PHYSICAL REVIEW B VOLUME 19, NUMBER 9 1 MAY 1979

The Stoner glass

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(Received 1 May 1978)

A theory of spin-glass condensation in a disordered itinerant-electron system without well-developed local moments is presented. The theory applies to finite-concentration impurity systems at temperatures well below their Kondo temperatures; such as *Rh*Co. The local spin fluctuations at impurity sites are coupled to each other via the magnetic response of the host-metal electrons, and since this interaction is effectively random, a phase transition to a frozen state characterized by an Edwards-Anderson spin-glass order parameter can occur. A mean-field description of this transition and state is given here, paying particular attention to the requirement of mutually consistent approximations for the susceptibility, order parameter, and order-parameter susceptibility. An interesting formal aspect of the theory is the fact that fluctuation corrections to the susceptibility, of the sort which occurs in the Moriya-Kawabata and Hertz-Klenin theory of itinerant ferromagnets, are necessary for a consistent description of the spin-glass case.

I. INTRODUCTION

Theoretical descriptions of ferromagnetism in solids span a range between two extreme pictures—one based on Heisenberg-Ising localized-spin models and the other on the Slater-Stoner-Wohlfarth itinerant-electron theory. Some materials conform better to one picture and others to the other one. Thus one also expects that among the many systems exhibiting spin-glass behavior, some should be reasonably good realizations of localized-spin models and others should fit more easily into an itinerant-electron description. While there has been extensive study of Heisenberg or Ising spin-glass models,[1-5] no itinerant models have been discussed in the literature. This paper therefore presents a theoretical framework for itinerant-electron spin-glasses, which one can call "Stoner glasses." We begin with a discussion of the sorts of systems likely to fit such a theory and write down the simplest model likely to contain the relevant physics. After a few approximations made for mathematical convenience, the model is then solved in mean-field theory.

The definition of a spin-glass is sometimes taken to include as a precondition the existence of well-defined localized moments on impurity atoms.[6] Such a definition would not classify the systems we are discussing here as spin-glasses, but this seems to be an artificial exclusion. (The corresponding definition for ordered magnetism would exclude nickel from the list of ferromagnets, for example.) We will take the point of view here that the existence of an Edwards-Anderson order parameter is a sufficient (though not necessary) condition to call a material a spin-glass. Thus, drawing on the analogy with the ferromagnetism of Ni or the antiferromagnetism of Cr, we investigate here the case where randomness of a suitable kind is sufficient to drive a system into a state where the frozen-spin density pattern is a random one, characterized by the Edwards-Anderson order parameter $q = \langle\langle S(x)\rangle^2\rangle_c$. [Here and henceforth, thermal averages are denoted in the conventional way $\langle\langle\ \rangle\rangle$, and averages over all possible impurity configurations by $\langle\ \rangle_c$.]

When do we expect that an itinerant-electron description of a spin-glass state will be appropriate? For ordinary magnetic order, itinerant models are generally more relevant for transition metals while localized ones are better for rare earths, so we expect cases where both host and impurity elements and transition metals to be potential Stoner glasses. Another way to answer the question is to look at the dilute limit of the system in question. If we have a very high Kondo temperature, so that a local-spin-fluctuation description is more relevant than a Kondo model, then any spin-glass order that sets in at a temperature $T_g \ll T_K$ cannot involve couplings between well-defined local moments, but must come about because of interactions between the virtual spin fluctuations. The latter situation is essentially the picture we develop here and can be taken as the defining condition for a Stoner glass: $T_g \ll T_K$. Several systems which meet this criterion have been studied experimentally, most notably *Au*Co, *Rh*Fe, and *Rh*Co.[7-9]

Another point to note, although it is in many cases an academic one, is that at sufficiently low temperature *any* impurity moment will be quenched,[10] and the localized-spin description will fail. Thus an itinerant picture is always necessary in principle as $T \to 0$.

II. MODEL

The model we consider here is based on a picture of an alloy where both host and impurity are

transition metals. We suppose that the principal difference between the two kinds of atoms is that one has a larger intratomic Coulomb repulsion energy I. Thus we write down a tight-binding model where this Coulomb (Hubbard) term is included only at the impurity sites

$$H=\sum_{ij\sigma} t_{ij}c_{i\sigma}^{\dagger}c_{j\sigma}+I\sum_{i\in S} n_{i\uparrow}n_{i\downarrow} \tag{2.1}$$

where S is the set of impurity sites. (For simplicity, we consider only a one-band model.) This can be thought of as either a dilute Hubbard[11,12] or a concentrated Wolff model.[13] The randomness which leads to spin-glass behavior originates in the random distribution of impurity sites S. It can be handled mathematically in several approximate ways. The qualitative results do not depend on which way we choose, as we shall argue later.

If I were very large, there would be well-defined moments on impurity atoms (above a very low Kondo temperature) and a Heisenberg model would be a sensible starting point. That is not the situation we deal with here. We consider an I in the intermediate region $IN(E_F)\approx 1$ where local spin fluctuations on the impurities persist for times long compared with electronic hopping states but short relative to the inverse temperature. In terms of the spin fluctuation energy ω_s

$$T\ll\omega_s\ll t_{ij}. \tag{2.2}$$

The one-impurity problem has a spurious instability at $I\chi_{\rm loc}^0=1$ where $\chi_{\rm loc}^0$ is the local susceptibility of the noninteracting system; fluctuations actually prevent the susceptibility from diverging at any finite I.[14] We will sweep all the problems associated with this difficult aspect of the problem under the rug by treating I as a phenomenological parameter with $I\chi_{\rm loc}^0<1$. This is standard practice in local-spin-fluctuation theory (LSF) for the single-site problem.[14] Some justification for this practice even for large I (provided $T\ll T_K$) is provided by recent work on the Kondo problem.[15]

What we want to do now is to examine whether the interactions between LSF sites mediated by the host-metal electrons can drive the system into a macroscopic condensed state. Depending on details of the interaction, this state could be ferromagnetic (as in RhNi and PdNi), antiferromagnetic, or, if appropriate randomness is present, a spin-glass. Since we are interested in the spin-glass, we shall emphasize the random aspect of the interaction in what follows.

To express this formally, we consider calculating the spin susceptibility $\chi(r_i,r_j)\equiv\chi_{ij}$ in mean-field theory. [The indices i,j label impurity (LSF) sites.] We have

$$\chi_{ij}=\chi_{ij}^0+\sum_k \chi_{ik}^0 I\chi_{kj}. \tag{2.3}$$

or, solving interatively, the series

$$\chi_{ij}=\chi_{ij}^0+\sum_k \chi_{ik}^0 I\chi_{kj}^0+\sum_{kl}\chi_{ik}^0 I\chi_{kl}^0 I\chi_{lj}^0+\cdots, \tag{2.3a}$$

where χ_{ij}^0 is the susceptibility when $I=0$, that is, the susceptibility of the host. [If every site had $I\neq 0$ (the Hubbard model), (2.3) would be soluble by Fourier transform, yielding the familiar Stoner instability. The randomness of the distribution of impurities prevents this simple solution in the present problem.] It is convenient now to sum separately all repeated interactions on the same impurity, e.g., $k=l$. Then, defining the exchange enhanced interaction

$$g_0=I/(1-I\chi_{ii}^0) \tag{2.4}$$

and the intersite part of the susceptibility matrix

$$\phi_{ij}=\chi_{ij}^0(1-\delta_{ij}), \tag{2.5}$$

we have

$$\chi_{ij}=\chi_{ii}^0\delta_{ij}+\phi_{ij}+\sum_{kl}(\chi_{ii}^0\delta_{ik}+\phi_{ik})g_{kl}(\chi_{jj}^0\delta_{jl}+\phi_{lj}), \tag{2.6}$$

where the g matrix is given by the simple series

$$g_{ij}=g_0\delta_{ij}+g_0\phi_{ij}g_0+\sum_k g_0\phi_{ik}g_0\phi_{kj}g_0+\cdots. \tag{2.7}$$

This rearrangement illustrates the fact that the intrasite part of χ^0 just determines the local exchange enhancement, producing slow local spin fluctuations, while the intersite terms lead to a large degree of randomness in the series (2.7) because of the random arrangement of impurity sites. It is this randomness which can produce spin-glass behavior. More explicitly, the Fourier transform of χ_{ij}^0 contains singularities at the Fermi surface, leading to oscillatory behavior in χ_{ij}^0 as a function of $\vec{r}_i-\vec{r}_j$—the same mechanism responsible for exchange interactions of varying sign in Ruderman-Kittel-Kasuya-Yosida (RKKY)-coupled Heisenberg spin-glasses. The only difference is that here it is the LSF sites, not well-defined moments, which are coupled in this fashion.

For our idealized model (2.1), the free part of H is that of the pure host (since we assumed that the only thing different about an impurity atom was its I), so χ_{ij}^0 will contain oscillations characteristic of a pure system. More realistically, we should consider the one-electron effects of disorder as well; these will tend to smear the Fermi-surface singularities and damp the RKKY oscillations. But as

long as ϕ_{ij} of either sign can still occur and not too many ϕ_{ij} are of one sign, spin-glass behavior is still likely.

To work out the consequences of these interactions, we need to set up a perturbation theory for calculating configurationally averaged quantities. In order to do this conveniently, it is helpful to further idealize our model. This simplication is commonly made for Heisenberg spin-glasses: instead of randomly situated impurity sites with the interaction a given (nonrandom) function of the distance between impurities, we consider instead the problem of LSF centers on a lattice, coupled by ϕ_{ij} which are random. In the simplest model we can make, we take

$$\langle \phi_{ij} \rangle_c = 0 ,$$
$$\langle \phi_{ij}\phi_{kl} \rangle_c = (\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})\Delta_0(\vec{r}_i - \vec{r}_j) , \tag{2.8}$$

with all higher cumulants vanishing. Although the question has not been investigated very thoroughly, the current folklore[16] has it that more elaborate models (e.g., with a distribution appropriate to the RKKY interaction) would not change qualitatively any properties of the resulting spin-glass.

We will also need to consider finite frequency susceptibilities because we are dealing with a quantum-mechanical problem. We therefore discuss the structure of $\chi^0_{ij}(\omega)$ at finite ω and how to incorporate this into our model. For small k, we have[17] (for a free Fermi gas)

$$\delta\chi^0(q,\omega) \equiv \chi^0(q,\omega) - \chi^0(q,0)$$
$$\approx N(0)(\pi|\omega|/2qv_F)\theta(qv_F - |\omega|) . \tag{2.9}$$

(In this discussion, ω is a Matsubara frequency $2\pi m T$.) On Fourier transforming, we find that

$$\delta\chi^0_{ij}(\omega) \propto |\omega| r_{ij}^{-2} \cos(2k_F r_{ij}) \tag{2.10}$$

for small ω and $i \neq j$, and

$$\delta\chi^0_{ii}(\omega) \approx N(0)|\omega|/(E_F/n) \tag{2.11}$$

for $i = j$. Equation (2.11) produces a frequency-dependent locally enhanced interaction or t matrix

$$g_0(\omega) = \frac{I}{1 - I\chi^0_{ii}(\omega)} \approx \frac{g_0}{1 + |\omega|\tau_s} , \tag{2.12}$$

where τ_s is a local-spin-fluctuation time, while (2.10) suggests a generalization of (2.8):

$$\langle \phi_{ij}(\omega) \rangle_c = 0 ,$$
$$\langle \phi_{ij}(\omega)\phi_{kl}(\omega) \rangle_c = (\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$$
$$\times [\Delta_0(\vec{r}_i - \vec{r}_j) + |\omega|\Delta_1(\vec{r}_i - \vec{r}_j)$$
$$+ |\omega|^2\Delta_2(\vec{r}_i - \vec{r}_j)]$$
$$\equiv (\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})\Delta(\vec{r}_i - \vec{r}_j ; \omega) . \tag{2.13}$$

The term linear in $|\omega|$ does not vanish because $\chi^0_{ij}(0)$ and $\delta\chi^0_{ij}(\omega)$ are not independent. We also note that both χ^0_{ij} and $\delta\chi^0_{ij}$ decay rapidly enough with r_{ij} that all the Fourier transforms $\Delta_i(k)$ are well behaved at $k = 0$.

To complete our model, we will need the lowest-order anharmonic couplings between spin fluctuations, pictured in Fig. 1(a):

$$\Lambda_{ijkl}(\omega_1, \omega_2, \omega_3)$$
$$= -T \sum_E G_{ij}(E)G_{jk}(E+\omega_1)G_{kl}(E+\omega_1+\omega_2)$$
$$\times G_{li}(E+\omega_1+\omega_2+\omega_3) . \tag{2.14}$$

[The sum is over the fermion Matsubara frequencies $2\pi(n+\frac{1}{2})T$.] Λ depends on all the ω_i and on all the spatial separations $r_i - r_j$, and so on. Just as in the case of its two-point counterpart

$$\chi^0_{ij}(\omega) = -T \sum_E G_{ij}(E)G_{ji}(E+\omega) ; \tag{2.15}$$

however, we expect Λ to oscillate in space. Thus, when we change the problem from one of randomly located impurity sites to one of a lattice of randomly coupled LSF centers, Λ_{ijkl} acquires an effectively random behavior for $i \neq j$, etc. To keep the problem manageable, we ignore the randomness and the frequency dependence in Λ, on the argument that the spatial variation tends to get averaged out anyway and the frequency dependence of the vertex (which varies on a scale $\sim E_F$) is unimportant relative to that in the susceptibility (which varies on a scale τ_s^{-1}). The simplest way to do this[18] (expressed in momentum space) to to replace the general Λ by its value when all incoming momenta vanish. That is, we use as our coupling

$$\lambda = -T \sum_{p,E} G^4(p,E) = -\tfrac{1}{6} N_T''(E_F) \tag{2.16}$$

where $N_T(E_F)$ is a thermally averaged density of states

$$N_T(E_F) = \int d\epsilon \left(-\frac{\partial f}{\partial \epsilon}\right) N(\epsilon) . \tag{2.17}$$

As is common in other spin-fluctuation problems

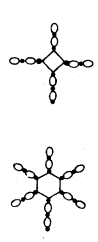

FIG. 1. (a) Four-spin fluctuation vertex; (b) the six-spin fluctuation vertex.

formulated in this way, we ignore higher-order paramagnon-paramagnon interaction vertices [Fig. 1(b)]. As long as the fluctuations are not too anharmonic, this should not be a source of serious error. [More precisely, as long as typical internal field fluctuations (expressed in energy units) are small compared to energies over which $N_T(E)$ can be approximated by a quadratic expansion around E_F, higher-order anharmonicity should be negligible.] We assume $\lambda > 0$ so that the anharmonicity stabilizes the fluctuations.

We now describe the perturbation theory for this model, beginning with the structure of the diagrams for a fixed set of ϕ_{ij}'s and then describing the averaging over the distribution of the ϕ_{ij}'s. We want to consider, in particular, the Green's function g_{ij}. The building blocks of perturbation theory are the bare Green's function $g_0(\omega)$, the vertex λ, and the random intersite susceptibility $\phi_{ij}(\omega)$. We represent $g_0(\omega)$ by a thin solid line, $\phi_{ij}(\omega)$ by a wavy line, and λ by a square. A ϕ_{ij} line can be inserted between any pair of g lines; thus, the diagrams of Fig. 2(a) represent the random-phase approximation (RPA) series (2.7). Some diagrams for g with anharmonic vertices are shown in Fig. 2(b).

In calculating susceptibilities and correlation functions, we have to remember that an external field couples into both parts of χ^0 (χ^0_{ii} and ϕ_{ij}), as is evident from the form of (2.6), and into the anharmonic vertex λ as well. Thus, for example, diagrams like Fig. 2(c) contribute to χ.

A few more comments are in order here before we discuss the configurational averaging: (i) The quantities $g_0(\omega)$, $\phi_{ij}(\omega)$, and λ all have weak

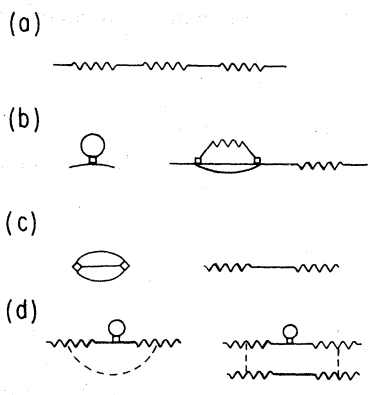

FIG. 2. (a) RPA series for g_{ij}; (b) diagrams for g involving λ vertices; (c) diagrams for χ involving λ and ϕ; (d) configurationally averaged diagrams for $\langle g_{ij}\rangle_c$ and $\langle g^2_{ij}\rangle_c$.

$[O(T/T_F)^2]$ temperature dependences which will turn out to be capable of driving a spin-glass transition as a certain function of these quantities passes through a critical value. (ii) One also has to keep track of component indices on the propagators and vertices. The rule is simply that a λ vertex with incoming g lines with spin component labels α, β, γ, and δ has the value

$$\lambda_{\alpha\beta\gamma\delta} = \tfrac{1}{4}\lambda \mathrm{Tr}(\sigma_\alpha\sigma_\beta\sigma_\gamma\sigma_\delta), \tag{2.18}$$

where the σ's are Pauli matrices. (iii) We have ignored density fluctuations.

It is also a simple matter to generalize the model to the (more realistic) case where $\langle\phi_{ij}\rangle_c$ is nonzero. The bare Green's function $g_0(\omega)$ is just replaced by a nonlocal propagator

$$g^0_{ij}(\omega) = \frac{I}{N}\sum_k e^{i\vec{k}\cdot(\vec{r}_i-\vec{r}_j)}[1 - I\langle\chi^0(k,\omega)\rangle_c]^{-1}. \tag{2.19}$$

The averaging of these correlation functions, or products of arbitrary numbers of such functions, over the distribution of the random variables is now straightforward to describe formally. One simply connects all the "bonds" ϕ_{ij} together in pairs by dotted lines. A pair ϕ_{ij} and ϕ_{kl} so joined stands for the variance given in Eq. (2.13). Figure 2(d) shows examples of $\langle g_{ij}\rangle_c$ and $\langle g^2_{ij}\rangle_c$. The latter quantity will turn out to be the Edwards-Anderson order-parameter susceptibility.

We conclude this section with a discussion of how the spin-glass instability reveals itself in the behavior of correlation functions. An ordinary magnetic instability would occur when a particular Fourier component of $\langle\chi_{ij}\rangle_c$ became infinite. That is, a finite external field would produce an infinite mean magnetization. We assume that this does not happen here. We consider instead subjecting the system to a *random* magnetic field

$$\langle h_i\rangle_c = 0, \quad \langle h_i h_j\rangle_c = h^2\delta_{ij} \tag{2.20}$$

and examining the mean-square resulting magnetization

$$q(h^2) = \langle\langle S_i\rangle^2\rangle_c = Dh^2, \tag{2.21}$$

where the second equality defines a linear-response coefficient for small variance h^2 of the random field. If the response $q(h^2)$ diverges, the system is unstable and stabilizes itself by generating a spontaneous value of q even when $h = 0$. This is the Edwards-Anderson order parameter. Therefore we will study the onset of the spin-glass order by calculating the order-parameter susceptibility D in the normal state and looking for a divergence in it. In the spin-glass state, we will write a mean-field equation to determine the spontaneous value of q.

The uniform order-parameter susceptibility D

can be generalized to finite wave number in the following way. Let the variance of the applied random field (2.20) vary from site to site. Then

$$\langle\langle S_i\rangle^2\rangle_c = \langle\chi_{ij}^2\rangle_c h_j^2, \tag{2.22}$$

where h_j^2 is the variance of the field at site j. Defining $D_{ij} = \langle\chi_{ij}^2\rangle_c$ and $D(k)$ as its Fourier transform, the parameter in (2.21) is just $D(0)$.

The preceding discussion may seem to the reader to be only very weakly connected with the notion of a spin-glass as a particular frozen irregular magnetization pattern $\langle S_i\rangle \neq 0$, which suggests that the conjugate field to the order parameter is an external magnetic field with the same irregular spatial variation. One can in fact proceed in this fashion in numerical simulation calculations, but we do not know how to formulate an analytical theory of the spin-glass on this basis. We need a formalism which allows us to calculate the probability distributions of various physical quantities over the statistical ensemble of macroscopically indistinguishable systems defined by, e.g., (2.8). Thus our order parameter and conjugate field have to be parameters of these probability distributions, not microscopic variables pertaining to only one particular realization of the ensemble. The simplest way to try to characterize a spin-glass in such a fashion appears to be through the (configurational) variance of the thermal average magnetization, that is, by the Edwards-Anderson order parameter. The conjugate field also has to be such a statistical parameter, and the previous discussion shows that the variance of the field, h^2 (2.20) is the appropriate choice if q is to be the order parameter.[19]

III. MEAN-FIELD THEORY

It is known that in spin-glasses, the notion of a "mean-field" theory is not as straightforward as it might appear.[3] Here we take the view that a "correct" mean-field theory must satisfy two criteria: (a) It should be correct to leading order in the reciprocal of the coordination number z for a hypothetical situation in which $\phi_{ij} = 0$ except for nearest neighbors. (b) It should not violate any conservation laws. In particular, approximations for the free energy, the susceptibility, and the order-parameter susceptibility D_{ij} can not be made independently on one another; they must be related by Ward identities.[20,21]

The more recent versions of mean-field theory for the Ising spin-glass conform to these requirements. The present theory has an analogous structure.

We begin, in the paramagnetic state, with an approximation to the average free energy [Fig. 3(a)]

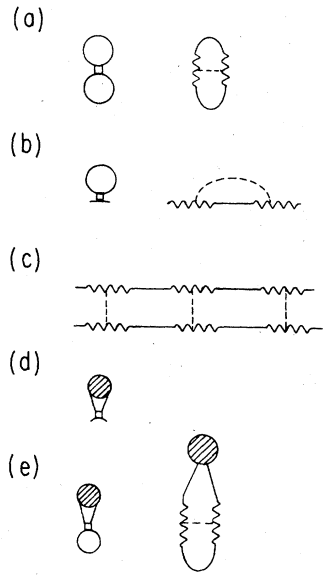

FIG. 3. Mean-field approximation (solid lines stand for the full Green's function G). (a) Diagrams for free energy, $T > T_g$; (b) self-energy diagrams, $T > T_g$; (c) typical ladder diagram for F; (d) extra self-energy diagram for $T < T_g$; (e) extra free-energy diagrams for $T < T_g$.

which is correct to lowest order in Δ and λ, and also to leading order in $1/z$. In these diagrams the thick solid lines stand for the full $G(\omega) = \langle g(\omega)\rangle_c$ (We confirm our attention to the simple case $\langle\phi_{ij}\rangle_c = 0$ for now.) In the standard way,[21] the approximation for the self-energy $\Sigma(\omega)$, defined by the Dyson equation

$$G(\omega) = g_0(\omega) + g_0(\omega)\Sigma(\omega)G(\omega) \tag{3.1}$$

is obtained by functional differentiation of the free-energy diagrams with respect to G. Thus $\Sigma(\omega)$ is given by Fig. 3(b):

$$\Sigma(\omega) = \tfrac{5}{2} T\lambda \sum_{\omega'} G(\omega') - \Delta(0)G(\omega). \tag{3.2}$$

Thus the randomness enhances G (or χ), while the anharmonicity suppresses it. (In the corresponding Ising case,[3] a spherical model constraint is imposed, forcing the two effects to cancel exactly. No such exact cancellation happens here.)

Leaving aside the solution of (3.1) and (3.2) for G, we proceed to derive the order-parameter susceptibility, which is proportional to the fluctuations in g_{ij}:

$$\begin{aligned} D_{ij}(\omega) &= [\Delta(k,\omega) + (\chi_{ii}^0)^2]^2 \langle g_{ij}(\omega) g_{ij}(-\omega)\rangle_c \\ &\equiv [\Delta(k,\omega) + (\chi_{ii}^0)^2]^2 F_{ij}(\omega). \end{aligned} \tag{3.3}$$

[The factorized form (3.3) is, strictly speaking, only a mean-field result; it ignores correlations between ϕ's at the external vertices of the χ_{ij} diagrams and those in the g_{ij}'s.] If we now define a vertex function by

$$F_{ij}(\omega) - \delta_{ij} G^2(\omega) = G^4(\omega)\Gamma_{ij}(\omega), \tag{3.4}$$

it then follows that Γ_{ij} is obtained by functional differentiation of the self-energy:

$$\Gamma_{ij}(\omega) = \delta\Sigma_i^{(d)}(\omega)/\delta G_j(\omega), \tag{3.5}$$

where $\Sigma^{(d)}$ consists of all diagrams for Σ that become disconnected (except for being tied together by averaging lines) when a G line is cut. In the present case, just one of the two diagrams for Σ (the one with the ϕ_{ij}'s) meets this requirement, and we obtain the simple ladder series [Fig. 3(c)]

$$F(k, \omega) = G^2(\omega)/[1 - G^2(\omega)\Delta(k, \omega)]. \tag{3.6}$$

This shows an instability when

$$1 = G^2(0)\Delta(0, 0) = G^2(0) \sum_{i \neq j} \langle [\chi_{ij}^0(0)]^2 \rangle_c . \tag{3.7}$$

This is the spin-glass counterpart of the ferromagnetic Stoner criterion $1 = I\sum_j \chi_{ij}^0$. Note that while the direct calculation of $G(0)$ from (3.1) and (3.2) is somewhat messy because of the anharmonic term, $G(0)$ is well defined experimentally as $I\chi/\chi_{\text{Pauli}}$. Since both Δ and G are temperature dependent, (3.7) can define a critical temperature below which spin-glass ordering sets in. Although it is in general a complicated matter to calculate T_g, taking into account the temperature dependence of all these quantities, we can study simply the case where the local spin fluctuations have a lifetime τ_s such that $T_g > \tau_s^{-1}$. In this case their statistics are essentially classical,[18] and only the $\omega' = 0$ term in the sum in (3.2) is sizeable. We then expand the Green's function as

$$G = g_0\{1 - g_0^2[\Delta(0) - \tfrac{5}{2}\lambda T] + \cdots\}^{-1}. \tag{3.8}$$

The explicit linear T dependence will dominate the quadratic one implicit in g_0, $\Delta(0)$, and λ, and we can solve (3.7) for T_g

$$T_g \approx \frac{g_0[\Delta(0)]^{1/2} - 1 + g_0^2\Delta(0)}{\frac{5}{2} g_0^2 \lambda}. \tag{3.9}$$

We turn now to the spin-glass phase, for which we simply augment the self-energy $\Sigma(\omega)$ by a term dependent on the order parameter [Fig. 3(d)]. [This term comes from functionally differentiating the first diagram of Fig. 3(e).] It is clear that consistency requires that we add this term to the single-loop one in order to keep all terms of first order in λ when a spin-glass order parameter is present. Conversely, it would not be a sensible approximation to retain only the new term, discarding the previous single-loop one. This is one simple way to see the point that a proper mean-field theory for a spin-glass must include terms which would be the first (single loop) corrections to mean-field theory of an ordinary phase transition.[3] Thus

$$\Sigma(\omega) = \tfrac{5}{2} T\lambda \sum_{\omega'} G(\omega') + \tfrac{5}{2}\lambda Q - \Delta(0)G(\omega), \tag{3.10}$$

where Q (Fig. 4) is proportional to the Edwards-Anderson order parameter q

$$q = [(\chi_{ii}^0)^2 + \Delta(0)]Q. \tag{3.11}$$

The functional differentiation of Σ to obtain the reduced-order-parameter susceptibility F is unchanged by this modification; we still have (3.6) except that G is now Q dependent because of the extra term in Σ. How do we determine Q itself? Again, by functional differentiation of the free energy including the new graphs [Fig. 3(e)]. Just as the connected part of the functional derivative with respect to G gives Σ, the disconnected part gives us simply

$$Q = \Delta(0)G_Q^2(0)Q \tag{3.12}$$

(remembering that G is Q dependent). Thus, if we expand G to first order in Q

$$G_Q = G_0(1 - \tfrac{5}{2}\lambda Q G_0 + \cdots), \tag{3.13}$$

where G_0 is the value of $G(0)$ when $Q = 0$, we obtain the characteristic linear growth of Q in the spin-glass phase

$$Q = [\Delta(0)G_0^2 - 1]/5\lambda G_0. \tag{3.14}$$

That is, if $\Delta(0)$ and G_0 vary smoothly with temperature, $Q \propto (T_g - T)$ near the transition. This is characteristic of mean-field theory for all spin-glasses.[1-4,22] Thus, because of the term in (3.10) linear in Q, the susceptibility will have a discontinuity in slope at the transition—again, a familiar result. The magnitude of the suppression of χ is just

$$(G_0 - G_Q)/G_0 = \tfrac{5}{2}\lambda G_0 Q. \tag{3.15}$$

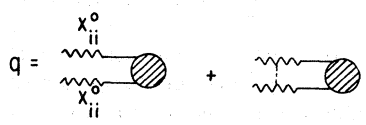

FIG. 4. Relation between Q and the Edwards-Anderson order parameter $q = \langle\langle S_i \rangle^2\rangle_c$.

We note further that the self-consistent equation for Q (3.12) is equivalent to the vanishing of the denominator of the order-parameter susceptibility (3.6) at $k=\omega=0$, *everywhere in the spin-glass phase*. The long-wavelength order-parameter fluctuations remain divergent, rather than being healed by the growth of the order parameter as in mean-field theory for an ordinary phase transition. This "soft" response also occurs in other mean-field theories[3,22] for spin-glasses. Lest any confusion arise, we also remark that this divergence has nothing to do with "Goldstone modes," since it occurs with a scalar order parameter as well.

The more general situation (2.19) where the susceptibility matrix elements χ^0_{ij} have a nonvanishing mean for $i\neq j$ as well as $i=j$ can be treated almost as easily as this simple case. The Dyson equation (3.1) is unchanged except that G and g_0 are momentum dependent, with

$$g_0(k,\omega)=I[1-I\langle\chi^0(k,\omega)\rangle_c]^{-1}, \tag{3.16}$$

Σ is (at this level of approximation) still momentum independent, so the modifications of the susceptibility are the same as in the previous case. However, the k dependence of G does modify the analytical form of the ladder series [Fig. 3(c)] for the reduced order-parameter susceptibility; instead of (3.6), we have

$$F(k,\omega)=\frac{\Pi(k,\omega)}{1-\Pi(k,\omega)\Delta(k,\omega)}, \tag{3.17}$$

where

$$\Pi(k,\omega)=\int\frac{d^dp}{(2\pi)^d}\,G(p,\omega)G(p+k,\omega). \tag{3.18}$$

Thus the instability condition (3.7) becomes

$$1=\left(\sum_j G^2_{ij}(0)\right)\left(\sum_{i\neq j}[\chi^0_{ij}(0)]^2\right). \tag{3.19}$$

Similar modifications occur in the spin-glass phase as well. The equation for the reduced order parameter Q (3.12) becomes

$$Q=\Delta(0)\Pi_Q(0,0)Q. \tag{3.20}$$

The qualitative conclusions remain unchanged. Q grows linearly with T_c-T and $F(0)$ remains infinite.

Thus all the qualitative features of the transition to a Stoner-glass state are the same as those found for other spin-glass models in the corresponding mean-field theories.[3,22] The fact that we have derived them in a fashion that imposed mutual consistency on approximations for different quantities enhances the credibility of the results for all the models. Of course, we do not know whether this mean-field condensation will survive the effects of fluctuations in three dimensions, but we do at least have a sensible, consistent mean-field theory. (The early mean-field theories[1,2] which were played by unphysical features like negative entropy at low temperatures, on the other hand, are not consistent in the sense we have required here, they omit the single-loop corrections.)

There are also a few points about the present theory which do not occur in the other models mentioned above, but are particularly relevant to the Stoner glass. The first of these is the presence of quantum effects (via the Matsubara sum) in the self-energy (3.2). In our treatment, these effects do not modify the general features of the spin-glass transition. Whether this conclusion would also apply to quantum Heisenberg models is unclear. Early work by Fischer[4] would support a universality hypothesis, but a more recent investigation by Klemm[23] leads to a second-order transition only in the limit where the spin magnitude S is infinite.

The second point to discuss is the temperature dependence of the susceptibility, particularly the effect of the single-loop term in Fig. 3(b) [Eq. (3.2)]. As we already noted, these terms, which would be the first corrections to mean-field theory for a ferromagnet, are necessary to a consistent mean-field theory here. The effects of such corrections for itinerant-electron ferromagnets and strongly enhanced paramagnets have been considered by a number of authors.[18,24] They find that the temperature dependence of χ is dominated by these corrections, leading to a large reduction of the effective spin fluctuation temperature τ_s^{-1} from its mean-field value, and that for $T\gg\tau_s^{-1}$, χ^{-1} is approximately a linear function of temperature. We expect these effects to be present in the Stoner-glass case as well, since strong local susceptibility enhancement is a basic ingredient of our model. Experimentally, this seems to be true: Coles' susceptibility curves for RhCo show something like Curie or Curie-Weiss above the spin-glass transition.[8]

We finally recall that the other term in Σ [Fig. 3(b) or Eq. (3.2)] has a sign opposite to that of the anharmonic term; thus it enhances the susceptibility. Hence, as we increase the randomness Δ and approach the spin-glass transition (by, say, varying the concentration of spin-fluctuation impurities at $T=0$), the spin-fluctuation (or Kondo) temperature will fall. This sort of effect is also observed experimentally.[9]

IV. DISCUSSION AND CONCLUSIONS

We now return to some questions about experimental studies of Stoner glasses. In particular, in what systems might we expect to observe a transition of the sort described here, and how would we

know that an itinerant model was better than a Heisenberg one?

As we noted in Sec. I, the important condition is that the spin-glass transition occur at a temperature lower than the Kondo temperature, so that it cannot be viewed as a freezing of ordinary local moments. If we restrict our attention to systems where both host and impurity are transition metals, so that the multi-impurity Wolff model picture we have used is a sensible one, the best candidates seem to be *Rh*Co and *Rh*Fe, both of which have been studied by Coles.[7-9] The former is a cleaner example, since its Kondo temperature is higher. Another possibility is *V*Fe. At first sight, alloys where the impurity is Ni seem attractive, but they always seem to go ferromagnetic beyond a critical Ni concentration, without any intervening spin-glass phase. Apparently, there is not enough random oscillation in χ^0_{ij} for these systems to produce a spin-glass transition. (But see note below.)

The present theory does not apply in detail to situations where the impurity is better described by an Anderson[25] than a Wolff model. In these cases (e.g., a transition-metal impurity in a noble-metal host), the spin fluctuations are associated with an "extra orbital," and, for high T_K, one may introduce anharmonic effects perturbationally in an Anderson-Friedel virtual level picture. Couplings between virtual levels on different sites will then lead to randomness in the same way that they do in the present description, and the description of the spin-glass transition can be expected to have a form quite similar to that given here. For low T_K, on the other hand, one can perform a Schrieffer-Wolff transformation[26] to a Kondo Hamiltonian. Now the host conduction electrons can both mediate an RKKY exchange interaction between local moments and quench those moments (below T_K). The spin-glass transition occurs at the impurity concentration where the former effect first wins out over the latter. It is the random counterpart of Donaich's Kondo lattice[27]; we can call it a Kondo glass. The theory of the Kondo glass remains to be worked out, but our understanding of the low-temperature properties of the single-impurity Kondo problem[15] suggests that the present Wolff-model picture is essentially correct. *Au*Co is one example of a Kondo glass which has been studied experimentally.[7] Some other examples of transition-metal impurities in noble-metal hosts have solubility problems[7] (*Cu*Fe and *Al*Mn), but it should be possible to find other examples. Another class of systems to examine would be alloys containing mixed-valent rare-earth impurities in transition metals. (In this case the mixed-valent situation is necessary for a high T_K.)

Whichever description is appropriate, a key experimental signature which sets itinerant glasses apart from simple RKKY-coupled Heisenberg systems is the fact that they require a finite concentration of impurities [in our model, this translates into a finite $\Delta(0)$] to drive the system to a spin-glass state at $T=0$. The usual Heisenberg models give a T_c which goes to zero only at zero concentration. The reason for the finite threshold here is that the magnetic ordering has to overcome band (kinetic) energies, or, in the case of the Kondo glass, the Kondo singlet binding energy.

A naive expectation might be that a Stoner glass was characterized by a relatively temperature-independent susceptibility above the spin-glass transition, rather than the Curie or Curie-Weiss law found for localized-spin systems. This is incorrect, since the fluctuation effects simulate the Curie or Curie-Weiss χ in the present description via the first term in Σ [Eq. (3.2)]. Any system where these fluctuation effects were negligible would presumably also not become a spin-glass, since the spin-glass terms in the free energy are of the same order.

Finally, a few words about "clustering": A popular *ad hoc* description[28] of many spin-glass materials invokes a picture of fairly large clusters of impurity moments coupled strongly together internally to account for the gross features of measurements (specific heat, resistivity, etc.) which are sensitive to properties on a wave-number scale of inverse interatomic spacings. The spin-glass transition is then seen as a freezing of the cluster moments. An extension of this idea has been used by Coles[7] for *Au*Co and *Rh*Fe. In it, one argues that a pair of impurities very close to each other will interact in such a way that the pair, viewed as a single entity, will have a lower Kondo temperature than a single impurity, that close triplets will have an even lower effective T_K, and so forth. Then, as one turns up the concentration, there will be more and more such cluster configurations present, with a concomitant lowering of the effective T_K for the alloy. In this picture (Fig. 5), spin-glass order only becomes possible after T_K has been driven to zero.

Sherrington[29] has formulated an approach to these clustering effects in the paramagnetic state which

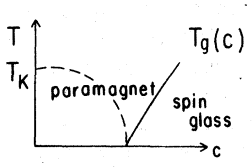

FIG. 5. Coles' picture of the phase diagram of a Kondo alloy, showing the hypothesized lowering of T_K to zero at the critical concentration for spin-glass formation.

is based on Anderson localization theory.[30] In it, a local cluster moment is associated with the magnetic instability of a localized solution of a random Landau-Ginzburg theory which is similar in some respects to the formulation of the problem given here. Although such an approach does not suffer from the shortcomings of Coles' *ad hoc* picture (it is possible to do a calculation in the Sherrington model), the approach has not yet been followed up to the point where its success or relevance can be judged. Furthermore, the connection with the spin-glass state is unclear, although presumably the transition is associated with the first extended eigensolution. In particular, there is no obvious order parameter in such a description.

It is not the intent of this discussion to argue that either of the above pictures is incorrect; rather, it is to point out that these clustering effects are implicit in the theory given here, at least insofar as they may be observed experimentally. As we noted at the end of the previous section, the second term in the self-energy (3.2) acts to enhance the susceptibility and thus to lower the Kondo temperature from its value in the (dilute) limit of zero random couplings between impurities, as observed experimentally and inferred from *ad hoc* arguments in the Coles picture. We do not need to resort to such arguments, as the observed concentration dependence of the effective T_K (inferred from susceptibility measurements) and the observed temperature dependence of χ fall out naturally from the theory.

Note added in proof. After this work was completed, spin-glass behavior was observed in *Ni*Cu alloys [C. J. Tranchita and H. Claus, Solid State Comm. 27, 583 (1978)].

ACKNOWLEDGMENT

It is a pleasure to acknowledge the hospitality of the Xerox Palo Alto Research Center, where this work was completed. This work was supported by the NSF under Grant No. DMR 77-12637 and through the MRL program at the University of Chicago.

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