**B. — DISORDERED METALLIC ALLOYS.
MAGNETISM IN DISORDERED METALLIC SYSTEMS :
THE LOCAL ENVIRONMENT EFFECTS.**

**INHOMOGENEOUS CHARACTER OF THE TRANSITION NON MAGNETISM-
MAGNETISM IN ALLOYS THROUGH MAGNETIC MEASUREMENTS**

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Résumé. — Les mesures magnétiques permettent une étude détaillée de la transition du non-magnétisme au magnétisme d'un alliage. L'étude d'alliages où cette transition a lieu pour de faibles concentrations permet de déduire un modèle simple reliant le comportement d'une impureté aux interactions avec les impuretés voisines. Pour les alliages où cette transition a lieu dans une gamme plus élevée de concentration, il n'est plus possible de déduire un modèle de la seule analyse des résultats expérimentaux. L'extension du modèle trouvé pour les alliages dilués permet une compréhension correcte des principaux phénomènes caractéristiques des alliages concentrés (superparamagnétisme de moments géants notamment). Les effets de polarisation de sites passifs par des sites actifs, mis en évidence sur des alliages ternaires, complètent un modèle du comportement des alliages au cours de la transition.

Abstract. — Magnetic measurements allow a detailed study of the transition from non-magnetism to magnetism of an alloy. The study of alloys in which this transition occurs for low concentrations allows the deduction of a simple model relating the behavior of an impurity with neighbouring impurities. For alloys in which the transition occurs in a higher concentration range, a model can no longer be deduced from the simple analysis of experimental results. The extension of the model found for dilute alloys allows a good understanding of the main characteristic phenomena of concentrated alloys (particularly the superparamagnetism of giant moments). Polarization effects of passive sites by active ones, clearly shown on ternary alloys, provide a more detailed model of the behavior of alloys in the transition range.

1. Introduction. — Magnetic measurements have been proved to be a very useful tool in the study of environment effects, because of their great sensitivity to local or far environments. Our purpose is to examine critically the total information contained in these measurements, and to discuss what can be strictly deduced from them, what is simply suggested or even what is only a support of a model proposed elsewhere. In fact, the distinction is rarely clear between a simple hypothesis and experimental evidences, so that the conclusions drawn are not always logical. It will be necessary to keep in mind the limitations of the experiments, and therefore the limitations of the interpretations given.

Another reason of incommunicability between physicists is that a lot of terms are not understood in the same way by different authors. Our attempt will be mainly to clarify our meaning of these terms, which are often more picturesque than realistic, and to handle them with care, owing to the limitations contained in their definitions.

In a first part, we try to explain what may be expected from magnetic measurements, and how to analyze them in a logical way. This type of analysis is then

applied to a dilute alloy, CuFe, for which the transition from a paramagnetic state to an ordered state can be observed for very low concentrations. The extension of this analysis to an intermediate alloy, AuCo, allows a refinement of the model deduced from the CuFe analysis.

— In a second part, we propose a simple-crude model of the same type as previously which is tested with available experimental results on several concentrated alloys. The model is shown to be good in view of its crudeness and of accuracy of the analysis of experimental data.

— In a third part, we describe another mechanism which plays a dominant role to build up the giant moments : The polarization of nearly magnetic atoms (passive sites) by the neighbouring magnetic ones (active sites). This effect appears clearly from the study of ternary alloys, and is able to correct some discrepancies of the previous crude model.

— In the fourth and last part, some words are said about order phenomena occurring in concentrated alloys, and the notions of order temperatures and critical concentrations briefly discussed.

2. Magnetic measurements and dilute alloys. —

2.1 WHAT MAY BE EXPECTED FROM MAGNETIC MEASUREMENTS ? — All usual magnetic measurements consist in drawing magnetic isotherms $M(H)$ for a variety of temperatures and concentrations. If we restrict ourselves to measurements well above the ordering temperatures the magnetization curves start linearly from the origin and tend to an asymptot for sufficiently high fields and low temperatures. The initial slope is denoted χ_i (susceptibility), and the asymptot is described by its slope χ_F and its zero intercept σ . These three quantities are the fundamental magnetic variables, which are then to be analyzed versus temperature and concentration.

The only use of these quantities looses a part of the information contained in the total curve, especially the way it approaches the asymptot. Unfortunately, it is not possible to describe this fact in a numerical way. The use of Brillouin functions is criticable in the presence of interactions between magnetic moments, and for other reasons to be discussed later. So, for the present time, this evaluation must be let to the experimentalist's eye.

The analysis of magnetic results assumes that the total magnetization curve is the superposition of several parts. Schematically, one may distinguish in a curve a magnetic and a non magnetic contribution. The magnetic part saturates in sufficiently high fields at a value independent on the temperature T , and has a Curie Weiss like initial susceptibility. The non magnetic part is linear *vs* H , with a Pauli-like temperature independent susceptibility. A typical example of a magnetic behavior (in our sense) is a paramagnetic salt, whereas a non magnetic behavior is typically found in pure copper. Therefore, the meaning of the three magnetic variables is clear : σ represents the total moment of the magnetic part, χ_F the susceptibility of the non magnetic part. In χ_i , we find the contribution of the two parts : $\chi_F +$ a Curie Weiss term. Of course, things are a bit more complicated in practical cases, but for the present time, it is sufficient to keep this basic scheme in mind.

To analyse the magnetic variables versus concentration, it is obviously always possible to expand a variable in series of the concentration c ; but this expansion is significant only if its number of terms is much smaller than the number of measured concentrations. Two or three terms seem to be a maximum number in view of the accuracy of available results. Moreover, we must not forget that this analysis is fruitful only if we are able to interpret the different terms. In fact, this occurs when the concentration is low enough to neglect higher powers of c in the expansion (dilute alloys, $c \ll 1\%$). Then a magnetic variable $V(c, T)$ may be written

$$V(c, T) = V_0(T) + cV_1(T) + c^2 V_2(T).$$

V_0 is usually taken as the matrix contribution, which can be subtracted ; $V_1(T)$, which is the only c depen-

dent term for vanishing concentrations, is interpreted as a *single impurity* term, and $V_2(T)$ first evidence of a departure from linearity in c , is taken as the manifestation of interactions which modify the behavior of some impurities. In fact, no impurity is really *single* in an alloy of non zero concentration, but is submitted to interactions from all other impurities. We must expect a distribution of magnetic behaviors, each impurity behaving according to its own environment. In the present state of experiment, it is not possible to deduce detailed informations on this distribution. But assuming a reasonable distribution makes possible consequent conclusions on the concentration dependence of the bulk alloy properties [1]. They can be written as higher, V_1 and V_2 being only mean values.

Without knowing anything on the interactions but their $1/r^3$ decrease, we can understand this in a less realistic, but perhaps more concrete way : When an impurity has no other impurity within a certain radius r_0 , experiment fails to detect any difference between its behavior and that of a really isolated impurity. On the contrary when it has another impurity within this radius, it feels the interaction in a detectable way, giving a c^2 term, usually called a pair term for evident reasons. Let us outline that r_0 is determined in relation with experimental accuracy and sensitivity, and is not an intrinsic range of the interactions. An important consequence is that r_0 will be large if the modification of behavior of impurities is important, and small (nearest neighbors) if the modification has weak effects on the magnetization variables. This empirical critical distance can be determined as shall be seen on the following examples.

2.2 ANALYSIS OF THE CuFe MAGNETIZATION DATA. [2, 3]. — The σ variable is successfully analyzed in concentration by the formula :

$$\sigma = \sigma_1 c + \sigma_2 c^2$$

and the final susceptibility is found to be proportional to the concentration (Fig. 1).

For a better interpretation, we can use an alternative point of view, in analyzing directly the magnetization curves versus c (Fig. 2).

$$M(H, T, c) = \chi_0 H + cM_1(H, T) + c^2 M_2(H, T).$$

Here, $\chi_0 H$ is the contribution of the Cu matrix. $M_1(H, T)$, contribution of isolated impurities, is almost linear in field (with a little σ_1 value which is the same as previously), with an initial susceptibility well described versus T by a Curie Weiss law (Fig. 3)

$$\chi_1 = \frac{C_1}{T + 29}$$

from the value of the Curie constant, we deduce an effective spin of 1.35 by isolated impurity. $M_2(H, T)$ presents a magnetic behavior (Fig. 3) saturating at 1.3 K in 60 kOe. The initial susceptibility is more or less described by a Curie-Weiss law with a negative

FIG. 1. — CuFe alloys. a) Linear variation of the high field susceptibility vs. concentration. b) σ/c vs. c linear plot, proof that σ may be written $\sigma = \sigma_1 c + \sigma_2 c^2$.

FIG. 2. — An example of a determination of $M_1(H)$ for CuFe alloys at a given temperature. $M_1(H)$ is the zero intercept of the curves and $M_2(H)$ their slope.

FIG. 3. — A summary of the analysis vs. concentration and temperature on CuFe alloys. a) Isolated impurities contribution. b) Pairs contribution. c) The susceptibility of isolated impurities is well described by a Curie Weiss law. d) The mean susceptibility of pairs obeys more or less a Curie Weiss law.

Curie temperature of a few K. If we assume that magnetic moments are described by an effective spin S with a concentration C , the Curie constant C_2 and the saturation magnetization σ_2 can be written :

$$c_2 = \frac{N_A C s(s+1) g^2 \mu_B^2}{3 k} \quad \sigma_2 = N_A C s g \mu_B$$

from these two data, it is possible to deduce S and C . We find $S = 2 \times 1.35$, and $C = 130 c^2$.

Up to now, the result is clear : the c term is due to isolated Fe atoms, with a characteristic temperature of 29 K, and the c^2 term is due to ferromagnetically coupled pairs of Fe atoms. If we try to represent this behavior in a statistical model, assuming moreover that there are as many ferromagnetic as antiferromagnetic pairs [1], we find that a Fe atom is isolated if it has no Fe atom in its 520 neighbouring sites. If there is one impurity among these sites, it modifies the behavior of the central impurity, the modification being symmetric on the two impurities, which make a pair. But it is not realistic to say that all Fe atoms in pairs are magnetic (characteristic temperature $T_K \approx 0$) and that suddenly, the T_K becomes 29 K. We expect intermediate behaviors when the distance between impurities decreases.

This expectation is strongly supported by two magnetic facts : very low temperature susceptibility measurements [4] analysed in the same way with the same effective spin S , lead to an initial susceptibility $\chi_i = \frac{C'_2}{T} + \chi'$, with a concentration of magnetic

pairs of $65 c^2$ instead of $130 c^2$. Only one half of the

previous pairs are magnetic at very low temperatures, the second half being composed of nearly magnetic pairs with characteristic temperatures between .1 and 29 K. A second argument is to consider that $M_2(H, T)$ saturates in ~ 60 kOe for temperatures as different as 50 mK and 1.3 K, while we could expect a diminution of the saturation field with temperature for true magnetic pairs. In fact, this means that we have saturated all the pairs, magnetic and nearly magnetic, whose T_K is lower than 60 kOe.

The existence of nearly magnetic pairs is supported also by resistivity measurements [5] especially sensitive to intermediate behaviors. The analysis of experimental results shows that the T^2 term in very low temperatures resistivity varies versus concentration as $c + c^2$, proving that not only isolated impurities, but also some pairs have a non magnetic behavior at low temperatures. A more detailed analysis allows a computation of the mean T_K of non magnetic pairs, of the order of 6 K, in good agreement with magnetic measurements [2].

2.3 ANALYSIS OF AuCo MAGNETIZATION DATA [6, 7]. — Low temperature measurements : the σ value is found to vary with the cube of the concentration and the high field susceptibility, temperature independent from 0.05 to 10 K is successfully analysed in a c term + a c^2 term (Fig. 4), once the matrix contribution subtracted, for low values of concentration (for higher values, there is a problem because the

FIG. 4. — AuCo alloys. a) χ_F/c vs. c linear plot, proof that χ_F may be written as the sum of a c and a c^2 term. b) Logarithmic plot of σ vs. c showing the c^3 variation of σ .

magnetization curve is not linear in high fields). This suggests strongly that the occurrence of magnetic moments requires the contribution of 3 atoms, while isolated impurities together with pairs have a non magnetic behavior. To go further in the analysis, the magnetic part is assumed to follow a Curie law and the initial susceptibility to be $\chi_i = \chi_0 + \frac{C_3}{T}$, which gives, in a $\chi_i T$ versus T diagram (Fig. 5) a linear variation from which χ_0 and C_3 may be deduced.

FIG. 5. — AuCo. Experimental separation, from low temperature susceptibility measurements, of the magnetic part (characterized by C_3) and of the non magnetic part (characterized by χ_0).

Using as previously σ and C_3 , we deduce an effective spin of $\simeq 4$, concentration independent, and a concentration of magnetic moments of $\simeq 20 c^3$. For several reasons of experimental origin (difficulties in the correction for Fe contamination, difference in the effective spins involved in C_3 and σ), these values must be considered only as an order of magnitude, within perhaps a factor 2 or 3. Nevertheless, an important conclusion can already be drawn: Any model involving 3 atoms to find the right value of $20 c^3$ for the concentration of magnetic moments must not take into account all groups in which one Co atom is not nearest neighbour of at least one Co of the group. In other words, in such a group, environment effects are not sufficient to achieve magnetism. And, as experimental accuracy does not allow a c^3 term in the non magnetic part, we must conclude that only nearest neighbour interactions give significant effects in this alloy. Therefore, we can determine the number N_1 of isolated impurities (having no

impurity in its 12 nearest neighbours) N_2 of impurities in isolated pairs (having no impurity in its 18 nearest neighbours) the rest of impurities being magnetic impurities, with a number N_3 [8].

Within this model, it is possible to study χ_0 versus N_1 and N_2 (Fig. 6). One finds that the susceptibility

FIG. 6. — AuCo. Decomposition of the non magnetic susceptibility in an isolated atoms term $\sim N_1$ and an isolated pairs term $\sim N_2$. This figure is to be compared with figure 4a. It is extended to more concentrated alloys.

of Co in pairs is 8.5 times enhanced with respect to that of isolated impurities (or else that the characteristic temperature T_{K_2} of pairs is 8.5 times lower than that of isolated impurities T_{K_1} . To find an order of magnitude of these temperatures, we plot $\chi_1 - \frac{C_3}{T}$ versus temperature. Though temperature independent at low temperatures, it decreases strongly for higher ones. It is possible to analyze this variation with

$$\chi_1 - \frac{C_3}{T} = \frac{N_1 \mu_1^2}{3k(T + 8.5 T_{K_2})} + \frac{N_2 \mu_2^2}{3k(T + T_{K_2})}$$

giving $T_{K_2} \approx 16$ K and $T_{K_1} \approx 133$ K. Therefore, at low temperatures, pairs have a nearly magnetic behavior, what explains the non linear high field curves (Fig. 7).

This model has been extensively confirmed by nuclear specific heat measurements [9] which are only sensitive to magnetic atoms. As in the case of CuFe, resistivity measurements are the ideal tool to see nearly magnetic impurities. The T term of the low temperature resistivity is found to vary fairly linearly with N_2 (Fig. 8) [10].

FIG. 7. — AuCo. A typical set of magnetization curves, showing the non linear high field behavior for low temperatures.

FIG. 8. — AuCo. The T term of resistivity is fairly proportional to the number of isolated pairs.

2.4 In these two alloys, we observe a superparamagnetic behavior due to pairs in CuFe or triplets in AuCo, which can be taken as the simplest examples of giant moments. In both cases we have intermediate behaviors between the non magnetic single impurity and the magnetic atoms involved in giant moments.

These nearly magnetic atoms are also grouped in pairs, and characterized by a mean critical temperature much lower than that of isolated impurities.

The main difference between the two alloys is the difference of the critical radius r_0 previously defined. For Fe atoms in Cu, this critical radius involves a great variety of possible neighbourhoods giving detectable departures from the single impurity behavior, whereas in AuCo, only nearest neighbours play a significant role. This is a consequence of the difference in characteristic temperatures of single impurities in CuFe (29 K) and AuCo (133 K). As interaction effects are of the same order of magnitude in both alloys, the interaction between two n th neighbour impurities varies the T_K of the same amount. Obviously, a variation of 30 K gives a large effect on CuFe and a nearly zero effect on AuCo.

3. Concentrated alloys. — **3.1 A SIMPLE CRUDE MODEL.** — This survey of two analysis of alloys allows us to improve our basic scheme. There is no abrupt difference between a magnetic and a non magnetic behavior. Any impurity in a given matrix is characterized by a temperature T_K , which depends on its neighbourhood. Experimentally, it is found that its susceptibility may be represented more or less by the law

$$\chi = \frac{C}{T + T_K}.$$

Therefore, in a temperature range much lower than T_K , χ is almost temperature independent, and the impurity has a *non magnetic* behavior. For a temperature range much higher than T_K , χ is almost a Curie law (molecular field effects may then give a positive Curie temperature) and the impurity behaves *magnetically*. In all other cases, when T_K is of the order of the experimental temperature range, we have an intermediate behavior that we call *nearly magnetic* for lack of another term.

An interesting qualitative description of this continuity is to represent the magnetization curves of impurities of a given T_K by a Brillouin function in which $\frac{H}{T}$ is replaced by $\frac{H}{T + T_K}$. It gives the right susceptibility and accounts well for the fact that well below T_K , impurities are saturated in a temperature independent field ($\mu H \approx kT_K$). However, it is well known that magnetic RKKY interactions create a distribution of molecular fields which is not accounted for by this type of Brillouin functions. Moreover, no theoretical model predicts such a Brillouin like behavior for an impurity with a given T_K . So, a quantitative fit of the total magnetization curves by Brillouin functions seems to be questionable, especially when a great number of adjustable parameters is used [11].

We have seen in dilute alloys how a model could almost be deduced from experiment, because of the little number of parameters involved. For more con-

centrated alloys, the quantity of experimental data is nearly constant, while the number of parameters is much greater. Therefore, a model must be chosen *a priori*. In the present state of experiments, it is hopeless to use a sophisticated realistic model of a concentrated alloy, a great variety of such models being possible and equally credible. The model must be simple and schematic, able to account roughly for the main facts observed in the alloys, in order to *understand* well the fundamental mechanisms, even though their detailed balance is not very well *described*.

The fact that the transition occurs in a higher range of concentration means that an impurity needs a richer environment to become magnetic. As is AuCo, r_0 will be reduced to the nearest neighbour distance. Therefore, we assume that the magnetic behavior of an atom, characterized by its T_K , depends only on the number of nearest neighbours of its kind. For some critical number, T_K becomes zero (or much lower than any experimental temperature range). The impurity is then magnetic and is assumed to carry its maximum moment. This model was first used by Jaccarino and Walker, [12] who explained the occurrence of magnetism on Fe atoms in NbMoFe alloys by assuming that the moment of Fe atoms is zero when surrounded by less than 7 Mo atoms, and has its maximum value otherwise. Moreover, in order to make quantitative evaluations, we suppose the alloy to be perfectly disordered. This assumption is reasonable because the effects occur in random alloys. Any chemical short range order, if present, only enhances the effects without being their intrinsic cause. Our purpose is now to show, within these simple ideas and a crude model, that there are no fundamental differences in the origins of the behavior of dilute and concentrated alloys.

3.2 SATURATION MAGNETIZATION MEASUREMENTS.

Probably because of the coexistence of magnetic and nearly magnetic impurities, the asymptotic behavior of the magnetization curves is not very clear. Therefore, the extrapolation of the high field part of the curves to obtain σ must be taken as an order of magnitude of the true magnetic part, especially when the number of magnetic impurities is much smaller than the number of nearly magnetic ones (in the paramagnetic side of the transition). Nevertheless it can be said in a general way that σ varies very quickly with the concentration.

A factor 2.5 is found for CuNi between 43 and 48 % [13], 2.88 for RhNi between 58 and 62 % [14], 6.9 for VFe between 25 and 27 % [15] and 11 for VNi between 86 and 88 % [16]. In all cases also, no evidence is found of the cutoff of σ when the concentration decreases, the experimental sensitivity being the only limitation.

The results on CuNi are well described with the assumption that magnetism occurs on a Ni atom when

it is surrounded by at least 8 nearest neighbours. A similar analysis on RhNi shows that a critical number of 10 nearest neighbours gives the right order of magnitude. For more concentrated alloys, the problem is more intricate because next nearest neighbours play a significant role, but it can be understood in the same way. It must be outlined that this model is schematic, and wants nothing but a quantitative description of the alloys in the range where a magnetic behavior appears, which we call the critical range. But it is basically the same as more sophisticated models [17, 18] which try to describe the magnetic behavior of the alloy at all concentrations above the critical range.

3.3 SUSCEPTIBILITY MEASUREMENTS FAR BELOW THE CRITICAL RANGE. — If we consider concentrations much lower than the critical range, the magnetic part cannot be detected and the only part is a Pauli type curve. As in AuCo, where it was analysed in a c and a c^2 term, the susceptibility can be analysed versus concentration within our crude model, assuming moreover a linear decrease of the characteristic temperature of an impurity with the number n of neighbouring impurities

$$T_K(n) = T_0 - n \Delta T_K.$$

The measurements have been performed on CuNi, showing that the non magnetic susceptibility by impurity increases with concentration (Fig. 9). The analysis [13] finds a critical number of 8 or 9 nearest neighbours for which $T_K(n)$ falls to zero, which is in

rather good agreement with saturation magnetization measurements. Therefore, the alloy may be seen as a mixture of impurities with a nearly zero T_K (magnetic impurities) and of impurities with a distribution of more or less equally spaced T_K (non- and nearly magnetic impurities). In view of the number of different T_K , it is hopeless to determinate them in an analysis versus temperature of the susceptibility. We have now to see what can be deduced of these measurements.

3.4 INITIAL SUSCEPTIBILITY MEASUREMENTS. — For CuNi [19], RhNi [14], VFe [15] and VNi [16], it is not possible to obtain a simple Curie-Weiss behavior of the initial susceptibilities χ_i . Therefore, the analysis is performed in the same way as for AuCo alloys : Low temperature values of χ_i are analyzed in a non magnetic susceptibility χ_0 and a Curie Weiss law :

$$\chi_i = \chi_0 + \frac{C}{T - \theta}.$$

Taking now C and θ as constants, we plot $\chi_i - \frac{C}{T - \theta}$ versus temperature, to see how the *non magnetic* susceptibility varies with temperature.

For CuNi, VNi and VFe, there is no evidence of a variation of χ_0 versus temperature in the temperature range 0-300 K. For RhNi, a significant variation is observed, perhaps in part due to impurities with a T_K situated in the range of measurements. For the three other alloys, the T_K of impurities involved in χ_0 are likely to be higher than 300 K, meaning that ΔT_K is at least of the order of 300 K.

Another interesting fact is that for all 4 alloys, the experimental Curie temperature θ varies from negative to positive values when the concentration increases. We can say that the term contains both magnetic and nearly magnetic (with a low value of T_K) atoms, which cannot be separated clearly. Nearly magnetic atoms follow more or less a Curie Weiss law with a negative Curie temperature $\theta \simeq -T_K$, while magnetic ones follow a Curie Weiss law with a positive θ , characteristic of ferromagnetic molecular field effects. Then, for low values of the concentration, nearly magnetic effects are dominant while for higher concentrations, magnetic effects prevail. It is clear, in this case, that there can be only one environment giving nearly magnetic impurities (7 for CuNi, 9 for RhNi). It can be seen on figure 10 that the ratio of magnetic impurities and of nearly magnetic ones increases strongly in the critical range.

3.5 GIANT MOMENTS. — From comparison between the saturation magnetization and the Curie constants, we can deduce an order of magnitude of the effective magnetic moments and of their concentrations. The result shows *giant moments*, the size of which increases according to the critical range of the alloy ($\sim 10 \mu_B$ for CuNi, $25 \mu_B$ for RhNi and VFe, $40 \mu_B$ for VNi). As already noticed for AuCo alloys, this must be

FIG. 9. — CuNi. a) Reciprocal susceptibilities of non magnetic atoms compared with theoretical curves for $n = 8$ and $n = 10$. b) Mean moment per atom compared with theoretical curves for $n = 7, 8, 9, 10$.

FIG. 10. — Theoretical probabilities of occurrence of nearly magnetic and magnetic sites in RhNi and CuNi, according to the *crude model*.

considered only as an order of magnitude, so that it is difficult to conclude for a given alloy whether these moments vary or not with concentration (they are experimentally found to vary in RhNi and VFe, and to be more or less constant for CuNi and VNi). We must not forget also that in our sense, nearly magnetic atoms are contained in the Curie constant and, to a certain extent, in saturation magnetization values, therefore also in giant moments.

The existence of such giant moments suggests that magnetic atoms are grouped in *clouds* within which they are strongly (probably ferromagnetically) coupled; these clouds are more or less independent in the paramagnetic range and give rise to superparamagnetism. Our crude model is able to account for this distribution [20] using the following argument: As the model is environment dependent, any two sites with overlapping environments have a correlation between their probabilities of being magnetic. This leads to a computation of short range order parameters between moments in a chemically random alloy, giving a good order of magnitude of the size of the mean giant moment.

Neutron diffraction experiments [21] have shown a strong variation of the diffuse elastic cross section with the wave vector $\mathbf{K}$, proof of an inhomogeneous distribution of magnetic moments in the alloy [22].

It is possible to evaluate the neutron experimental curve from magnetic order parameters [23]. The comparison with experiment shows the good order of magnitude, and a discrepancy of a factor 2 in width (Fig. 11). Several explanations may be given for it. Of course chemical clustering may play a significant role, but we think that a more fundamental phenomenon occurs, which must be combined with the *crude model* to obtain a realistic understanding of concentrated alloys.

FIG. 11. — Comparison of the experimental (a) and theoretical (b) diffuse scattering cross sections for CuNi alloys.

4. Polarization effects. — A main thing must not be forgotten in the initial susceptibility in the critical range: The non magnetic part has a susceptibility of the order of that of Pd. The difference is that the enhanced susceptibility is not equally distributed on each site, but has different values according to the environments of atoms. This implies that some atoms are much more enhanced than Pd atoms, and can therefore be strongly polarized or even saturated in the field of neighbouring magnetic impurities. The role

of these polarizations appears more clearly in the study of ternary alloys, made of the alloy under study to which are added some impurities which are known to be magnetic (Fe) and play the role of sources of molecular fields. Systematic studies have been made on different ternary alloys : CuNiFe [24], [25], AuCoFe [9] [26], PdNiFe [27]. On these 3 alloys, the difference between the total magnetization and the part due to Fe atoms is larger than the magnetization of the corresponding iron free alloys.

In CuNiFe alloys, the supplement of magnetization is quantitatively well described in a wide range of concentration (0 to 60 % Ni) by assuming that any non magnetic Ni atom nearest neighbour of a Fe atom carries its maximum moment, while no effect occurs for other Ni atoms (of course, this is valid when Fe concentrations are high enough to assume that all Fe atoms are magnetic).

In AuCoFe alloys, the conjunction of nuclear specific heat and susceptibility measurements provides a greater amount of experimental data. To account correctly for those data, it is necessary to assume a different behavior for isolated and paired Co atoms, together with a distribution of magnetic moments deduced from the distribution of molecular field due to Fe atoms. For the concentrations used, the number of triplets is negligibly smaller than the number of Fe atoms. It is found that the magnetic moment is proportional to the concentration of Fe atoms, proving that the moments on Co atoms are nucleated by the Fe atoms. For that reason, Fe atoms are called active sites, while polarized Co atoms are called passive ones.

A similar phenomenon may be expected in binary alloys, where active atoms, having sufficiently rich an environment, can polarize to a certain extent nearly magnetic neighbours. However, this conceptually simple distinction is not yet available, and perhaps impossible to detect. Moreover, as it involves two super-imposed phenomena (apparition of magnetic moments on active sites by environment effects and polarization of other atoms by those magnetic moments) there are too much adjustable parameters so that a great variety of quantitative models may be proposed to explain equally well the available experimental results. Let us just mention some experimental facts which are better understood with the help of polarization effects.

If one tries the crude model to fit saturation magnetization data for concentrations lower than the critical range for CuNi for instance, we find that a critical number of 9 for low concentrations is more likely than 8 (Fig. 9). This can be understood if we assume that the critical number for an active site is 9 or even 10, and that it polarizes (to saturation) nearly magnetic neighbours, which become more and more numerous with increasing concentration.

Another fact is that the concentration of giant moments varies more or less as the concentration of Ni

atoms having more than 10 nearest neighbours in CuNi and 12 nearest neighbours in RhNi, suggesting [14, 19] that 10 (for CuNi) and 12 (for RhNi) are the critical number of neighbours for active sites. The extent of the polarization clouds nucleated by these sites is not clear, because the analysis of neutron experimental data was made in an oversimplified way [28]. However, the polarization phenomena increase certainly the short range order of magnetic moments, modifying in the right way our theoretical diffuse elastic cross section.

5. Order phenomena. — In disordered alloys, the magnetic order occurs progressively between randomly distributed moments as the temperature is decreased. It is therefore impossible to define accurately an order temperature as in bulk magnetic metals, and several methods are used by experimentalists to determine temperatures below which order phenomena are dominant. In a general way, these temperatures are proportional to the concentration of magnetic moments. In CuFe, they vary as c^2 , proving that the magnetic entities participating to the magnetic order are magnetic pairs of strongly coupled magnetic moments, while well above the order temperature, a superparamagnetic behavior is observed involving both magnetic and nearly magnetic pairs. In AuCo, they vary as c^3 , proving that the order occurs between triplets of magnetic moments. Well above these temperatures, the superparamagnetic behavior involves magnetic triplets and nearly magnetic pairs.

We expect the same phenomenon to occur in more concentrated alloys, the order arising from magnetic interactions between giant moments. Therefore the order temperature should be proportional to the concentration c_m of these giant moments. Unfortunately, accurate determinations of this temperature are difficult [29] and we have already said that the concentration of giant moments is nothing but an order of magnitude, so that no decisive test can be made of this assumption. The only available results [15] are rather promising for its validity.

In a general way the magnetic order is found to be of the ferromagnetic type. The reason lies in the predominance of the nearest neighbour interaction in concentrated alloys. It was justified higher by experimental arguments, but a more fundamental reason for it is the diminution of the mean free path of electrons in concentrated alloys, which shortens the range of the interaction effects.

The meaning of the term *critical concentration* is then very questionable. In VFe, the order temperature decreases with concentration without any evidence of a cutoff for a given concentration, as in CuFe or AuCo, where the experimental temperature range is the only limitation. We think that this phenomenon is general for disordered alloys, and that it is better to speak of a « critical range of concentration ».

Conclusion. — It has been shown that magnetic measurements provide a great amount of experimental data, which are sufficient to build up a model account for all the main features of a magnetic alloy. A great interest of magnetic measurements is a direct interpretation of the measured variables, without using intermediate theories as it is the case for resistivity, neutron or hyperfine measurements. We do not intend to neglect these methods and the original information that they can provide, but insist on the necessity in any study of a magnetic alloy to never forget the basic magnetic measurements.

We have tried also to examine critically the origin of some usual terms, as *giant moments* or *critical concentrations*, and we have shown that they are only a poor though evocative description of reality ; so it is necessary to keep in mind their self contained limitations to avoid giving to these terms an excessive meaning.

We may summarize our present knowledge of a disordered concentrated magnetic alloy by asserting that a concentrated alloy (CuNi) is nothing but a dilute alloy of active magnetic (Ni) atoms embedded in a nearly magnetic (CuNi) host, containing passive sites able to be polarized by active ones. The differences with the classical PdFe alloy are twofold :

a) The nearly magnetic CuNi host is composed of a variety of different atoms with an enhancement of susceptibility strongly varying from an atom to another instead of being constant in a Pd matrix.

Therefore, polarization effects are not so regular as in PdFe, and more difficult to analyse.

b) All the magnetic properties are governed by environment effects, which imply always a non randomness of the site having a given property. Then it can be said that the active sites are never at random, but short range ordered throughout the alloy, even in a perfect chemical disorder.

The short range order due to environment effects is probably observed on nearly magnetic impurities, with a subcritical environment. In part 3-4, we have attributed the Curie-Weiss term with a negative Curie temperature θ to nearly magnetic impurities with a characteristic temperature of $-\theta$. But the Curie constant used leads to Giant moments [14]. The explanation should lie in the presence of nearly magnetic giant moments, with a superparamagnetic behavior above their characteristic temperatures, and non magnetic at low temperatures (not participating to order). These entities are exactly the same as nearly magnetic pairs in CuFe and in AuCo. Resistivity measurements [30], [31], [32] give a good support to the presence of these nearly magnetic giant moments and this point is discussed in detail in a paper of Amamou and Loegel [33] in this conference.

We think that the next step in the understanding of concentrated alloys is a clear study of the nearly magnetic behavior, and of the clustering of nearly magnetic giant moments. This can be done by a systematic study of very low temperature and high field measurements.

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