

Saturation of Ruderman-Kittel-Kasuya-Yosida interaction damping in high-resistivity spin glasses

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(Received 5 August 1985)

Saturation of the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction damping at large resistivity or short mean free path in metallic alloys, as predicted in the Kaneyoshi model, is shown to account for the recently observed spin-glass freezing temperature, T_0 , in such systems as $AuFe$ quench-condensed films and ternary XY_cZ_x alloys, where $X = Au, Ag, \dots$, is a nonmagnetic metal host, $Y = Fe, Mn, \dots$, is the dilute magnetic species of concentration c , and $Z = Cu, \dots, Ti, Sb, \dots$, is a nonmagnetic impurity of concentration x . Some deeper aspects of the c dependence of the characteristic RKKY interaction energy scale are discussed, with emphasis on the necessary distinction between quenched and ergodic situations in the randomly dilute alloys. A consequent logarithmic correction to the c -scaling laws (at the marginal dimensionalities $d = p = 3$, where d is the electronic dimension of RKKY interaction varying as R^{-d} , and p is the space dimension of the magnetic structure), in the form of $T_0 \sim c(-0.577 - \ln c)^{1/2}$, is shown to be due to broken dilatation invariance, by finite atomic size. The finite mean free path in real systems also breaks this invariance by providing a length scale. However, at the damping saturation limit a pseudo- c -scaling $T_0 \sim c$ reappears, as was found in the amorphous spin-glass $LaAuGd$. This, and related predictions of the "typical environment" approach to the quenched-random-averaging problem agree remarkably well with the new data that have recently become available.

In a recent paper¹ the authors bypass an interesting opportunity to compare their experimental data with the theoretical predictions of the spin-glass freezing temperature, among which the estimate due to the present author^{2,3} has already been tested by several investigators.²⁻¹² Elsewhere,¹³ Vier and Schultz (VS) recognize the good agreement that has thus been obtained. Nevertheless, there is, apparently, not a full consensus about the "right" way to proceed. The controversial issue is one of alternative models, and is best decided on empirical grounds. Since the situation is really quite subtle and seems in need of clarification we shall briefly explain its essence. It will be shown that our theoretical estimate does, in fact, also provide a detailed account of the experimental results reported by VS.¹

Consider a randomly dilute configuration of spins that interact strongly at short range. It may be time dependent, as in a fluid, in which case we shall take ergodicity for granted, or static, as in a quenched solid. Spins that are particularly close to other spins experience a "strong environment." In the ergodic situation, all the spins have a strong environment part of the time. In the quenched situation part of the spins have a strong environment all the time.

If a decision to freeze is based on the rule that a majority must experience a total interaction strength above a given threshold, then there are cases in which the ergodic and quenched situations will give opposite decisions. All that is required is sufficient dilution to decrease the frequency of close pairs (by roster in the quenched situation, by time share in the ergodic) and sufficient strength at short range.

For example, for the sake of simplicity, suppose all of the interactions are zero except on neighboring lattice

sites, where it is X . Let the threshold be X_0 and let $c = N_s/N_a$ be the fraction of sites having a spin. The probability (by roster or time share) that such a site has a neighbor spin is qc , where q is the lattice coordination number. For $qc \ll 1$, one can disregard multiples. In the quenched situation, the majority, therefore, have a weak environment, and the decision is "No." If, however, $qc |X| > X_0$, then every spin in the ergodic situation at times experiences a strong environment which is sufficiently strong, compared to the frequency with which it occurs, for the decision to be "Yes."

This example resembles the situation prevalent in *real* spin-glass systems. One should note that if every site contains a spin, or if the interaction is constant and of infinite range, then the distinction vanishes, because all environments are equivalent. The latter cases are the ones considered in most current spin-glass theories, and in this respect do not alert the necessary caution in their application to the real systems. If the interactions are graduated with range each environment is unique. In the case of a fluid, we know how to evaluate the situation by the averaging of statistical mechanics, which rests on ergodicity. In the quenched situation it is the most prudent to assume that, without empirical confirmation, we do not, except in highly symmetric crystalline structures. If we have to decide between alternative suggestions in this matter it is an empirical question.

A range dependence appropriate to the Ruderman-Kittel-Kasuya-Yosida (RKKY) coupled spin-glasses is

$$\mathcal{J}(R_{ij}) \sim \pm \mathcal{J}_0 \left[\frac{r_0}{R_{ij}} \right]^3, \quad (1)$$

where R_{ij} is the distance between spins (i) and (j), and r_0

is an atomic radius. The sign is assumed randomly independent for each pair, in order to represent the effects of the oscillation in the true RKKY interaction. The local energy scale Δ_i for a given site (i) is the single sum over neighbor spins (not lattice points):³

$$\Delta_i^2 = \sum_{j (\neq i)} [\mathcal{J}(R_{ij})]^2. \quad (2)$$

Consider replacing the j sum in (2) by an integral over a spherically symmetric density. Three approaches are of interest.

(i) *Ergodic situation*: An "average environment" is presented to each spin. Its density is essentially c/r_0^3 extending down to the atomic radius r_0 , therefore, apart from the constants,

$$\Delta_i^2 \sim \mathcal{J}_0^2 \frac{3c}{r_0^3} \int_{r_0}^{\infty} dR R^2 \left(\frac{r_0}{R} \right)^6 = \mathcal{J}_0^2 c. \quad (3)$$

Hence,

$$\overline{\Delta}_i^{\text{ergodic}} \sim \mathcal{J}_0 \sqrt{c}. \quad (4)$$

This is at odds with the well-established concentration scaling laws,¹⁴ which have shown the energy scales in RKKY coupled real spin glasses to be nearly linear in c . Only in very exceptional circumstances do freezing temperatures approach the \sqrt{c} predicted in (4), and these are much more consistently explained otherwise.³

It was suggested¹⁵ using a cutoff at the nearest-neighbor distance ξ_i , which is a random variable depending on (i), and subsequently to average over its distribution (to be done in tandem, before the root is taken). This can be shown¹⁶ to be equivalent to constructing a "typical environment" density.¹⁷ It represents the essence of quenching, that a given spin never experiences a closer encounter than a definite nearest neighbor. We have shown how this may affect freezing decisions.

(ii) *Quenched-uniform situation*: Assume all ξ_i are equal to the average nearest-neighbor distance $\langle \xi \rangle = (4\pi r_0^3/3c)^{1/3}$ as would be appropriate with a spin-atom superlattice. Then,

$$\Delta_i^2 \sim \mathcal{J}_0^2 \frac{3c}{r_0^3} \int_{\langle \xi \rangle}^{\infty} dR R^2 \left(\frac{r_0}{R} \right)^6 \propto \mathcal{J}_0^2 c^2, \quad (5)$$

and

$$\overline{\Delta}_i^{\text{quenched uniform}} \sim \mathcal{J}_0 c. \quad (6)$$

This result does agree with the scaling laws, apart from the small corrections to be discussed below.

(iii) *Quenched-random situation*: Averaging over the distribution of ξ_i appropriate to the random configuration gives close pairs a more proper representation, and a logarithmic correction to c scaling,^{12,15}

$$\overline{\Delta}_i^{\text{quenched random}} \sim c \sqrt{-\gamma - \ln c} \mathcal{J}_0 \quad (7)$$

(γ represents Euler's constant) in good agreement with quite extensive sets of data.^{3,10} The case of $d=3$ with the R^{-3} range dependence is marginal in this sense. The phenomenon is due to broken dilatation invariance:

$\overline{\Delta}_i^{\text{quenched random}}$ does not exist in the limit of point atoms.¹⁶ If atoms have a finite size, the r_0 here, then it introduces a length scale. Any such inherent length, atomic size, or mean free path, or RKKY short-range modification, breaks dilatation invariance. In the present case, pairs closer than $2r_0$ are missing, compared to what would occur in a random distribution of points. This makes a big difference because the RKKY is strong at short range. It causes the root factor in (7), which is absent from (6) because under (ii) there are no close pairs anyway, as illustrated in Fig. 1. The c dependences of (4), (6), and (7) differ because their hypotheses about the environment of a given spin differ. Each approach attempts to estimate what continuous neighbor density might be best assumed to be equivalent for all spins under the given circumstances.

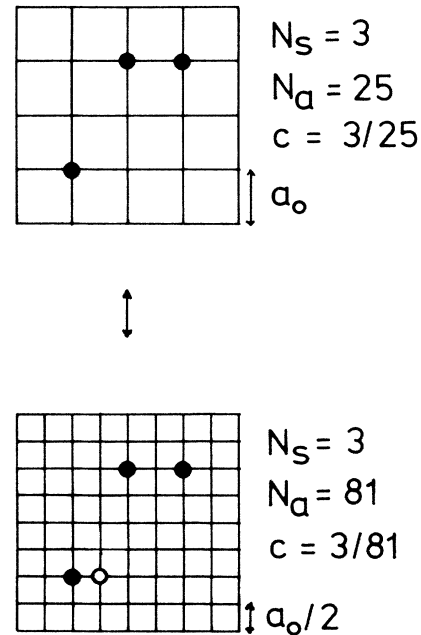


FIG. 1. Dilatation transformation changing lattice spacing and concentration c in conjunction, such as to keep fixed the invariant space density. The actual lattice spacing is related to the finite atomic size. The configuration of \bullet atoms misses some close pairs (shown as \bullet - \circ) which would be possible on the finer lattice, and both miss some closer ones that would occur with point atoms in a continuous space. This will break dilatation invariance in all of those quantities, such as our present quenched-random $k_B T_0$, in which close pairs contribute significantly at the $c \rightarrow 0$ limit. This is marginally the case with the form (1) of the RKKY interaction because of its strength at short range. Evidently, the quenched uniform situation has no similar effect, since the atoms are never closer than the superlattice spacing. A distribution of this sort, such as the \bullet atoms exclusively, is therefore dilatation invariant and gives rise to the pure c -scaling law.

There are two further possibilities one may consider:

(iv) The first moment of Δ_i is

$$\begin{aligned} \mu_1 &= \lim_{N_s \rightarrow \infty} \left[\frac{1}{N_s} \sum_i \Delta_i \right] \\ &= \lim_{N_s \rightarrow \infty} \left[\frac{1}{N_s} \sum_i \left[\sum_{j (\neq i)} [\mathcal{J}(R_{ij})]^2 \right]^{1/2} \right]. \end{aligned} \quad (8)$$

Since the distribution of Δ_i in the quenched-random situation is anomalous, due to close pairs and the infinite range of the interaction, a direct numerical evaluation is demanding.¹⁸ A careful evaluation with N_s large enough to get sufficient accuracy of μ_1 at low c to distinguish the c dependences of (4), (6), and (7) from each other showed that μ_1 agrees with (7) (cf. Fig. 2).

(v) The second moment of Δ_i is

$$\begin{aligned} \mu_2 &= \lim_{N_s \rightarrow \infty} \left[\frac{1}{N_s} \sum_i \Delta_i^2 \right] \\ &= \lim_{N_s \rightarrow \infty} \left[\frac{1}{N_s} \sum_{i \neq j} [\mathcal{J}(R_{ij})]^2 \right]. \end{aligned} \quad (9)$$

It involves the double sum $\sum_{i \neq j}$ and therefore implies an averaging over the center spin (i) of our previous considerations. Thereby it becomes equivalent to an "average environment," which is the same as the one used in the ergodic situation (i). It is difficult to see how the locally relevant quantity could be

$$\Delta = (\mu_2)^{1/2} \sim \sqrt{c} \mathcal{J}_0, \quad (10)$$

rather than μ_1 itself. Perhaps it may acquire a global significance by way of the replica method¹⁹ (both the Δ_i and Δ are displayed in Ref. 19, but the distinction appears to have been overseen in some subsequent works). Even if one maintains (10) for such reasons, the decision between the different models remains an empirical question.

With proper attention to details, in the quenched-random situation (iii) with a "typical environment" density one finds^{2,3,17}

$$k_B T_0 = A [3cT(z, \xi)]^{1/2}. \quad (11)$$

Here, A is essentially the RKKY strength \mathcal{J}_0 , z is the damping strength (see below), $\xi = c/(1-c)$, and

$$T(z, \xi) = \int_1^\infty dx \frac{1}{x^4} e^{-zx} (1 - e^{\xi(1-x^3)}). \quad (12)$$

The Euler-McLaurin expansion is

$$\begin{aligned} T(z, \xi) &= \frac{\xi}{4m^2} e^{-z} \\ &+ \frac{1}{m} \sum_{k=1}^{m-1} \left[\frac{k}{m} \right]^2 e^{-zm/k} (1 - e^{\xi[1-(m/k)^3]}), \end{aligned} \quad (13)$$

and in some ranges of z and c is accurate already at $m=10$.

In the Kaneyoshi model²⁰ one has asymptotically²¹ the

RKKY interaction²²

$$\mathcal{J} \sim -\mathcal{J}_0 \frac{\cos(x + \varphi)}{x^3} e^{-x \tan \varphi}, \quad (14)$$

where

$$x = 2k'_F R, \quad \varphi = \frac{1}{2} \tan^{-1} \delta. \quad (15)$$

This contains three parameters: \mathcal{J}_0 , k'_F , and δ (the linewidth), which, at present, we have no choice but to

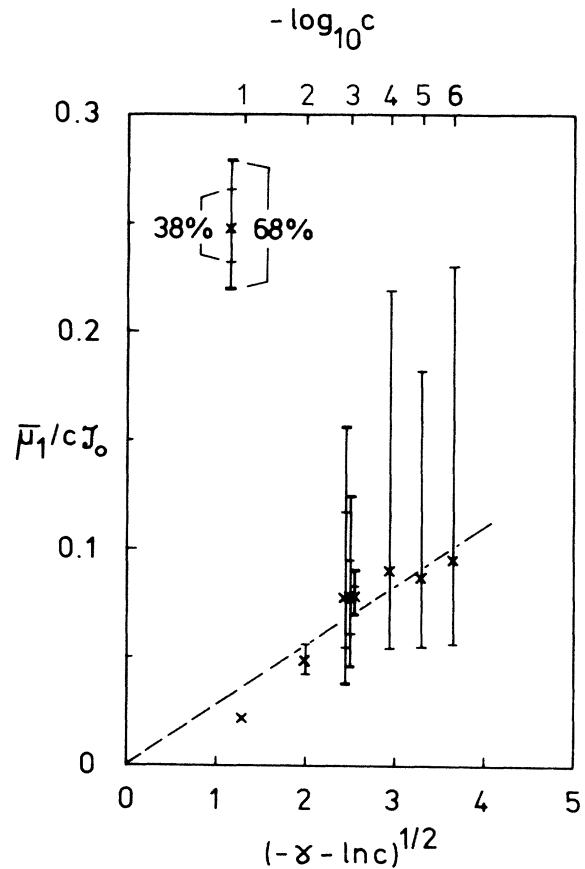


FIG. 2. Mean first moment $\bar{\mu}_1$, according to Ref. 18, of spin-glass RKKY interaction energy (8) over randomly constructed systems with finite N_s . In samples with fixed fcc lattice volume N_s is distributed as appropriate to the given global concentration c (i.e., Poisson for low c) with mean $\bar{N}_s = 40$. There are typically ~ 400 samples at each c . The development with size \bar{N}_s is indicated for $c = 10^{-3}$, including systems of $\bar{N}_s = 100$ (100 samples) and $\bar{N}_s = 500$ (14 samples), which become quite consuming of time and storage. Special care was taken to compensate for boundary effects by embedding each sample volume in a shell containing 26 times as many spins (also in average over the N_s distribution). Displayed is $\bar{\mu}_1 / c \mathcal{J}_0$ vs $\sqrt{-\gamma - \ln c}$ to emphasize the c -scaling correction [low c form according to (7)], as well as intervals containing the central 38.2% of the sampled values (together with 68.2% intervals at $c = 10^{-3}$). The trend is $\sim \bar{N}_s^{-3/4}$. The accuracy achieved here, despite the anomalous distribution of μ_1 at low c , seems sufficient to establish in (8) the c -scaling correction in the form predicted in (7) with the "typical environment" construction.

leave adjustable to the experiments.²³ \mathcal{F}_0 and k'_F contribute to A , while it follows from (14) and (15) that

$$z = \frac{z_0}{\delta} [(1 + \delta^2)^{1/2} - 1], \quad (16)$$

where z_0 contains k'_F (and r_0).

Previous investigations²⁻¹² were done under the simplifying assumption that $\delta \ll 1$, in which case $z = \frac{1}{2}z_0\delta$, and one could let $\delta \propto \rho$, the resistivity. The present experiments¹ indicate that in the ternary alloys δ may reach values large enough to actually bring about the saturation of the damping, as predicted in (14), i.e.,

$$z \rightarrow z_0 \text{ for } \delta \rightarrow \infty. \quad (17)$$

We continue to let $\delta \propto \rho$, since we have no evidence to the contrary, but the constant of proportionality should be reserved as an adjustable parameter

$$\delta = \rho / \rho_0. \quad (18)$$

Free-electron estimates are not meaningful in systems this complicated. We have then three parameters: A , z_0 , and ρ_0 with which to reproduce the entire experimentally observed ρ and c dependences.

Inspection of the data of VS for $\text{AgMn}_{0.026}\text{Sb}_x$, which is the most extensive set they present, reveals a saturation at large ρ already without assistance from *ad hoc* fitting formulas. Without attempting a fully optimized fit the values

$$z_0 = 0.5, \quad \rho_0 = 6.4 \mu\Omega \text{ cm}, \quad (19)$$

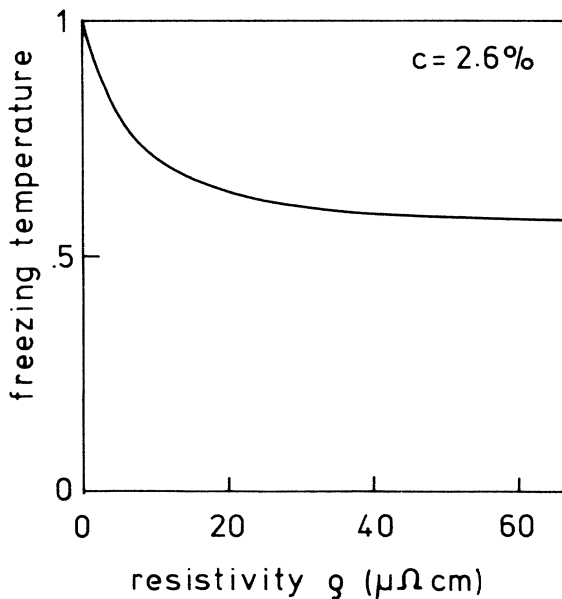


FIG. 3. Resistivity dependence of spin-glass freezing temperature with z_0 and ρ_0 as in (19) adjusted to fit approximately the AgMn_xSb_x ($c = 0.026$) data presented in VS (Fig. 1). Saturation of the RKKY damping implies a finite value at infinite resistivity.

produce the ρ dependence shown in Fig. 3, and the c dependences shown in Fig. 4. These theoretical estimates of $k_B T_0/A$ must be compared with Figs. 1 and 2 in VS, respectively. In Fig. 4 we show $\rho = 0$ and $\rho = \infty$ limits, as well as the intermediate curve corresponding to the binary system AgMn_c , where the resistivity is²⁴

$$\rho_{\text{AgMn}} \approx 154c \mu\Omega \text{ cm}. \quad (20)$$

We believe it is fair to conclude that the agreement is striking. It even extends to the slight curvatures in the c dependences, and this is not something which can be arranged by adjusting the parameters since they are already determined at $c = 0.026$. For example, at $\rho = \infty$ in $1\% \leq c \leq 10\%$ we find $T_0 \sim c^{0.93}$, whereas with their extrapolation formula VS report $T_g(\infty) \sim c^{0.91}$. Since the saturation of T_0 in the present estimate is not exponential, but slower, the values at $\rho = \infty$ are a little smaller than what their exponential formula might suggest, so it is to be expected that their exponent may be slightly less. But, in any case the c^α dependence is only approximate, and the exponent depends somewhat on the interval of c in which it is taken. The phenomenon that T_0 becomes nearly linear in c at strong damping was first noticed by Poon and Durand,²⁵ and here receives an explanation. (See Fig. 4.)

Compared to the earlier analysis we have now introduced one more parameter in order to extend the test of our estimate to the range where the damping is predicted to saturate. For small δ they coalesce into the self-damping parameter

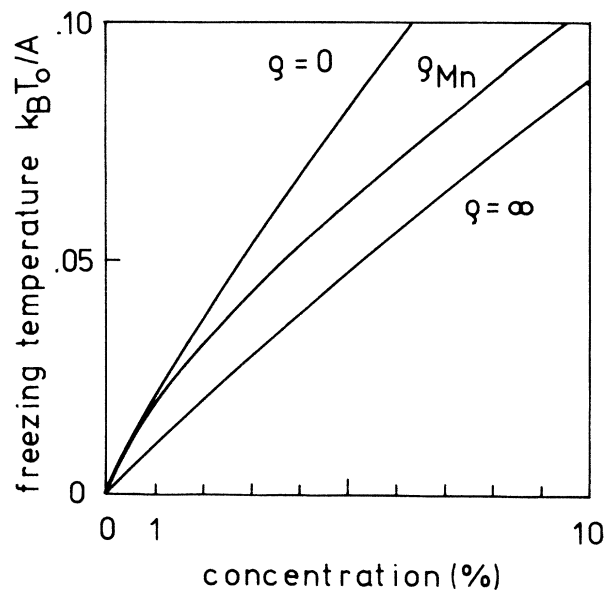


FIG. 4. Concentration dependence of the zero resistivity limit of the spin-glass freezing temperature, the infinite resistivity limit, and the estimate for the AgMn_c resistivity (20) with self-damping of the RKKY interaction by the Mn impurities. Same parameter values as in Fig. 3 to reproduce approximately the data in Fig. 2 of VS for AgMn_xSb_x .

$$r = \frac{z}{c} = \frac{1}{2} z_0 \rho_{\text{AgMn}} / \rho_0 c \simeq 6. \quad (21)$$

In the absence of unequivocal evidence for the saturation in the earlier data this simplification was continued to high ρ , without introducing the quantity δ . With hindsight, one can see the hints of the saturation also there,^{6,26,27} a point which was noted but left for future investigation.³

In conclusion, we have pointed out the distinction be-

tween ergodic and quenched environments which is necessary when estimating the RKKY interaction strength in dilute alloys. The source of a marginal correction to c scaling has been identified in this context. The model using the "typical environment" density in the quenched random situation was compared with the experiment of VS and found entirely adequate in an extended range, demonstrating the damping saturation effect predicted in the Kaneyoshi model.

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¹⁷A direct way of showing this is the following: Select at random a definite spin (i) as origin, and arrange the lattice sites $k=1, 2, \dots$, in a sequence of never-decreasing distances. Remove the spin (i) from the origin at $k=1$. The probability of having no spin on the first N lattice sites is $(1-c)^{N-1}$. The typical sum, $\sum_{j(\neq i)}$ over a quenched neighbor configuration can be represented by a weighted sum over all lattice sites: $\sum_k p_k \dots$, if a lacuna is thus maintained around the origin: $p_k = c[1 - (1-c)^{k-1}]$ equals the probability of a neighbor spin (j) on site k , conditional on the site k not being in the lacuna! In the ergodic situation the lacuna gets filled in the course of time, except the origin $k=1$, of course, by spins (j) flowing in from infinity, as well as by the itineracy of the spin (i) we consider. The three-dimensional "typical neighbor-density" is then $\omega_{\text{quenched}} = \omega_0(1 - e^{-\zeta(1 - (R/r_0)^3})$, in $R \geq r_0$, and 0 in $R \leq r_0$, where $\omega_0 = 3c/4\pi r_0^3$. To ensure correct normalization, $\zeta = c/(1-c)$, i.e., the lacuna contains exactly one spin: $4\pi \int_0^\infty dRR^2(\omega_0 - \omega_{\text{quenched}}) = 1$, namely, the excluded spin (i). There is an adjustment of ζ from the lattice value $-\ln(1-c)$, in order to have a correct continuous density all the way up to $c=1$. The subtlety of the problem is due to the fact that, in the quenched local situation there is no

natural averaging process, nor any repeated sampling on which one could rely if one attempted to use traditional statistical methods. The absence of an implicit ergodic justification of such methods does not seem to have been appreciated in the present context. This principle that, in quenched-random systems "no real atom is an average atom, nor is an experiment ever done on an ensemble of samples" [P.W. Anderson, Rev. Mod. Phys. **50**, 191 (1978)] was ignored in the calculation of W. Kinzel and K. H. Fischer [J. Phys. F **7**, 2163 (1977)], where they generate the ergodic average environment by using a cut off at the atomic radius r_0 . The latter is also what was endorsed in the paper by VS (Ref. 1). In our way of estimating T_0 (Ref. 2), an effort to deal with the c -scaling phenomena while awaiting the complete and exact solution of the RKKY spin-glass problem we are still looking forward to, we observed Anderson's principle by using the nearest-neighbor distance distribution of Riess and Ron (Ref. 15), and we have now demonstrated how it corresponds to a "typical environment" appropriate, as it seems, to the quenched-random situation. Evidently, what is "typical" is not necessarily the same as the "average." The recent observation of h/e Aharonov-Bohm magnetoresistance oscillations in small normal-metal rings of structurally disordered Au [R. A. Webb, S. Washburn, C. P. Umbach, and R. B. Laibowitz, Phys. Rev. Lett. **54**, 2696 (1985)] testifies to the absence of self-averaging in quenched randomness on the local scale. The present estimate of T_0 has been extended to one- and two-dimensional electronic systems, and layered spin configurations (see Ref. 16).

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²¹U. Larsen, J. Phys. F **15**, 101 (1985).

²²The Kaneyoshi model (Refs. 20 and 21) assumes independent electrons, but wave-functions appropriate to a (highly) disordered structure. Since they are not known exactly, it represents them, for simplicity, by means of a Breit-Wigner spectral density of width $\sim \delta$ on the plane-wave free-electron representation. All of the results one obtains from it are exact. As such, it is a model in its own right, and appears to be a very realistic one. Thus, there are both physical and mathematical advantages to defining models directly in terms of Green functions, rather than in terms of insoluble Hamiltonians that tend to seriously complicate the investigation of interesting situations from the very beginning. As a matter of principle, one can have reservations about the explanatory power that usually motivates the Hamiltonian formulation, if such a model is not soluble in practice. Anyway it is valuable to construct a physically simple and well-defined intermediate

stage which can be handled in a mathematically exact way. With respect to the relation to other models, it was noted (Ref. 20) that, using the coherent-potential approximation [B. Velický, *Phys. Rev.* **184**, 614 (1969)] on free electrons scattered by random impurities one obtains a similar, but more complicated result. Although there are good reasons to presume that if such a model could be treated exactly it would give essentially equivalent results, the RKKY interaction obtained directly, and exactly, from the Kaneyoshi model does not depend on the validity of such arguments. This nicely sidesteps the problem of having to deal with averaged Green functions. In particular, the complication noted by de Gennes [J. Phys. Rad. **23**, 630 (1962)] in the weak scattering perturbative approximation (i.e., \overline{GG} not equal to $\overline{G} \overline{G}$), although perhaps eliminated by the CPA method, does not occur at all here, because there are no impurities and no free electrons in the first place for which a scattering must be averaged over. This problem only occurs if one does not have the correct wave functions to start with, but if one does, the electrons are independent, which means $G_{\text{exact}} G_{\text{exact}}$. Thus, it is a clear advantage to pose the model in terms of such postulated G_{exact} , and this lends itself to a direct experimental test, without our having to worry over the validity of difficult approximations. Curiously enough, the very recent results of Webb *et al.* mentioned in Ref. 17 suggest another application of the quenched-randomness principles emphasized there, according to which one is tempted to regard G_{exact} as a "typical" electronic Green function, appropriate to the local situation between the RKKY interacting spins. The usual averaging over samples with different random impurity configurations, i.e.,

to produce \overline{G} and \overline{GG} , gets to look extremely suspect. Anyway, there seems to be a preference for G_{exact} in other contemporary work as well [cf. L. Coffey, K. Levin, and K. A. Muttalib, *Phys. Rev. B* **32**, 4382 (1985)].

²³When $\rho \neq 0$, the physically relevant parameter is k'_F , not k_F , since $1/k'_F$ is the actual RKKY period, and must be essentially independent of ρ and c , for obvious physical reasons. The relation is (Ref. 21) $k'_F = k_F(1 + \delta^2)^{1/4} \cos(\frac{1}{2} \tan^{-1} \delta)$, and k_F must be assumed to depend on δ in such a way as to perform the cancelling. Integrating the exact $\mathcal{F}(R)$ over the volume V , using Eq. (11) of Ref. 21, gives the static susceptibility [c.f. U. Larsen, *Phys. Lett.* **85A**, 471 (1981) and *J. Math. Phys.* **21**, 1925 (1980)]: $\chi_0 = V \mu_B^2 m k'_F / 3\pi^2$. Thus keeping k'_F constant implies χ_0 constant, which makes good sense as a first approach. It can be arranged simply by adjusting the electron density according to $N_e/V = (k'_F)^3 / 3\pi^2 \cos^3(\frac{1}{2} \tan^{-1} \delta) \times (1 + \delta^2)^{3/4}$. These are all elementary considerations.

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