$\gamma_0 = \gamma_1 N_1/N + \gamma_2 N_2/N,$$

where γ_1 and $2\gamma_2$ are, respectively, the electronic specific-heat coefficient for a mole of isolated atoms and for a mole of isolated pairs. From a plot of $\gamma_0/(N_1/N)$ vs N_2/N_1 (the crosses in Fig. 12), Costa-Ribeiro *et al.*⁵² deduced from Crane's results⁵⁹:

$$\gamma_1 = 38 \text{ mJ/K}^2 \text{ mole of isolated impurities,}$$

$$\gamma_2 = 520 \text{ mJ/K}^2 \text{ mole of impurities in pairs,}$$

$$\gamma_2/\gamma_1 = 13.7.$$

The errors in γ_1 and γ_2 are more important here because of the difficulties inherent to the specific-heat determination at very low concentration. The fact, however, that we could find an appropriate scale to superpose the experimental points of $\gamma_0/(N_1/N)$ and $\chi_0/(N_1/N)$ on the same line (Fig. 12) indicates that the ratios γ_1/χ_1 and γ_2/χ_2 are the same within the experimental error (a factor 2 on the determination of γ_1 and γ_2). Our analysis permits us to conclude that the nonmagnetic properties are mainly due to two types of impurities which are the isolated atoms and the atoms included in pairs.

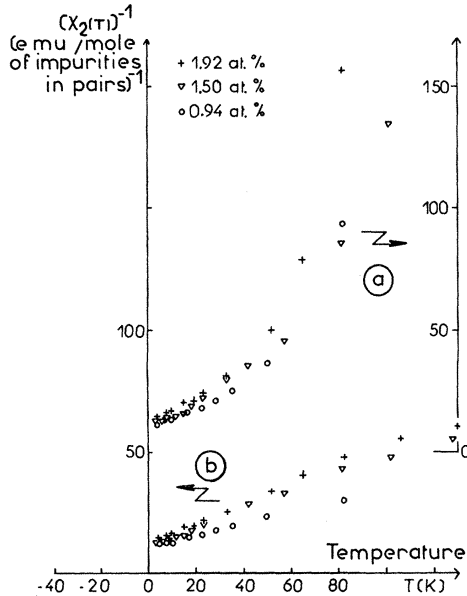


FIG. 13. The inverse of the susceptibility of a mole of impurities in pairs,

$$\frac{1}{\chi_2(T)} = \frac{N_2/N}{\chi_1(T) - C_3/T - \chi_1(0)(N_1/N)},$$

is plotted vs temperature. The χ_1 , χ_2 , and C_3 are expressed in emu/mole. (a) Plotted with the assumption that isolated atoms have a susceptibility independent of the temperature [$\chi_1(T) = \chi_1(0)$ determined in Fig. 3], the figure shows that the Curie-Weiss law of pairs is not followed above 40 K. (b) With the assumption that χ_1 is dependent on the temperature [$\chi_1(T) = C_1/(T + T_{K1}) = \chi_1(0) \times T_{K1}/(T + T_{K1})$], the figure shows that the pair susceptibility obeys the law $2\chi_2 = 2C_2/(T + T_{K2})$ within experimental error. We have taken $T_{K2} = 25$ K and $T_{K1} = T_{K2}\chi_2(0)/\chi_1(0) = 225$ K.

D. Au-Co Alloys and LSF Model

In the LSF model, when the degeneracy and the intra-atomic Hund's rule are taken into account,²⁷ the susceptibility at 0 K is

$$\chi = 2\mu_B^2(2l+1)/(\pi k_B T_K) \quad (9)$$

($l=2$; k_B is the Boltzmann constant).

Let T_{K1} and T_{K2} be the Kondo temperatures for isolated atoms and pairs, respectively. From the measured values of χ_1 and χ_2 we find

$$T_{K1} = 190 \text{ K}, \quad T_{K2} = 22.5 \text{ K}.$$

In the same model,²⁷

$$\xi = \left(\chi \frac{d\chi}{dc} \right) \left(\gamma \frac{d\chi}{dc} \right)^{-1} = \frac{3\mu_B^2}{\pi^2 k_B^2} \frac{d\gamma}{d\chi} \quad (10)$$

is a constant and equal to $3/[2(2l+1)] = 0.3$. Experimentally, we find $\xi = 0.06$ for the isolated atoms, and $\xi = 0.08$ for the pairs. This value compares with the values observed in other systems: $\xi = 0.34$ for Cu-Ni,³⁷ 0.24 for Au-V,⁶⁸ and 0.06 for

Cu-Co.²⁸ (Incidentally, the fact that ξ is different by a factor of 4 to 5 from the theoretically predicted value explains why the values for T_K that we obtain from magnetization differ from the values deduced in Ref. 52 from specific-heat results alone.) We now may try to obtain the thermal variation of $\chi_2(T)$ between 4 and 100 K from the observed variation of the total susceptibility $\chi_1(T)$ in this range:

$$\chi_1(T) = \chi_1(T)N_1/N + \chi_2(T)N_2/N + C_3/T. \quad (11)$$

The magnetic contribution C_3/T has already been determined (Sec. III A). As a first step, we may assume the thermal variation of $\chi_1(T)$ is small in the range 0–100 K because of the high value of T_{K1} . In Fig. 13, we have plotted

$$\frac{1}{\chi_2(T)} = \frac{N_2/N}{\chi_1(T) - C_3/T - \chi_1(0)(N_1/N)}$$

vs temperature for different concentrations.

The same Curie-Weiss law of the form⁶⁷ $\chi_2(T) = C_2/(T + T'_{K2})$ with $T'_{K2} \approx 25$ K is observed for the different concentrations at temperatures below 40 K. [The deviation from the Curie law observed above 40 K may be removed if we assume that the thermal variation of $\chi_1(T)$ is also given by a Curie-Weiss law $C_1/(T + T'_{K1})$,⁶⁷ with $T'_{K1} = T'_{K2}\chi_2(0)/\chi_1(0) \approx 225$ K; T'_{K1} and T'_{K2} are not very different from the values obtained from Eq. (9).]

From the Curie constants, we can obtain the effective moments and the spin values.

In a *simple paramagnetic model*, we write

$$C = N\mu_{\text{eff}}^2/3k \quad \text{with } N = N_1, N_2, \text{ or } (N_3 + N_4),$$

which gives an effective moment and spin value per Co atom, respectively, when isolated,

$$\mu_{\text{eff}} = (4 \pm 0.1) \mu_B, \quad S = 1.57 \pm 0.05,$$

when in pairs,

$$\mu_{\text{eff}} = (3.9 \pm 0.1) \mu_B, \quad S = 1.5 \pm 0.07,$$

when in groups of three or more,

$$\mu_{\text{eff}} = (3.5 \pm 0.4) \mu_B, \quad S = 1.35 \pm 0.15.$$

In a *superparamagnetic model*, we have

$$C_2 = \frac{1}{2}N_2\mu_{\text{eff}}^2/3k = \frac{1}{2}N_2^2g^22S(2S+1)\mu_B^2/3k,$$

which gives $\mu_{\text{eff}} = (5.5 \pm 0.2) \mu_B$ per pair and $2S = 2.35 \pm 0.25$. In the same way, we have

$$C_3 = [3S(3S+1)\frac{1}{3}N_3 + 4S(4S+1)\frac{1}{4}N_4]g^2\mu_B^2/3k,$$

which leads to a spin value per atom of $S = 0.855 \pm 0.075$. In the paramagnetic model, we get the same spin value 1.5 ± 0.25 per atom within experimental error. But the superparamagnetic model leads to a smaller value of S for the Co which are in groups, which agrees better with the value that may be deduced from saturation measurements in hexagonal Co ($gS\mu_B = 1.7\mu_B$). The accuracy of the

experiments seems insufficient to determine the exact paramagnetic behavior of Co atoms.

E. Magnetic Ordering at Low Temperatures

The groups of localized magnetic moments are statistically distributed in the matrix. As for *Cu-Mn* alloys, the magnetic ordering at very low temperatures is characterized by the appearance of a remanent magnetization. For a Rüdernann-Kittel-Yosida (RKY) type of interaction,⁶⁸ it has been shown that the magnetization and the specific heat, per atom of impurity, scale as universal functions of T/c and H/c (H is the external field). This has been verified, for example, on *Cu-Mn*⁶⁸ for the magnetization, and in particular for the remanent magnetization.

The *Au-Co* remanent magnetization divided by $N_3 + N_4$ is approximately described by a unique curve as a function of $T/(N_3 + N_4)$ (Fig. 14), where the concentration of magnetic impurities varies by a factor $(3.86/1.50)^3 = 17$ for the results on this figure. Over the same range of concentration, we observe that the initial susceptibility in the ordered state is only varying by a factor 3 (Fig. 15). The small increase with concentration is partly due to the reduction of the electronic mean free path of conduction electrons by the nonmagnetic impurities,⁶⁹ but is mainly due to the susceptibility of the nonmagnetic impurities, which is contained in the initial susceptibility. Then the fact that the suscep-

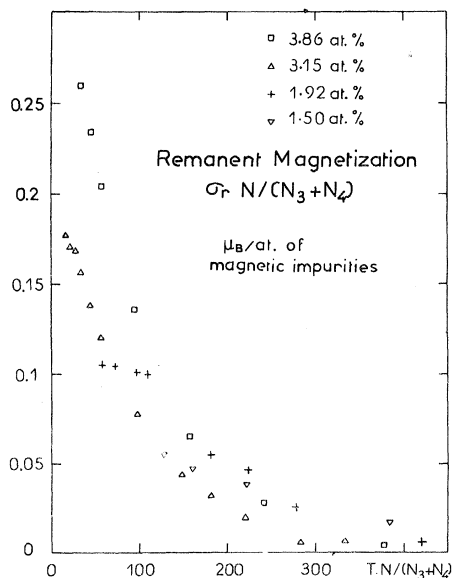


FIG. 14. The remanent magnetization σ_r of different *Au-Co* alloys divided by the concentration $(N_3 + N_4)/N$ of magnetic Co impurities is plotted vs $T[(N_3 + N_4)/N]^{-1}$. As in the case of *Cu-Mn* alloys (Ref. 67), we observe an approximately universal curve, indicating that the remanent magnetization appears at a temperature proportional to $N_3 + N_4$.

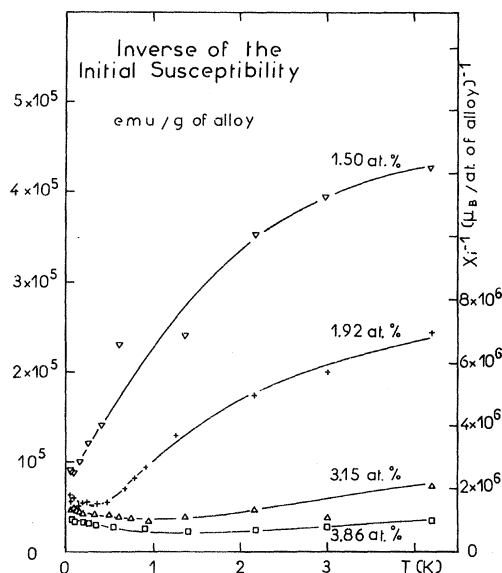


FIG. 15. Temperature dependence of the inverse of the initial susceptibility at low temperatures. The ordering temperature is approximately proportional to the cube of the concentration. The susceptibility at the lowest temperature depends only slightly on the concentration in magnetic impurities, although the latter varies by a factor of 17.

tibility due to magnetic impurities is nearly independent of c , as for the *Cu-Mn* alloys,⁵⁵ clearly indicates the $1/r^3$ main character of the interaction between groups of three or four atoms.

F. Pair Magnetization in High Fields

It is known that the magnetization of *Pd-Ni* alloys is only approximately a linear function of the external field at low temperatures.²³ Doniach⁷⁰ attributes this deviation from linearity to a decrease of the exchange enhancement factor η [Eq. (1)] with increasing field; this property should be better observed on impurities with a high susceptibility at low temperatures because a larger magnetic moment is produced in an external field, and then larger deviations from the linearity are observed.

In *Au-Co* alloys, the susceptibility of Co pairs is of the same order of magnitude as the susceptibility of Ni in Pd.²² The curvature is not important at low concentrations, because the number of Co pairs is small compared to the number of isolated atoms. But the curvature which appears in high fields for the 0.74- and 0.94-at.% alloys (Fig. 5) shows that for these alloys the factor η is reduced by an increasing external field.

V. CONCLUSION

In conclusion, all the magnetic properties of *Au-Co* alloys may be described by a simple model of nearly magnetic impurities. Isolated atoms have

a Kondo temperature of several hundred K and produce at low temperatures some physical properties proportional to the concentration. Pairs of first-neighbor Co atoms have a Kondo temperature nine times lower and the physical properties vary as c^2 . When a third atom is in the proximity of a pair in a vaguely known position, magnetism appears; magnetic ordering and hyperfine fields on Co nuclei are produced at very low temperatures. The number of magnetic impurities varies as the third power of the concentration.

The interaction produces a transition from non-magnetism to magnetism for the Co atom. These results may be compared with the Knight-shift results on Au-V alloys.⁵¹ In this case, the conclusions are exactly opposite; a V atom in the position of a first neighbor decreases the exchange enhancement factor. Other changes of magnetic properties have been observed on Ni and Co impurities. A magnetic moment appears on a Ni atom when it has a critical number 8, 9, or more first-neighbor Ni atoms.⁴⁶ In the Cu-Ni nonmagnetic region, the electronic specific heat and the susceptibility vary more rapidly than strict proportionality to the concentration.³⁷ Here, it is due to the increase of the exchange enhancement factor with the concentration.⁴⁷ The Cu-Co and Au-Co alloys have similar properties.²⁸ It is also possible to distinguish the different contributions of pairs and groups of three atoms in Cu-Co alloys. It has been recently shown⁵⁸

that nearly magnetic Fe impurities become magnetic in Cu, when a Fe atom has a neighbor inside a critical radius r equal to 6 Å. Nearly magnetic pairs of Fe atoms ($6 \text{ Å} < r < 11 \text{ Å}$) increase the low-field magnetization and cancel the one-impurity effects of very isolated atoms ($r > 11 \text{ Å}$).

Fe impurities introduced into nonmagnetic Pd-Ni²³ and Cu-Ni^{45, 71} alloys create some magnetic moments on Ni atoms inside a critical radius $r \approx 9 \text{ Å}$ for the first alloy, and on the first-neighbor shell for the second alloy.

Thus, this phenomenon of the influence of interactions on the appearance of magnetism, which we have described in Au-Co alloys, plays an important role in all physical properties of this type of alloy because many impurities are near the limit of magnetism.

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