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LOCAL SPIN SUSCEPTIBILITY IN DISORDERED ALLOYS II (*)

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Résumé. — Nous explorons l'analogie entre une nouvelle formulation du problème de la susceptibilité locale dans les alliages concentrés et la théorie de la localisation des états électroniques dans les systèmes désordonnés. La discussion ouvre la voie à une meilleure description et une meilleure compréhension de l'effet des propriétés magnétiques locales dans les systèmes désordonnés. En particulier, on considère l'effet des interactions entre amas et la nature de la transition de phase magnétique-non magnétique dans les alliages. Finalement, on discute la possibilité dans les alliages concentrés d'une généralisation de la notion de verre de spin.

Abstract. — We explore the analogy between a new formulation of the problem of local susceptibility in concentrated alloys and the theory of localization of electronic states in disordered systems. This discussion opens the way to a better description and understanding of the effect of local magnetic properties in disordered systems. In particular, the effect of cluster-cluster interactions and the nature of the magnetic-non magnetic phase transition in alloys is considered. Finally, we discuss the possibility in concentrated alloys of a cluster generalization of the spin glass phase.

1. **Introduction.** — It is now accepted that the local magnetic properties in transition metal binary substitutional alloys depend strongly on the neighbourhood of each atom. These local environment effects determine the nature of the magnetic-non magnetic phase transition in the critical concentration region and when averaged, determine the macroscopic magnetic properties of the alloy [1-7].

A recent investigation [5-6] of this problem using the methods and results of the theory of disordered alloys has shown that the local magnetic instabilities depend not only on the explicit number of neighbours of both kinds of atoms but also on the variation with environment of the partial densities of states and the local atomic potentials. Fluctuations of environment give rise to fluctuations of these three factors and, therefore, to fluctuations of local magnetic properties, e.g. local susceptibilities and possible local moments. However, the theory of reference [5-6] is not yet able to provide a good description of local environment effects close to the ferromagnetic transition. When nearly magnetic clusters are present in the system, they give rise to non-negligible cluster-cluster interactions and when magnetic clusters are formed, they polarize the non-magnetic clusters. Until now, these effects have been considered in a semi-phenomenological manner [4-6]. In reference [5-6], cluster-cluster interactions were considered indirectly in an approximate way by averaging the medium outside the cluster. This method, however, does not contain an important aspect of cluster-cluster interactions, i.e. the statistical nature of local environment fluctuations in the alloy. In a recent paper [7], one of us has derived an expression for the local susceptibility which allows a statistical approach of local magnetic properties of substitutional alloys. This expression has a formal structure very similar to an expression of the diagonal Green's function used in the discussion of the localization of electrons in disordered systems.

The purpose of this paper is to explore this analogy and to see how the ideas, concepts and conclusions which have been used in the theory of disorder induced localized states could be used to describe magnetic properties of random systems.

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2. **Local spin susceptibility.** — The molecular field theory yields the following simple expression for local susceptibility : the magnetization $\chi_{\alpha\beta}$ of the α-th cell resulting from an external unit field in the β-th cell is obtained from the corresponding non-interacting quantity $\chi^0_{\alpha\beta}$ by the relation

$$\chi_{\alpha\beta} = \chi^0_{\alpha\beta} + \sum_\gamma \chi^0_{\alpha\gamma}\, u_\gamma\, \chi_{\gamma\beta} \,. \tag{1}$$

This equation is the direct consequence of the molecular field approximation : the local magnetic field h_γ acting on conduction electrons is equal to

$$h_\gamma = h^0_\gamma + \sum_\beta u_\gamma\, \chi_{\gamma\beta} \tag{2}$$

where h^0_γ is the value of the external field in the γ-th cell ($h^0_\gamma = \delta_{\gamma\beta}$) and the molecular field in the γ-th cell $\sum_\beta u_\gamma\, \chi_{\gamma\beta}$ is proportional to the local magnetization $\chi_{\gamma\beta}$ and to an effective intra-atomic Coulomb interaction ($u_\gamma = U^{\text{eff}}_\gamma/2\,\mu_B^2$, μ_B being the Bohr magneton). We have shown in reference [7] that a simple and compact form of $\chi_{\alpha\beta}$ can be derived from (1).

If we define $\chi^{(\alpha)}$ as the susceptibility of a medium with an intra-atomic electron-electron interaction on each site except on site α, one can write

$$\chi = \chi^{(\alpha)} + \chi^{(\alpha)} \mid \alpha \rangle\, u_\alpha \langle\, \alpha \mid \chi \tag{3}$$

and, therefore,

$$\chi_{\alpha\beta} = \frac{\chi^{(\alpha)}_{\alpha\beta}}{1 - u_\alpha\, \chi^{(\alpha)}_{\alpha\alpha}} \,. \tag{4}$$

The interacting susceptibilities can be expressed in terms of the non-interacting susceptibilities using the $\mathcal{T}$-matrix :

$$\chi^{(\alpha)} = \chi^0 + \chi^0\, \mathcal{T}^{(\alpha)}\, \chi^0 \,. \tag{5}$$

The matrix element of $\mathcal{T}^{(\alpha)}$ can be expanded in terms of the atomic τ-matrix

$$\mathcal{T}^{(\alpha)}_{\mu\nu} = \tau_{\mu\nu}\, \delta_{\mu\nu} + \tau_\mu\, \chi^0_{\mu\nu}\, \tau_\nu + \sum_{\gamma \neq \mu,\nu}^{(\alpha)} \tau_\mu\, \chi^0_{\mu\gamma}\, \tau_\gamma\, \chi^0_{\gamma\nu}\, \tau_\nu + \cdots \tag{6}$$

with

$$\tau_\mu = \frac{u_\mu}{1 - u_\mu\, \chi^0_{\mu\mu}} \,. \tag{7}$$

The summations in (6) exclude the site α.

From (5) and (6), one can write

$$\chi^{(\alpha)}_{\alpha\beta} = \chi^0_{\alpha\beta} + \sum_{\gamma,\delta} \chi^0_{\alpha\gamma}\, \mathcal{T}^{(\alpha)}_{\gamma\delta}\, \chi^0_{\delta\beta} = \chi^0_{\alpha\beta} + \Sigma^{(\alpha)}_{\alpha\beta} \tag{8}$$

with

$$\Sigma^{(\alpha)}_{\alpha\beta} = \sum_{\gamma \neq \alpha} \chi^0_{\alpha\gamma} \frac{u_\gamma}{1 - u_\gamma\, \chi^0_{\gamma\gamma}}\, \chi^0_{\gamma\beta} + \sum_{\gamma,\delta \neq \alpha} \chi^0_{\alpha\gamma} \times \times \frac{u_\gamma}{1 - u_\gamma\, \chi^0_{\gamma\gamma}}\, \chi^0_{\gamma\delta} \frac{u_\delta}{1 - u_\delta\, \chi^0_{\delta\delta}}\, \chi^0_{\delta\beta} + \cdots \,. \tag{9}$$

Substituting (8) into (4), we obtain the compact form

$$\chi_{\alpha\beta} = \frac{\chi^0_{\alpha\beta} + \Sigma^{(\alpha)}_{\alpha\beta}}{1 - u_\alpha(\chi^0_{\alpha\alpha} + \Sigma^{(\alpha)}_{\alpha\alpha})} \,. \tag{10}$$

The condition for the onset of local instability, i.e. the divergence of the susceptibility, is given by

$$u_\alpha(\chi^0_{\alpha\alpha} + \Sigma^{(\alpha)}_{\alpha\alpha}) = 1 \,. \tag{11}$$

The expansion for $\Sigma^{(\alpha)}_{\alpha\alpha}$ has the same structure as for the self-energy of the site-diagonal Green's function of the one-electron tight binding Hamiltonian

$$\mathcal{H} = \sum_n E_n \mid n \rangle \langle\, n \mid + \sum_{m \neq n} V_{nm} \mid n \rangle \langle\, m \mid \tag{12}$$

in the site representation

$$G_{00} = \langle\, 0 \mid \frac{1}{z - \mathcal{H}} \mid 0 \,\rangle = \frac{1}{z - E_0 - \Sigma_0} \tag{13}$$

with

$$\Sigma_0 = \sum_{n_1 \neq 0} V_{0n_1} \frac{1}{z - E_{n_1}}\, V_{n_1 0} + \sum_{n_1 \neq 0, n_2 \neq 0} \times \times V_{0n_1} \frac{1}{z - E_{n_1}}\, V_{n_1 n_2} \frac{1}{z - E_{n_2}}\, V_{n_2 0} + \cdots \,. \tag{14}$$

This has been used in the discussion of the localization of electrons in disordered systems [8-13]. To reduce the statistical dependence of factors in the series (14), one renormalizes it using Watson's [13] multiple scattering theory so as to obtain in the series only terms corresponding to non-repeating paths. The renormalized series can be written in the form

$$\Sigma_0 = \sum_{n_1 \neq 0} V_{0n_1}\, g^{(0)}_{n_1}\, V_{n_1 0} + \sum_{\substack{n_1 \neq 0 \\ n_2 \neq n_1, 0}} V_{0n_2}\, g^{(0,n_1)}_{n_2} \times \times V_{n_2 n_1}\, g^{(0)}_{n_1}\, V_{n_1 0} + \cdots \tag{15}$$

where

$$g^{(0,n_1,\ldots,n_{i-1})}_{n_i} = (E - E_{n_i} - \Sigma^{(0,n_1,\ldots,n_{i-1})}_{n_i})^{-1} \tag{16}$$

and $\Sigma^{(0,n_1,\ldots,n_{i-1})}_{n_i}$ is the self-energy for a site 0 if in the self-avoiding paths from n_i to n_i the sites

$$n_0, n_1, \ldots, n_{i-1}$$

have been removed (or equivalently, the potentials $E_0, E_{n_1}, \ldots, E_{n_{i-1}}$ have been put $= \infty$).

A similar renormalization can be used for the path-expansion of $\Sigma^{(\alpha)}_{\alpha\alpha}$. If we put

$$u_\nu^{-1} - \chi^0_{\nu\nu} = e_\nu \tag{17}$$

we have

$$\tau_v = (e_v)^{-1} \tag{18}$$

and the renormalized series reads

$$\Sigma_{\alpha\alpha}^{(\alpha)} = \sum_{\beta \neq \alpha} \chi_{\alpha\beta}^0 \tau_\beta^{(\alpha)} \chi_{\beta\alpha}^0 + \sum_{\substack{\beta \neq \alpha \\ \gamma \neq \beta_1 \alpha}} \chi_{\alpha\gamma}^0 \tau_\gamma^{(\alpha,\beta)} \times \times \chi_{\gamma\beta}^0 \tau_\beta^{(\alpha)} \chi_{\beta\alpha}^0 + \cdots \tag{19}$$

where

$$\tau_v^{(\alpha,\beta,\gamma,\ldots,(v-1))} = (e_v - \Sigma_{vv}^{(\alpha,\beta,\gamma,\ldots,(v-1))})^{-1} \tag{20}$$

and

$$\Sigma_{v\gamma}^{(\alpha,\beta,\gamma,\ldots,(v-1))}$$

is the self-correction of the susceptibility for site v if in the self-avoiding closed paths from v to v the sites α, β, γ, ..., $(v-1)$ have been removed (or, equivalently, the susceptibilities χ_{vv}^0 (the electron-electron interactions u_v) have been put $= \infty$ $(= 0)$).

In the alloy the quantities e_v and $\chi_{\mu\nu}^0$ are random quantities; they depend on the kind of atoms occupying sites μ and v and, as was recalled in the introduction, on the neighbourhood of each atom. For a given concentration one can, therefore, define a probability distribution for e_v and $\chi_{\mu\nu}^0$. As most theoretical treatments of localization investigate only the effect of diagonal disorder, we shall assume in the following that e_v is the random variable and that the non-diagonal $\chi_{\mu\nu}^0$ are constant or averaged quantities.

3. **Formulation of the problem.** — The effect of local environment on $\Sigma_{\alpha\alpha}$ and thus on χ_α has been studied using various assumptions. If one considers an isolated cluster formed by a given A or B atom surrounded by a given number of A and B atoms first neighbours, the simplest approximation consists in decoupling this cluster magnetically from the outside i.e. in neglecting in the summation in (19) contributions due to sites outside the cluster. In the framework of this isolated cluster model various approximations have been considered :

a) $\Sigma_{\alpha\alpha}^{(\alpha)}$ has been calculated up to fourth-order paths assuming that the non-interacting susceptibilities χ_{vv}^0 and $\chi_{v\mu}^0$ are environment independent [4]. This formulation has been generalized in order to take account of higher order paths and a closed expression for $\Sigma_{\alpha\alpha}^{(\alpha)}$ within this approximation has been obtained using the multiple scattering theory [7].

b) A more realistic theory has been considered by Brouers *et al.* [5, 6]. The effect of local environment on the diagonal and non-diagonal non-interacting susceptibilities has been incorporated into the theory as well as the dependence of local potentials and charge transfers on local environment.

One can go beyond the isolated cluster approximation by regarding the external medium as an average medium and neglecting the effect of local environment fluctuations outside the cluster. Although the consideration of the medium outside the cluster improves the theory, the molecular field approximation on which the theory is based leads to a fundamental difficulty, namely that the susceptibility of any cluster diverges when the outside medium contains even an infinetisimal concentration of such magnetic clusters.

The purpose of the present paper is to explore how it would be possible to use formula (10) and (19-21) together with some results of the theory of localization to improve the description of local susceptibility in disordered systems by taking account of the statistical nature of cluster-cluster interactions.

4. **Probability of local instability.** — In the theories developed in reference [1-6], one can calculate the condition for the divergence of the susceptibility. Here we shall be concerned with the calculation of the probability for the local susceptibility to diverge and therefore the probability of having a localized moment. If the probability distribution of the χ_{vv}^0 is known, one should be able to calculate the probability distribution of $\Sigma_{\alpha\alpha}^{(\alpha)}$ in the medium outside the cluster and then the probability for the susceptibility to diverge.

The probability distribution of local non-interacting susceptibilities χ_{vv}^0 can be determined starting from the theory of Brouers *et al.* [5]. One calculates the variation of $\chi_{\mu\nu}^0$ with concentration and local environment; starting from these results one can build, for any concentration, the probability distribution of χ_{vv}^0. Once this probability distribution is known, one can use for instance the method of Abou-Chacra *et al.* [14] as discussed in Brouers and Kumar [12] to determine the probability distribution of Σ_{vv}.

The series in (19) is truncated to the first term of the expansion i.e.

$$\Sigma_{\alpha\alpha} = \sum_\beta \frac{(\chi_{\alpha\beta}^0)^2}{(u_\beta^{-1} - \chi_{\beta\beta}^0) - \Sigma_{\beta\beta}^{(\alpha)}}. \tag{21}$$

Such a truncation is only valid for a Bethe lattice. However one assumes that the self-consistent determination of the probability distribution gives a good representation of the true probability distribution.

In the present paper, we want to indicate how such an idea can be used. We do not calculate the parameters $\overline{\chi}_0^0$ and $\Delta_{\chi_0^0}$ and their probability distribution from first principle or from a model hamiltonian but we start from the simplest possible model which can be solved within this approximation. We consider the substitutional alloy $A_x B_{1-x}$. We assume that $u = u_A$, and we use units such that $u = 1$ for A atoms and $u = 0$ for B atoms. This situation could represent for instance NiCu alloys. We consider only the random character of the diagonal non-interacting susceptibility χ_0^0. The non-diagonal susceptibility χ_0^1 is independent of composition. We consider an equiconcentration alloy and we assume that the proba-

bility distribution of χ^0_{vv} is given by a Cauchy distribution

$$P_{\chi^0_0}(\chi^0_0) = \frac{1}{\pi} \frac{\Delta_{\chi^0_0}}{(\chi^0_0 - \bar{\chi}^0_0)^2 + \Delta^2_{\chi^0_0}} . \tag{22}$$

Once the probability distribution of Σ, $P_\Sigma(\Sigma)$, is known by solving the self-consistent equation (21), one can determine the probability for the determinator of $\chi_{\alpha\alpha}$ to be $\leqslant 0$ i.e.

$$P[(\chi^0_{\alpha\alpha} + \Sigma_{\alpha\alpha}) \geqslant 1] . \tag{23}$$

If one starts from (21) and assumes that in the medium $\Sigma_{\alpha\alpha}$ is not different from $\Sigma^{(\beta)}_{\alpha\alpha}$, one can determine the probability distribution of the susceptibility self-correction Σ, following the same arguments as in Brouers and Kumar [12]. One has

$$P_\Sigma(\Sigma) = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \delta\left(\Sigma - \sum_{\beta=1}^{K_A} \frac{(\chi^0_1)^2}{[1 - (\chi^0_0)_\beta] - \Sigma'_\beta}\right) \times \times \prod_\beta P_{\chi^0_0}(\chi^0_0)\, P_{\Sigma'}(\Sigma'_\beta)\, \mathrm{d}(\chi^0_0)_\beta\, \mathrm{d}\Sigma'_\beta . \tag{24}$$

The summation is done over the first neighbours. On average this number is $K_A = xK$ where K is the connectivity constant of the lattice.

Using the Fourier transform of the δ-function, one can write

$$P_\Sigma(\Sigma) = \frac{1}{2\pi} \int_{-\infty}^{+\infty} \mathrm{e}^{it\Sigma} \left(\int_{-\infty}^{+\infty} \mathrm{d}(\chi^0_0) \int_{-\infty}^{+\infty} \mathrm{d}\Sigma' \times \times \exp\left[- \frac{it(\chi^0_1)^2}{(1 - \chi^0_0) - \Sigma'} \right] P_{\chi^0_0}(\chi^0_0)\, P_{\Sigma'}(\Sigma') \right)^{K_A} \mathrm{d}t . \tag{25}$$

Assuming a Lorentzian form for $P_\Sigma(\Sigma)$

$$P_\Sigma(\Sigma) = \frac{1}{\pi} \frac{\Delta_\Sigma}{(\Sigma - \Sigma_0)^2 + \Delta^2_\Sigma} \tag{26}$$

and solving (25) self-consistently, one gets

$$\Sigma_0 + i\Delta_\Sigma = \frac{K_A(\chi^0_1)^2}{(1 - \bar{\chi}_0 - \Sigma_0) - i(\Delta_\Sigma + \Delta_{\chi^0_0})} \tag{27}$$

which gives for Σ_0 a fourth-order equation

$$\Sigma_0(1 - \bar{\chi}^0_0 - \Sigma_0)(1 - \bar{\chi}^0_0 - 2\Sigma_0)^2 + (\Delta_{\chi^0_0})^2 \times \times [1 - \bar{\chi}^0_0 - \Sigma_0]\Sigma_0 = K_A(\chi^0_1)^2 (1 - \bar{\chi}^0_0 - 2\Sigma_0)^2 \tag{28}$$

which can be written as

$$[(1 - \bar{\chi}^0_0)^2 - x]\, x + (\Delta_x)^2 [(1 - \bar{\chi}^0_0)^2 - x] = = 4 K_A(\chi^0_1)^2 x \tag{29}$$

if

$$x = (1 - \bar{\chi}^0_0 - 2\Sigma_0)^2 . \tag{30}$$

The width of the distribution is then given by

$$\Delta_\Sigma = \frac{\Sigma_0 \Delta_{\chi^0_0}}{1 - \bar{\chi}^0_0 - 2\Sigma_0} . \tag{31}$$

a) One can now determine the probability for the local susceptibility of an atom A to diverge. This probability is given by

$$P = \int_1^\infty \frac{1}{\pi} \frac{(\Delta_{\chi^0_0} + \Delta_\Sigma)}{(x - \bar{\chi}^0_0 - \Sigma_0)^2 + (\Delta_{\chi^0_0} + \Delta_\Sigma)^2} \mathrm{d}x \tag{32}$$

$$= \frac{1}{2} - \frac{1}{\pi} \operatorname{arctg} \frac{1 - \bar{\chi}^0_0 - \Sigma_0}{\Delta_{\chi^0_0} + \Delta_\Sigma} . \tag{33}$$

This is the probability for any A-atom to have a local moment. If there are no fluctuations of non-interacting susceptibilities $\Delta_{\chi^0_0}$ and Δ_Σ are equal to zero and therefore $P = 1$ or 0 according to whether $\bar{\chi}^0_0 + \Sigma_0 \geqslant 1$ or < 1. This analysis can be generalized to the case of a cluster of given composition. We consider for instance a cluster having N_A atoms on the shell of first neighbours. In that case, one knows the exact value of $\chi^{0(N)}_0$ on the central atom of the cluster and the local enhanced susceptibility can be written

$$\chi^{(N)}_{\alpha\alpha} = \frac{\chi^{0(N)}_0 + \Sigma^{(N)}}{1 - (\chi^{0(N)}_0 + \Sigma^{(N)})} \tag{34}$$

with

$$\Sigma^{(N)} = \frac{N(\chi^0_1)^2}{1 - \bar{\chi}^0_0 - \Sigma} . \tag{35}$$

The quantity Σ characterizes the medium outside the cluster and its probability distribution is determined by eq. (29-31). Once this probability distribution is known, one can determine the probability distribution of $\Sigma^{(N)}$:

$$P(\Sigma^{(N)}) = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \delta\left(\Sigma^{(N)} - \frac{N(\chi^0_1)^2}{1 - \chi^0_0 - \Sigma}\right) \times \times P_\Sigma(\Sigma)\, P_{\chi^0_0}(\chi^0_0)\, \mathrm{d}\Sigma\, \mathrm{d}\chi^0_0 \tag{36}$$

$$= \frac{1}{\pi} \frac{\Delta_{\Sigma^{(N)}}}{(\Sigma^{(N)} - \Sigma^{(N)}_0)^2 + \Delta^2_{\Sigma^{(N)}}} . \tag{37}$$

which gives the two coupled self-consistent equations :

$$\Delta_{\Sigma^{(N)}} = \frac{(\Delta_\Sigma + \Delta_{\chi^0_0})\, N(\chi^0_1)^2}{(1 - \bar{\chi}^0_0 - \Sigma_0)^2 + (\Delta_\Sigma + \Delta_{\chi^0_0})^2} \tag{38}$$

and

$$\Sigma^{(N)}_0 = \frac{N(\chi^0_1)^2 (1 - \bar{\chi}^0_0 - \Sigma_0)}{(1 - \bar{\chi}^0_0 - \Sigma_0)^2 + (\Delta_\Sigma + \Delta_{\chi^0_0})^2} \tag{39}$$

and the probability of having a divergence of the cluster susceptibility is given by

$$P[(\chi^{0(N)}_0 + \Sigma^{(N)}) > 1] = = \frac{1}{2} - \frac{1}{\pi} \operatorname{arctg} \frac{1 - \chi^{0(N)}_0 - \Sigma^{(N)}_0}{\Delta_{\Sigma^{(N)}}} \tag{40}$$

$\Sigma_0^{(N)}$ (38) and $\Delta_{\Sigma(N)}$ (39) depends on Δ_Σ and Σ_0 which are obtained by solving (28) and (31). We shall apply formula (40) to show how it is possible to account for the influence of magnetic clusters on the susceptibility of a non-magnetic cluster in the alloy's paramagnetic phase.

5. **Illustration of the effect of cluster-cluster interactions.** — Before examining a particular cluster, it is necessary to investigate the relation between the macroscopic condition for ferromagnetism and the local instability condition discussed in the previous sections.

If we consider the average homogeneous system characterized by the susceptibility $\bar{\chi}_0^0$ corresponding to the susceptibility of the cluster with the most probable local environment, there are three ways of defining the instability of the susceptibility. It can be given by :

a) the divergence of the series (9) for this homogeneous system. This yields :

$$1 - \bar{\chi}_0^0 = N\chi_1^0 ; \tag{41}$$

b) the condition for (29) to have a physical solution in the limit of no χ_0^0 fluctuation i.e.

$$1 - \bar{\chi}_0^0 = 2\sqrt{K_N}\,\chi_1^0 ; \tag{42}$$

or, *c*) the condition expressing the divergence of the cluster susceptibility in the same limit which reads

$$1 - \bar{\chi}_0^0 = \frac{N(\chi_0^1)^2}{1 - \bar{\chi}_0^0 - \Sigma_0} . \tag{43}$$

As one can show that (cf. eq. 34 of ref. [12])

$$\Sigma_0 = \frac{N - K_N}{N}(1 - \bar{\chi}_0^0) \tag{44}$$

this condition reads

$$1 - \bar{\chi}_0^0 = \frac{N\chi_1^0}{\sqrt{K_N}} . \tag{45}$$

These three conditions should coincide. The discrepancy comes from the truncation of the series (19) which makes the condition (43) and (45) true only on a Bethe lattice.

As an illustration we consider a cluster corresponding to the most probable configuration

$$(N_A = 3, N_B = 3)$$

of an equiconcentration simple cubic alloy. We assume the distribution of χ_0^0 to be lorentzian of half-width $\Delta_{\chi_0^0}$. We choose for the average diagonal $\bar{\chi}_0^0$, the non-diagonal χ_1^0 and K_A the effective number of A atoms the following values

$$\bar{\chi}_0^0 = 1/2, \quad \chi_1^0 = 1/7 \quad \text{and} \quad K_A = 0.5. \quad K = 2 .$$

They are such that none of the three conditions *a*, *b*, *c* yields an instability i.e. the average medium is paramagnetic and the considered cluster has no local moment if one neglects the fluctuations of χ_0^0. However due to the presence of local moments corresponding to the presence in the alloy of clusters having a local environment such that an instability of the susceptibility occurs, the probability for the considered cluster to have a moment is small but non-zero. This probability can be calculated using equation (40) and depends on the width of the distribution of χ_0^0 and is shown on figure 1.

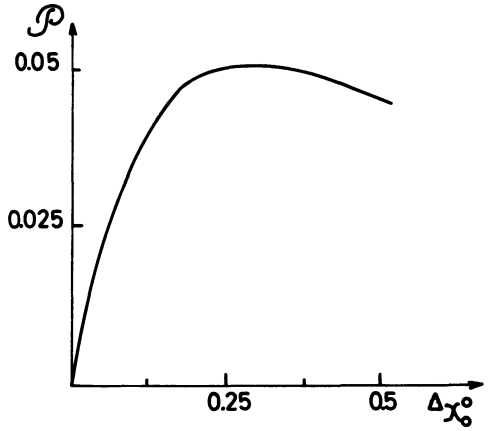

FIG. 1. — Probability of local moment.

The behaviour of the probability which gives a measure of the effective magnetization in the paramagnetic phase of the alloy. Such a small magnetization has been measured by B. Cornut *et al.* [15] in paramagnetic NiCu. In the example we have chosen, the maximum of the probability function for $\Delta_{\chi_0^0} = 0.25$ reflects the competition between the number of magnetic and non-magnetic clusters as $\Delta_{\chi_0^0}$ increases.

In our model where $\chi_0^{0(N)}$ has a sharp value, due to the form of the lorentzian distribution, the probability (40) tends to its non-disorder value (0 or 1) when the width of the susceptibility distribution $\Delta_{\chi_0^0}$ tends to infinity. This gives rise to a maximum or a minimum value of the function *P*.

6. **Relation to the spin-glass problem.** — A few years ago, the spin-glass phase in dilute alloys was discussed [16] from the point of view of the theory of localization starting from the RKKY interaction Hamiltonian. In this section we want to indicate that the concepts of localization could be used also in concentrated transitional alloys starting from the molecular field expression (1). Ideas in the same direction have been also expressed by Sherrington and Mihill [17].

In this last section we restrict ourselves to general speculations. We neglect the long range oscillatory character of χ_0^0 by only considering the fluctuations of the diagonal susceptibility $\chi_{\alpha\alpha}^0$ and averaging the first neighbours susceptibilities $\chi_{\alpha\beta}^0$. Such a restriction

can be tolerated only for concentrated alloys when long range behaviour of susceptibilities is damped and blurred by the disorder.

Starting from the equation which connects the magnetic moment on any site with the moments on neighbouring sites we can write in the molecular field approximation

$$m_\alpha = \sum_\beta \chi^0_{\alpha\beta}(h^0_\beta + u_\beta m_\beta) . \tag{46}$$

Since the external field h^0 is the same on every site, one has

$$\frac{m_\alpha}{h_0} = \chi_\alpha = \sum_\beta \mathcal{A}_{\alpha\beta}\, \chi^0_\beta \tag{47}$$

where the matrix elements of $\mathcal{A}$ are given by

$$\mathcal{A}_{\alpha\beta} = \frac{\text{cofactor } D_{\alpha\beta}}{\det D} \tag{48}$$

and

$$D = [I - \chi^0 u] \tag{49}$$

$\mathcal{A}$ is a random-matrix. The nature of the spectrum of eigenvalues determines the nature of the solution of (47). From (47), we can write

$$\chi = \sum_\alpha \chi_\alpha = \sum_{\alpha,\beta} \frac{\mathrm{d}}{\mathrm{d}D_{\alpha\beta}} \ln \det D_{\alpha\beta}\, \chi^0_\beta \tag{50}$$

or

$$\chi/\chi^0 = -\,2 \sum_{\alpha,\beta} \frac{\dfrac{\mathrm{d}}{\mathrm{d}D_{\alpha\beta}} (\det D)^{-1/2}}{(\det D)^{-1/2}} . \tag{51}$$

Using the integral representation

$$(\det D)^{-1/2} = \frac{1}{\pi^{N/2}} \int \mathrm{d}x_1 \cdots \mathrm{d}x_N\, \mathrm{e}^{-\sum_{\alpha\beta} D_{\alpha\beta}\bar{x}_\alpha x_\beta} \tag{52}$$

one gets after an orthogonal transformation

$$\frac{\mathrm{d}}{\mathrm{d}D_{\alpha\beta}} (\det D)^{-1/2} = -\,\frac{1}{\pi^{N/2}} \times \times \int \mathrm{d}y_1, \cdots, \mathrm{d}y_N \left(\sum_\alpha y^2_\alpha\right) \mathrm{e}^{-\sum_\gamma \lambda_\gamma y^2_\gamma} \tag{53}$$

and

$$\chi/\chi^0 = 2 \sum_\alpha \frac{\displaystyle\int \mathrm{d}y_\alpha\, y^2_\alpha\, \mathrm{e}^{-\lambda_\alpha y^2_\alpha}}{\displaystyle\int \mathrm{d}y_\alpha\, \mathrm{e}^{-\lambda_\alpha y^2_\alpha}} \tag{54}$$

and finally

$$\chi/\chi^0 = \sum_\alpha \frac{1}{\lambda_\alpha} = \int \frac{1}{\lambda} g(\lambda)\, \mathrm{d} \tag{55}$$

where λ are the eigenvalues and $g(\lambda)$ the density function of the spectrum of the random matrix

$$D = I - \chi^0 u . \tag{56}$$

This matrix has a structure similar to the Anderson model of cellular disorder with diagonal and non-diagonal (between nearest neighbours) terms. In the NiCu type alloy considered in the first part of this paper, the matrix D would be written for a one-dimensional system as

$$\begin{pmatrix} 1 - u\chi^0_0\,\zeta_1 & u\chi^1_0\,\zeta_1\,\zeta_2 & 0 & 0 & 0 & \dots \\ u\chi^1_0\,\zeta_2\,\zeta_1 & 1 - u\chi^0_0\,\zeta_2 & u\chi^1_0\,\zeta_2\,\zeta_3 & 0 & 0 & \dots \\ 0 & u\chi^0_1\,\zeta_3\,\zeta_2 & 1 - u\chi^0_0\,\zeta_3 & u\chi^1_0\,\zeta_3\,\zeta_4 & 0 & \dots \\ \vdots & \vdots & \vdots & \vdots & \vdots & \vdots \end{pmatrix} \tag{57}$$

where ζ_i is equal to 1 or 0 according to whether site i is occupied or not by a Ni atom.

One can, as a first approximation replace the non-diagonal term by an average and represent the diagonal term by a distribution of width Δ. In this example as well as in more complicated situations where the diagonal disorder can be represented by a random distribution, the results of the theory of localization [8-12] can be invoked to make some general statements. According to the relative value of Δ which in alloys depends on concentration, the spectrum of D is composed of extended states, a mixture of localized and extended states separated by a mobility edge or a set of localized states.

From (55), one can see, that the alloy is paramagnetic when the eigenvalues of D are all positive while the magnetic solutions occur for negative eigenvalues. In this case two possible situations can occur.

i) The local environment fluctuations can be such that the negative eigenvalues correspond to localized solutions (cf. Fig. 2).

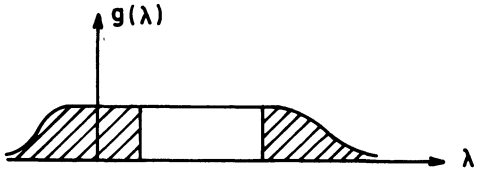

FIG. 2. — Density function of the eigenvalue spectrum of D. Hachured regions correspond to localized states.

Then one has local moment but no long-range ordering and there exists the possibility of a cluster generalization of the spin glass phase.

ii) If the concentration of atoms bearing local moments increases, percolation paths corresponding to magnetic clusters with sufficiently similar neighbourhood will appear, some negative eigenvalues will correspond to extended states and the alloy will undergo a ferromagnetic transition.

7. **Conclusions.** — Any quantitative theory of the magnetic-non magnetic transition in concentrated alloys would require a more realistic description of the local environment fluctuations and of the cluster-cluster interaction. However we believe that the consideration developped in the present paper could be a useful starting point for further investigations in this field.

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