Spin-glass-like behavior of dilute Cr-Er and Cr-Yb alloys

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Magnetic susceptibility measurements of dilute Cr-Er and Cr-Yb alloys are reported. A peculiar magnetic behavior is observed below the Néel temperature. A model is proposed in order to explain this behavior. The random $3d$-$4f$ interaction is shown to be responsible for the formation of a peculiar magnetic structure which has common features with a spin-glass. Critical temperatures and magnetic properties are derived.

I. INTRODUCTION

The interest in chromium itinerant antiferromagnetism is well known. Extensive studies of the influence of alloying on the various properties of the spin-density wave (SDW), such as the Néel temperature, spin-flip temperature, and commensurability, have been carried out. Among these studies, some are concerned with the reciprocal influence of magnetic impurities and itinerant antiferromagnetic electrons. This problem is interesting because of its relation with the one of s-d interaction in the situation when conduction electrons form an ordered state.

$3d$ magnetic impurities have quite complicated effects, especially because the magnetic moment of the impurity is very sensitive to the interaction with the host. Therefore, the d-f interaction of the SDW with a well-defined $4f$ magnetic moment might reveal some features which are helpful toward a better understanding of the coexistence between itinerant antiferromagnetism and localized paramagnetism.

The aim of this paper is to discuss the interaction between the $3d$ spin-density wave of chromium and the $4f$ localized magnetic moments of Er and Yb. Our attempt, on both the theoretical and experimental side, showed that such an interaction may result in the formation of a peculiar magnetic structure, called "spin-glass-like structure" in this paper.

The next section is devoted to the experimental results which shed light on the model and the discussions are presented in the following sections.

II. SAMPLE PREPARATION AND EXPERIMENTAL RESULTS

As indicated in Ref. 1, Er and Yb have a very low solubility in chromium. To obtain homogeneous samples thus becomes a difficult task.

The samples have been prepared by arc melting, under pure-argon atmosphere, 99.999% pure chromium and 99.9% pure Er and Yb. In order to obtain homogeneous samples we remelted them several times, annealed them at 1200 K for 12 h, and finally quenched them to room temperature. Homogeneity has been checked by comparing the room-temperature magnetic susceptibility of serveral parts of an ingot, the error for the magnetic susceptibility of the parts of a homogeneous ingot being 5%.

Thermal hysteresis was searched for in order to determine the solubility limit. For Cr-Yb we have found this limit to lie between 0.112 and 0.225 at.% Yb. The magnetic properties of the last sample (0.225 at.% Yb) differed from those of the samples with lower Yb concentration. For Cr-Er alloys this limit could not be defined accurately enough.

For concentration control we have used a neutron activation method.[2] For each system, three concentrations were thus determined. The other ones have been obtained by comparing their high-temperature magnetic susceptibilities.

Magnetic susceptibility measurements have been carried out, with Weiss-Forrer equipment, between 80 and 800 K. The experimental error lies within 3%.

The temperature dependence of the magnetic susceptibility for the investigated systems, Cr-Er and Cr-Yb, is given in Figs. 1 and 2.

Figures 1 and 2 show also the magnetic susceptibility of the pure chromium we used in sample preparation. The critical Néel temperature is easily recognized and lies around 312 K. For $T > T_N$ the magnetic susceptibility has a quadratic temperature dependence, specific to the Pauli paramagnetism.

The magnetic susceptibility of the alloys also shows some critical temperature. The zero concentration limit of this temperature is identical to the Néel temperature of pure chromium. Therefore, we shall consider this temperature as the Néel temperature of the alloy.

The temperature dependence of the magnetic susceptibility of an alloy is quite different from that of pure chromium. Above the Néel temperature, a Curie-Weiss dependence is present, showing the ex-

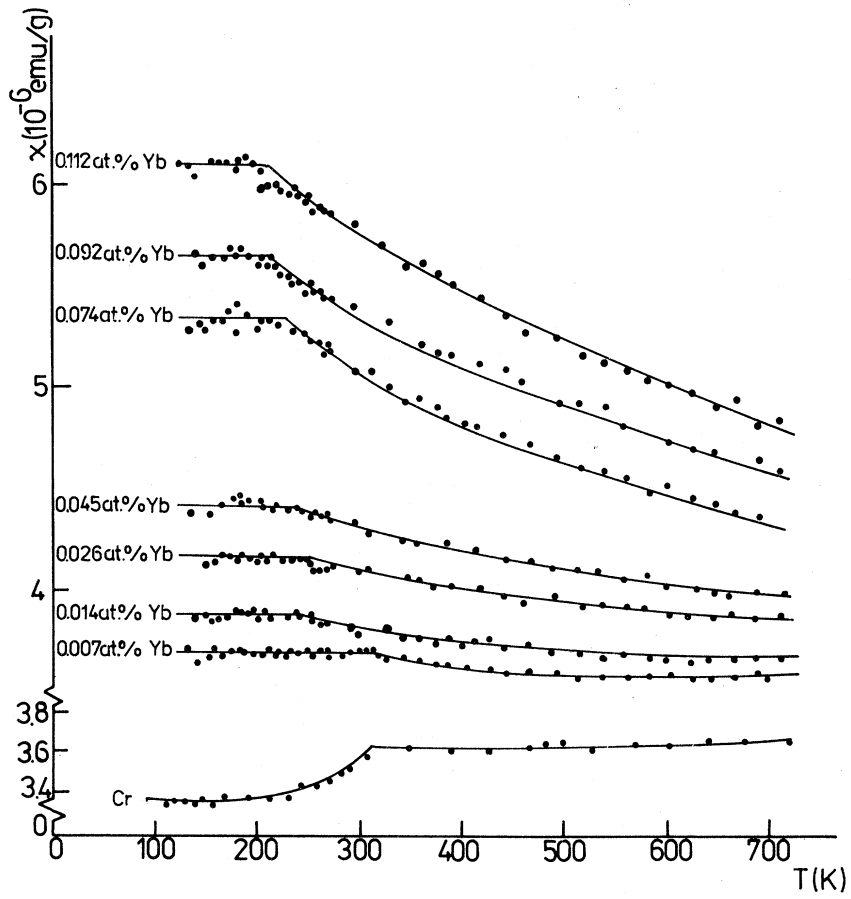

FIG. 1. Temperature dependence of the magnetic susceptibility for Cr-Yb alloys at 10 kG. The full lines are given by Eq. (18). The temperature dependence of the magnetic susceptibility of the chromium used for the preparation of the samples is also plotted.

istence of localized magnetic moments. Below this temperature, the behavior of the magnetic susceptibility shows no common features either with Curie-Weiss-like paramagnetism or with itinerant antiferromagnetism. The existence of a Néel temperature and of the localized magnetic moments indicates that this peculiar magnetic behavior is the result of the interaction between the $4f$ localized magnetic moments of Er or Yb and the SDW specific for itinerant antiferromagnetism.

In the following sections we suggest a model which tries to explain the observed magnetic behavior of dilute Cr-Er and Cr-Yb alloys.

The most important experimental fact, observed for this alloy, is the suppression of the normal paramagnetic behavior of the impurity spins below the Néel temperature of the conduction-electron system. Taking into account the interaction between the randomly substituted $4f$ spins and an incommensurate SDW, one might imagine the $4f$ spins feeling a random effective field proportional to the local value of the SDW amplitude. If strong enough, this field might freeze in the localized magnetic moments along randomly distributed directions. As a consequence the local mean values of the $4f$ spins will be nonzero ($\langle S_f \rangle \neq 0$) but, due to the randomness of the system, the total spin of any part of the $4f$ spin system is zero. This means that there are no magnetic (ferri- or antiferro-) sublattices although the local mean values of the impurity spin are nonzero. This feature brings the system of localized magnetic moments closer to the spin-glass state than to any antiferromagnetic state. We shall call this behavior "spin-glass-like."

Thinking of a spin-glass state induced by a random Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, one might find that although the spins see random fields, as they do in our model, their origin is quite different. The most striking difference occurs because the random interaction we discuss, is not a spin-spin one but a spin-SDW one.

Using "mean-field" approximations, the following

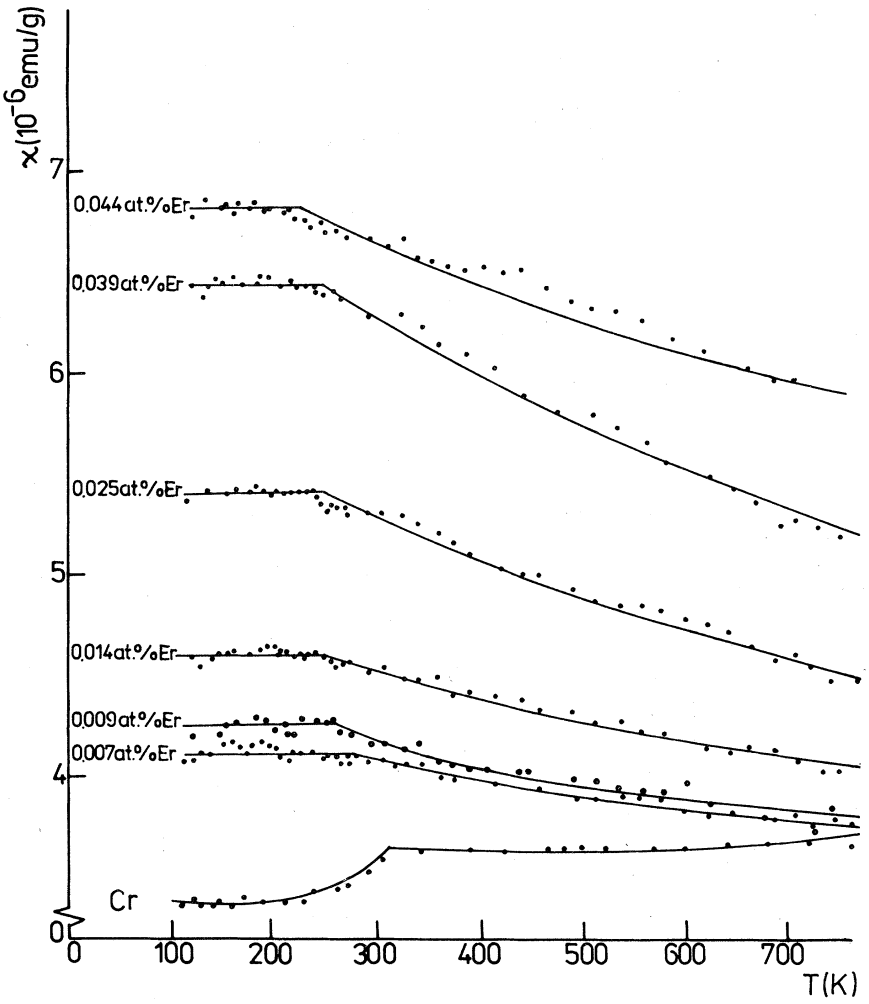

FIG. 2. Temperature dependence of the magnetic susceptibility for Cr-Er alloys at 10 kG. The full lines are given by Eq. (18). The temperature dependence of the magnetic susceptibility of the chromium used for the preparation of the samples is also plotted.

sections describe the magnetic behavior of the investigated systems considered as a "spin-glass-like" structure.

III. HAMILTONIAN

In order to discuss the random spin-SDW interaction let us consider a disordered alloy with chaotically distributed impurity spins, S_f, interacting with d-like electrons, in the presence of the electron-hole coupling.

The Hamiltonian of our problem reads

$$\mathcal{H}=\mathcal{H}_C+\mathcal{H}_L+\mathcal{H}_{LC} \ . \tag{1}$$

The two-band Hamiltonian for conduction electrons is

$$\mathcal{H}_C=\sum_{K,\alpha}[\epsilon_1(K)-\mu^*]a^\dagger_{K,\alpha}a_{K,\alpha}+\sum_{K,\alpha}[\epsilon_2(K)-\mu^*]b^\dagger_{K+Q,\alpha}b_{K+Q,\alpha}+\frac{V}{2}\sum_{\substack{K,K'\\ \alpha,\beta,\gamma,\delta}}\vec{\sigma}_{\alpha\beta}\vec{\sigma}_{\gamma\delta}a^\dagger_{K'-Q,\alpha}b^\dagger_{K+Q,\gamma}b_{K'\beta}a_{K,\delta}+\text{c.c.} \ , \tag{2}$$

where $a^\dagger_{K,\alpha}$ and $b^\dagger_{K,\alpha}$ are the creation operators for the two bands (electrons and holes). The chemical potential μ^* is affected by alloying, while for pure chromium the two bands considered satisfy

$$\tilde{\epsilon}(K)=\epsilon_1(K)-\mu=-[\epsilon_2(K)-\mu] \ .$$

The last term in Eq. (2) describes a tripletlike interband interaction. Intraband and singletlike interactions were neglected. As shown in Ref. 3, the experimental results concerning pure-chromium magnetic susceptibility[4] possibly indicate the coexistence of a singlet coupling in pure chromium, which however, becomes important only at low temperatures.

For the localized spin Hamiltonian we choose

$$\mathcal{H}_L = -g\mu_B \vec{H} \sum_f \vec{S}_f - \frac{1}{2} \sum_{f_1 f_2} B(R_{f_1} - R_{f_2}) \vec{S}_{f_1} \vec{S}_{f_2} \quad . \tag{3}$$

The second term shows a possible RKKY coupling between the impurity spins $\vec{S}_f$. We shall consider $B(R) > 0$ and only weakly R dependent in order to discuss the competition between a homogeneous ferromagnetic coupling of impurity spins (which might occur if the critical impurity concentration for ferromagnetism is surpassed) and the random interaction with the SDW.

The interaction Hamiltonian is written as

$$\mathcal{H}_{LC} = \sum_{\substack{K,K' \\ \alpha,\beta,i,j}} \sum_f (\langle K,i|U|K',j\rangle + \langle K,i|J|K',j\rangle \vec{S}_f \vec{\sigma}_{\alpha\beta}) e^{-i(K-K')R_f} a^{i\dagger}_{K,\alpha} a^j_{K',\beta} \quad , \tag{4}$$

where the following notations were used: $i,j \in \{1,2\}$ are band indices; $a^1_{K,\alpha} \equiv a_{K,\alpha}$ and $a^2_{K,\alpha} \equiv b_{K,\alpha}$; U and J describe the potential and spin scattering. In Eqs. (2) – (4) K, Q, and R_f are vectors.

To solve Eq. (1) we replace H by two effective Hamiltonians for the systems of conduction electrons and localized spins. To obtain the effective Hamiltonians we shall use a local self-consistent mean-field method, rather than that of unitary transformations. Thus, the Hamiltonian (1) is equivalent to

$$\begin{aligned} \mathcal{H}_{\text{eff}C} &= \mathcal{H}_C + \langle \mathcal{H}_{LC} \rangle_L \quad , \\ \mathcal{H}_{\text{eff}L} &= \mathcal{H}_L + \langle \mathcal{H}_{LC} \rangle_C \quad , \end{aligned} \tag{5}$$

where $\langle\ \rangle_L$ and $\langle\ \rangle_C$ are partial mean values obtained by mediating localized spin operators and, respectively, conduction-electron operators. In first order, mean values are computed with unperturbed Hamiltonians $\mathcal{H}_L$ and $\mathcal{H}_C$. Higher order is obtained by iterating this process.

We consider now the case of a $\frac{1}{2}$ impurity spin. Taking the external magnetic field along the Oz axis, we may consider[5] $Q \| Oz$, and choose the σz representation for the conduction-electron spin operators. In order to obtain Eq. (5) we shall stop the iteration at the first significant order for each coupling constant.

In first order we get

$$\langle \mathcal{H}_{LC} \rangle_C = -\frac{1}{2} \sum_i J_{ii}(0) \sum_K (n^i_{K,\uparrow} - n^i_{K,\downarrow}) \sum_f S^z_f - J_{ab}(Q) \sum_f \frac{\Delta}{V} S^z_f \cos Q R_f \quad , \tag{6}$$

where $n^i_{K,\alpha} = \langle a^{i\dagger}_{K,\alpha} a^i_{K,\alpha} \rangle$ and $\Delta/V = \sum_{K,\alpha,\beta} \sigma^z_{\alpha\beta} \langle a^\dagger_{K,\alpha} b_{K+Q,\beta} \rangle$. The next iteration would produce a correction to the RKKY coupling, considered in Eq. (3), which we neglect.

The second term of Eq. (6) shows an interaction between localized spins and the SDW, discussed previously in Ref. 6.

To obtain $\langle \mathcal{H}_{LC} \rangle_L$ we go to second order in J, while for U the first order will be significant because of the special band structure considered. Thus the repeated scattering on impurity potential is neglected. For the high-temperature limit, the linearized local response of the spin is

$$\langle S^z_f \rangle = \frac{\mu_B H}{4kT} + \frac{\langle S^z \rangle B_0}{16kT} + \frac{1}{8} \langle K,i|J|K',j\rangle e^{-i(K-K')R_f} \sigma^z_{\alpha\alpha} \frac{\langle a^{i\dagger}_{K,\alpha} a^j_{K',\alpha} \rangle}{2kT} \quad ,$$

where $\langle S^z \rangle = 1/\mathfrak{N} \sum_f \langle S^z_f \rangle$, $\mathfrak{N}$ being the impurity concentration and $B_0 = \sum_f B(R_f)$. For the second-order term in J we obtain

$$-\frac{1}{16} \sum_{\substack{K_1,K_2,K,K' \\ l,m,i,j,\alpha,\beta}} \frac{1}{2kT} \langle K_1,l|J|K_2,m\rangle \langle K,i|J|K',j\rangle e^{-i(K-K'+K_1-K_2)R_f} \sigma^z_{\alpha\alpha} \sigma^z_{\beta\beta} \langle a^{i\dagger}_{K,\alpha} a^j_{K',\alpha} \rangle a^{l\dagger}_{K_1,\beta} a^m_{K_2,\beta} \quad ,$$

where $l,m,i,j \in \{1,2\}$ are band indices and k is the Boltzmann constant. The second-order term in J is the "mean-field" equivalent of the Kim Hamiltonian.[7] Differences occur because of the two-band system and the σ^z symmetry considered.

Performing an impurity mediation we get finally

$$\langle \mathcal{H}_{LC} \rangle_L = \sum_{K,\alpha} (\mathfrak{N} U_a - H_a \sigma^z_{\alpha\alpha}) a^\dagger_{K,\alpha} a_{K,\alpha} + \sum_{K,\alpha} (\mathfrak{N} U_b - H_b \sigma^z_{\alpha\alpha}) b^\dagger_{K+Q,\alpha} b_{K+Q,\alpha}$$
$$- \frac{\mathfrak{N}}{4} \frac{S(S+1)}{3kT} |J_{ab}(Q)|^2 \sum_{K,\alpha} \frac{\Delta}{V} \sigma^z_{\alpha\alpha} b^\dagger_{K+Q,\alpha} a_{K,\alpha} - \text{c.c.} \ , \tag{7}$$

where $U_a = \langle K,1|U|K,1\rangle$; $U_b = \langle K,2|U|K,2\rangle$; $H_a = \mu_B H + J_a(\mathfrak{N}/2)\langle S^z\rangle$; $H_b = \mu_B H + J_b(\mathfrak{N}/2)\langle S^z\rangle$; $J_a = \langle K,1|F|K,1\rangle$; $J_b = \langle K,2|J|K,2\rangle$; $J_{ab}(Q) = \langle K,1|J|K+Q,2\rangle$. The presence of the temperature as a coefficient in the Hamiltonian is a characteristic feature of the mean-field method we used.[8]

If $U_a = U_b$, and that happens if the SDW is commensurable, the first significant term is the second-order one. We shall not consider this case, since the effect of the second-order term has been discussed several times (see for instance Ref. 9).

The effect of the molecular fields H_a and H_b has been studied previously.[10] However, the second-order term is important, since in the paramagnetic state $\langle S^z\rangle$ is proportional to the external magnetic fields.

The conduction-electron Hamiltonian, thus obtained, is not spatially homogeneous but, up to terms in J^2, the off-diagonal terms are nonzero only if $K - K' = Q$.

IV. MAGNETIC PROPERTIES OF THE CONDUCTION-ELECTRON SYSTEM

To keep the same mean-field approximation we shall rewrite the electron-hole coupling term as in Ref. 3, setting $H = 0$:

$$\mathcal{H}_{\text{eff}C} = \sum_{K,\alpha,i} \tilde{\epsilon}_i(K) a^\dagger_{K,\alpha} a^i_{K,\alpha} - \tilde{\Delta} \sum_{K,\alpha,\beta} \sigma^z_{\alpha\beta} b^\dagger_{K+Q,\alpha} a_{K,\beta} - \text{c.c.} \ , \tag{8}$$

where $\tilde{\epsilon}_1(K) = \tilde{\epsilon}(K) - \delta_\mu + U_a\mathfrak{N}$; $\tilde{\epsilon}_2(K) = -\tilde{\epsilon}(K) - \delta_\mu + U_b\mathfrak{N}$; $\delta_\mu = \mu^* - \mu$; and $\tilde{\Delta} = \Delta[1 + (\mathfrak{N}/4V)\,|J_{ab}(Q)|^2 S(S+1)/3kT]$. Denoting $\xi = \tilde{\epsilon} + \mathfrak{N}(U_a - U_b)/2$ we may write the gap equation as

$$\Delta = V \sum_{K,\alpha} \sigma^z_{\alpha\alpha} \frac{-\tilde{\Delta}\sigma^z_{\alpha\alpha}}{2(\xi^2 + \tilde{\Delta}^2)^{1/2}} \{ f[\tilde{\delta}_\mu + (\xi^2 + \tilde{\Delta}^2)^{1/2}] + f[\tilde{\delta}_\mu - (\xi^2 + \tilde{\Delta}^2)^{1/2}] \} \ ,$$

where $\tilde{\delta}_\mu = \delta_\mu - \mathfrak{N}(U_a + U_b)/2$ and where f is the Fermi function.

For the Néel temperature it follows:

$$\ln \frac{T_N}{T_{N_0}} = \frac{1}{V\rho(\mu)} \frac{\rho(\mu - \mathfrak{N}(U_a - U_b)/2) - \rho(\mu)}{\rho(\mu)} + \frac{1}{V\rho(\mu)} \frac{\mathfrak{N}|J_{ab}(Q)|^2 S(S+1)}{8V 3kT_N} - \frac{\delta_\mu^2}{8k^2 T_N^2} \ . \tag{9}$$

In Eq. (9) only the first significant order in J and δ_μ was considered. δ_μ might be evaluated by considering the change in the particle number with alloying.

Within the rigid-band model we get $\delta_\mu = \mathfrak{N} z/4\rho(\mu)$ where z is the change in the number of $3d$ electrons per impurity atom.

Relation (9) shows that the effect of the interaction with the randomly distributed impurity spins is to augment the Néel temperature. This effect occurs if the ferromagnetic correlation between localized spins is absent. As shown in Ref. 10, ferromagnetic correlations, or to be precise, a ferromagnetic molecular field, have the tendency to destroy the electron-hole coupling.

It must be also underlined that the whole treatment breaks down for $T \to 0$ because the linear response of the impurity considered in order to obtain Eq. (8), is no more appropriate.

To obtain the magnetic susceptibility we may use standard results[11] by replacing the renormalized gap

$$\chi(T) = \chi_P \left[\frac{2}{3} + \frac{1}{3} \int_0^D d\left(\frac{\epsilon}{2kT} \right) \Big/ ch^2 \left(\frac{\epsilon^2 + \tilde{\Delta}^2}{4k^2T^2} \right)^{1/2} \right] \ .$$

To obtain the Pauli susceptibility χ_P we consider $T > T_N$ and use instead of approximation (8) the following decoupling in the equation of motion for the Green's function

$$[\omega - \tilde{\epsilon}_1(K)] \langle\langle a_{K,\alpha} a^\dagger_{K,\alpha} \rangle\rangle_\omega = \frac{i}{2\pi} + \frac{V}{2} \langle b^\dagger_{K+Q,\alpha} b_{K+Q,\alpha} \rangle \langle\langle a_{K,\alpha} a^\dagger_{K,\alpha} \rangle\rangle_\omega \ .$$

Using a similar equation for $\langle\langle b_{K+Q,\alpha} b^\dagger_{K+Q,\alpha} \rangle\rangle$ one gets

$$\langle a^{\dagger}_{K,\uparrow}a_{K,\uparrow}\rangle - \langle a^{\dagger}_{K,\downarrow}a_{K,\downarrow}\rangle = -2f'(\xi-\delta\mu)\frac{H_a+\frac{1}{2}VH_b f'(\xi+\delta\mu)}{1-\frac{1}{4}V^2 f'(\xi-\delta\mu)f'(\xi+\delta\mu)} \ .$$

Neglecting the enhancement, specific to the second order, and using the similar formula for $\langle b^{\dagger}_{K+Q,\uparrow}b_{K+Q,\uparrow}\rangle - \langle b^{\dagger}_{K+Q,\downarrow}b_{K+Q,\downarrow}\rangle$ we finally get for $T \gg T_N$ but $T \ll T_F$ (the Fermi temperature):

$$\chi_P = 4\mu_B^2\rho\left[\mu - \mathfrak{N}\frac{U_a-U_b}{2}\right]\left[1-\frac{\chi_L}{\mu_B^2}\frac{J_a+J_b}{4}-\frac{V}{4kT}\right] \ , \tag{10}$$

where χ_L is the magnetic susceptibility of the localized spin system and μ_B the Bohr magneton. The last two terms in Eq. (10) show the influence of the interaction with the impurity and of the interband interaction. Comparing the most significant terms of Eqs. (9) and (10) we get a useful relation

$$\ln\frac{T_N}{T_{N_0}} \simeq \frac{1}{V\rho(\mu)}\frac{\chi_P-\chi_P^0}{\chi_P^0} \ , \tag{11}$$

where χ_P^0 is the Pauli susceptibility of pure chromium.

V. MAGNETIC PROPERTIES OF THE LOCALIZED-SPIN SYSTEM

Using Eqs. (6) and (10) we may write the effective Hamiltonian for the impurity spins as

$$\mathcal{H}_{\text{eff}_L} = -\left[g+\frac{\chi_P}{2\mu_B^2}(J_a+J_b)\left(1-\frac{V}{4kT}\right)\right]\mu_B H\sum_f S_f^z - \frac{1}{2}\mathfrak{N}\langle S^z\rangle\left[\rho(\mu)(J_a^2+J_b^2)+\frac{B_0}{\mathfrak{N}}\right]\sum_f S_f^z$$

$$-J_{ab}(Q)\frac{\Delta}{V}\sum_f S_f^z\cos(QR_f) \ . \tag{12}$$

In Eq. (12) we observe the Zeeman term with the expected g shift, a "molecular-field" term due to conduction-electron polarization and to homogeneous spin-spin interaction, and a last term, impurity-spin–SDW interaction, which destroys the homogeneity of the system.

However, for $T > T_N$, the last term disappears and one can easily show that the magnetic susceptibility of the system of localized magnetic moments has the Curie-Weiss form

$$\chi_L = \frac{C}{T-\theta} \ , \tag{13}$$

where

$$C = \frac{\mathfrak{N}}{3k}\mu_B^2 g^2[1+\rho(\mu)(J_a+J_b)]S(S+1)$$

and

$$\theta = \frac{B_0}{4k}+\mathfrak{N}\rho(\mu)\frac{J_a^2+J_b^2}{8k}-\rho(\mu)\frac{(J_a+J_b)V}{2k} \ .$$

The first term of θ describes the spin-spin interaction considered in Eq. (3), the second one might be ascribed to an effective spin-spin coupling via intraband polarization of electrons and holes, and the last term is related to a similar effective coupling via interband polarization.

For $J_a \simeq J_b$ we find a useful connection between θ and the effective magnetic moment per impurity atom

$$\mu' = g\mu_B[1+2J\rho(\mu)]\sqrt{S(S+1)}-\theta \ ,$$

$$-\theta = \frac{V}{2k}\frac{\mu'-\mu_0'}{\mu_0'}-\frac{B_0}{4k}-\frac{\mathfrak{N}J}{8k}\left(\frac{\mu'-\mu_0'}{\mu_0'}\right)^2 \ , \tag{14}$$

where μ_0' is the magnetic moment of the free impurity atom.

To solve the problem for $T < T_N$ we shall use some methods utilized for spin-glasses.[12] To be precise, we shall use the idea of replacing the effect of the nonhomogeneous molecular field $\tilde{H}(R_f) \equiv J_{ab}(Q)(\Delta/V)\cos(QR_f)$, which depends on the random variable R_f, by the action of a local, fluctuating field $\tilde{H}$, acting on each impurity spin but being spatially homogeneous.

For an incommensurable SDW one can easily show that the distribution function of such a field is

$$P(\tilde{H}) = \frac{\theta(H_0-\tilde{H})}{\pi(H_0^2-\tilde{H}^2)^{1/2}} \ ,$$

where $H_0 = J_{ab}(Q)\Delta/V$.

The magnetization is defined as[12]

$$M(H) = \int_{-\infty}^{+\infty} M(H,\tilde{H})P(\tilde{H})\,d\tilde{H} \ .$$

Thus, the magnetization becomes

$$M(A)=\mathfrak{N}\mu_B\frac{1}{\pi}\int_{-H_0}^{H_0}\frac{d\tilde{H}}{(H_0^2-\tilde{H}^2)^{1/2}}\tanh\frac{A+\tilde{H}}{2kT}\ , \tag{15}$$

where

$$A=\tilde{g}\mu_B\tfrac{1}{2}H+[\rho(\mu)(J_a^2+J_b^2)+B_0/\mathfrak{N}]\tfrac{1}{2}\mathfrak{N}\langle S^z\rangle\ ,$$

$\tilde{g}=g[1+\frac{1}{2}\rho(\mu)(J_a+J_b)]$ being the renormalized g factor.

To discuss the possible solutions of the self-consistent Eq. (15), let us start from the situation when the effect of the interaction with the SDW is absent, it is $J_{a,b}(Q)=0$. In this region the system of localized spins becomes ferromagnetic below the Curie temperature T_{C_0}, because of the homogeneous, ferromagnetic spin-spin interaction.

Let us couple gradually now the spin-SDW interaction by augmenting $J_{ab}(Q)$ and consequently H_0. In order to see what happens with initial ferromagnetism let us analyze the behavior of the Curie temperature T_C. In order to do that let us develop Eq. (15) around $M=0$. Thus we get for $\zeta_C=4kT_C/B_0=T_C/T_{C_0}$

$$\zeta_C^2(1-\zeta_C)=\left(\frac{H_0}{B_0}\right)^2\ . \tag{16}$$

If $|H_0/B_0|\ll 1$ then $\zeta_C\simeq 1-(H_0^2/B_0)$. Thus the effect of the random field is to lower the Curie temperature. Equation (16) has no positive solution if

$$H_0\equiv J_{ab}(Q)\frac{\Delta}{V}>\frac{B_0\sqrt{2}}{3}\ . \tag{17}$$

If Eq. (17) is true the ferromagnetic state is no longer possible, nor is the state of the impurity spins a paramagnetic one since, in the absence of the external magnetic field, the local mean values of the spin projection are nonzero. We conclude that Eq. (17) becomes thus a necessary condition for the "spin-glass-like" magnetic structure mentioned in Sec. II.

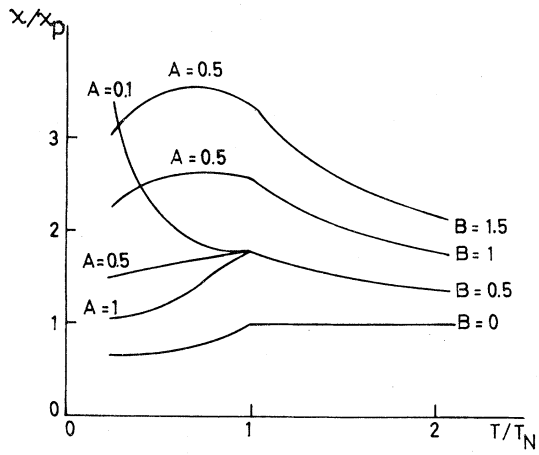

FIG. 3. Temperature dependence of the magnetic susceptibility as given by relation (18) with $A=J_{ab}(Q)/V$ and $B=(2/\pi)[\chi_L(T_N)/\chi_P]$.

Concerning the behavior of the localized magnetic moments, if Eq. (17) is not true, one might imagine a sort of ferromagnetic behavior affected by the presence of the random field. The detailed features of this state will be discussed elsewhere.[13]

The total magnetic susceptibility of the spin-glass-like magnetic structure, containing itinerant and localized contributions, is

$$\chi=\chi_P\left[\tfrac{2}{3}+\tfrac{1}{3}\int_0^D d\epsilon/2kT/ch^2\left(\frac{\epsilon^2+\tilde{\Delta}^2}{4k^2T^2}\right)^{1/2}\right]+\frac{\mathfrak{N}\tilde{g}^2\mu_B^2S(S+1)}{3k(T-\theta I)}I\ , \tag{18}$$

where

$$I=\frac{2}{\pi}\int_0^{H_0}\frac{dx}{ch^2x/2kT}\frac{1}{(H_0^2-x^2)^{1/2}}\ .$$

If we choose the limit $\theta I/T\ll 1$, we may approximate Eq. (18), for $T\to T_N$, by

$$\chi=\chi_P+\chi_L(T_N)+\frac{\mathfrak{N}\tilde{g}\mu_B^2S(S+1)}{3kT_N}\frac{T_N-T}{T_N}-\frac{\Delta^2}{4k^2T^2}\left(\tfrac{2}{3}\chi_P+\frac{J_{ab}^2(Q)}{2V^2}\frac{N\tilde{g}^2\mu_B^2S(S+1)}{3kT}\right)\ .$$

Using the known development[11] of Δ for $T\to T_N$,

$$\Delta=\pi kT_N\left(\frac{4}{F\xi(3)}\right)^{1/2}\left(\frac{T_N-T}{T_N}\right)^{1/2}\ ,$$

we get up to terms in $[(T_N-T)/T_N]^2$,

$$\chi(T)=\chi(T_N)+\frac{T_N-T}{T_N}\left[\chi_L(T_N)-\frac{\pi}{7\xi(3)}\left(\tfrac{2}{3}\chi_P+\frac{J_{ab}^2(Q)}{2V^2}\chi_L(T_N)\right)\right]\ . \tag{19}$$

The linear temperature dependence below T_N is very weak around some critical concentration for which the bracket is zero. Using Eq. (15) one gets for $T=0$

$$\chi(0)=\tfrac{2}{3}\chi_P+0.23[V/J_{ab}(Q)]\chi_L(T_N)\ ,$$

which indicates that Eq. (18) does not exhibit important changes as a function of temperature. Figure 3 shows the results of a numerical analysis of relation (18).

VI. DISCUSSION AND CONCLUSIONS

The theoretical results obtained are in agreement with the experimental ones. Figures 4 and 5 show the concentration dependence of the Néel temperature and of the high-temperature limit ($T\rightarrow\infty$) of the magnetic susceptibility (χ_0) for Cr-Yb and Cr-Er alloys, respectively. As is known, χ_0 contains temperature-independent contributions such as orbital paramagnetism, ionic diamagnetism, Pauli paramagnetism of s and d electrons, etc. If all the contributions, excepting the Pauli paramagnetic susceptibility of $3d$ electrons χ_P, are only weakly affected by alloying, we may consider $\Delta\chi_0\simeq\Delta\chi_P$. Thus experimental χ_0 values might be used instead of χ_P in Eq. (11). Full lines in Figs. 4 and 5 are given by Eq. (11). The fit values of χ_0 are plotted with dashed lines.

The plot $\ln(T_N/T_{N_0})$ vs χ_0, Fig. 6, shows that the linear relationship predicted by Eq. (11) is only roughly fulfilled for Cr-Yb but quite accurate for Cr-Er. This difference might occur because of the concentration-dependent terms in Eq. (10) since the concentrations corresponding to the same χ_0 are higher for Cr-Yb than for Cr-Er.

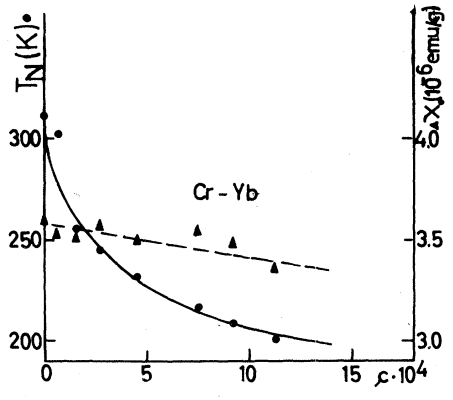

FIG. 4. Concentration dependence of the Néel temperature and of the high-temperature limit of the magnetic susceptibility χ_0, for Cr-Yb alloys. The full line is a fit with Eq. (11).

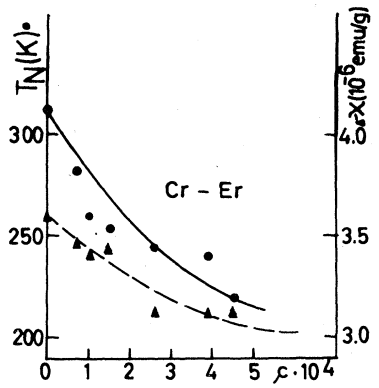

FIG. 5. Concentration dependence of the Néel temperature and of the high-temperature limit of the magnetic susceptibility χ_0, for Cr-Er alloys. The full line is a fit with Eq. (11).

The agreement between Eq. (11) and the experimental results indicates that the main mechanism in the Néel-temperature shift is the potential scattering by the impurities. Analyzing from this point of view our experimental results concerning $3d$ impurities in chromium,[14–16] we may conclude that Fe and Mn as the impurity do not exhibit this property while Co does.

For $T>T_N$ experimental results are compared with a Curie-Weiss law (full line in Figs. 1 and 2). The paramagnetic Curie temperatures, for both systems, are given in Figs. 7 and 8. An effective moment per impurity atom was determined and displayed in Figs. 7 and 8. The high values of the effective magnetic moment are not to be taken for the magnetic moment of an impurity atom but, rather, for the whole paramagnetic response of the region surrounding the impurity and interacting with it. It is

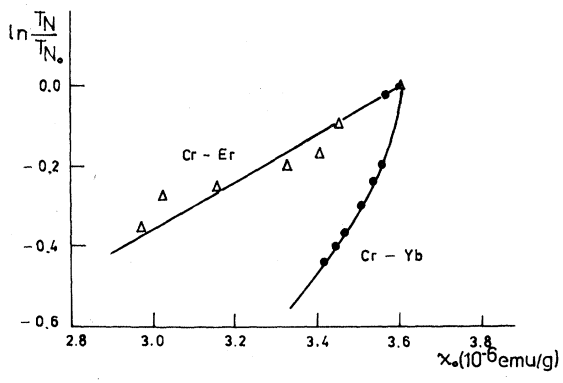

FIG. 6. $\ln(T_N/T_{N_0})$ vs χ_0 for Cr-Yb and Cr-Er alloys.

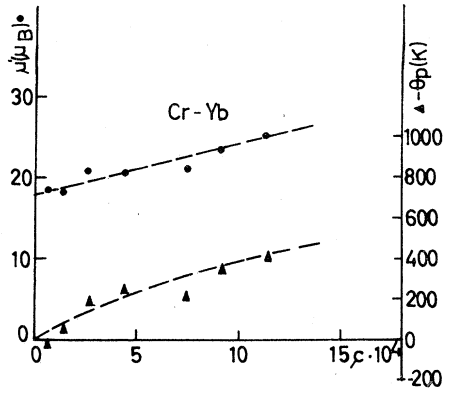

FIG. 7. Concentration dependence of the effective magnetic moment μ' per Yb atom and of the paramagnetic Curie temperature for Cr-Yb alloys.

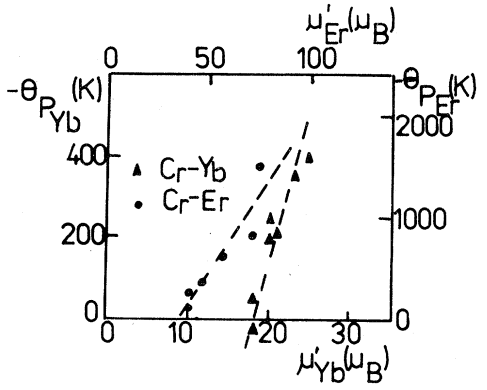

FIG. 9. Paramagnetic Curie temperature vs effective magnetic moment per impurity atom for Cr-Yb and Cr-Er alloys.

also possible that alloying causes a slight localization of chromium $3d$ electrons. Thus, the computation of a magnetic moment "per impurity atom" has only a formal sense in those situations when strong interactions with the host occur.

Relation (14) indicates the usefulness of a plot like the one shown in Fig. 9. The high negative value of θ_p and the almost linear dependence of θ_p as a function of μ' indicate that the most important term in Eq. (14) is the one related with the f-d interaction. The second term in Eq. (14), it is the f-f interaction, is negligible. This fact shows that the impurity concentrations are small enough to consider the observed magnetic behavior as a one-impurity effect.

The good agreement between Eq. (14) and experimental data allows the evaluation of the electron-hole coupling constant V out of the slopes of the curves shown in Fig. 9. The values obtained are $V_{\mathrm{Cr-Yb}}$ $=0.20$ eV and $V_{\mathrm{Cr-Er}}=0.23$ eV.

To try to explain the concentration dependence of the Néel temperature, or θ_p and μ', is a hard task since it supposes the complete knowledge of the density-of-states function and of the spatial behavior of the f-d exchange integrals (J). Thus, we consider universal relations like Eqs. (11) and (14) useful in the attempt to decide whether or not an interaction mechanism is present in the studied system.

Concerning the temperature regions with $T < T_N$, Figs. 1 and 2 show a very weak, quasilinear temperature dependence. The paramagnetic behavior of the impurity spins is suppressed. As shown in Sec. III such a behavior might occur if random impurity-spin–SDW interactions are considered. The agreement between the prediction (19) of the considered model and the actual magnetic behavior of Cr-Er and Cr-Yb alloys indicates the conclusion that these alloys have a "spin-glass-like" magnetic structure.

Because of experimental difficulties we could not obtain samples with lower or higher concentrations than the reported ones. Thus, the limits of this magnetic behavior could not be defined for the investigated systems. Low-temperature measurements are also required in order to complete and verify the statements of this paper.

Concerning the model we might conclude the following.

(i) The interaction between the randomly distributed $4f$ spins and the $3d$ electrons leads to an enhancement of the order parameter

$$\tilde{\Delta}=\Delta\left(1+\frac{\mathfrak{N}}{4V}|J_{ab}(Q)|^2\langle S^z\rangle\right) .$$

The potential scattering of conduction electrons on impurities has an opposite effect.

(ii) The random interaction impurity-spin–SDW leads to an interesting and peculiar magnetic behavior

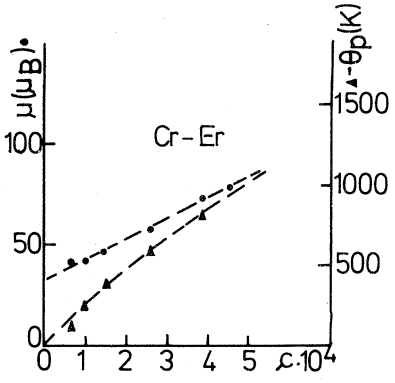

FIG. 8. Concentration dependence of the effective magnetic moment μ' per Er atom and of the paramagnetic Curie temperature for Cr-Er alloys.

of localized magnetic moments which has common features with the spin-glass state. Thus if Eq. (17) is true the configurational and thermodynamical mean-value of the local spin projection S_f^z is

$$\langle\langle S_f^z\rangle\rangle \equiv \int_{-\infty}^{+\infty} P(\tilde{H})\langle S_f^z\rangle d\tilde{H} = \int_{-\infty}^{+\infty} P(\tilde{H})\tfrac{1}{2}\tanh\frac{\tilde{H}}{2kT}d\tilde{H} = 0 \ .$$

The Anderson order parameter for spin-glasses is

$$\langle\langle S_f^z\rangle^2\rangle \equiv \int_{-\infty}^{+\infty} P(\tilde{H})\langle S_f^z\rangle^2 d\tilde{H} = \frac{1}{\pi}\int_0^{\pi/2} dx \tanh\left(\frac{H_0}{2kT}\sin x\right) \neq 0 \ .$$

The order parameter vanishes for $H_0 \to 0$. In this case the last integral might be performed

$$\langle\langle S_f^z\rangle^2\rangle \simeq \frac{H_0^2}{(4kT)^2} = \frac{|J_{ab}(Q)|^2}{V^2(4kT)^2}\Delta^2(T) \ .$$

Thus, the "spin-glass temperature" of the investigated magnetic structure is identical with the Néel temperature for itinerant electrons.

However, it must be underlined that the magnetic behavior investigated in this paper is not the behavior of a spin-glass system mainly because the random interaction is not a spin-spin one but a spin-SDW one. The "spin-glass-like" magnetic structure is also not reducible to an antiferromagnetic one since there are no antiferromagnetic correlations between impurity spins.

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