Superconductivity and magnetic order in ferromagnets and spin-glasses

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Taking into account the effect of both *time* and spatial-dependent correlations of the magnetic impurities on pair breaking, we derive an expression for the superconducting transition temperature. Ferromagnets and spin-glasses are discussed. We find, in agreement with previous workers, that spatial correlations which are ferromagnetic have a tendency to enhance the pair breaking effect, while spatial correlations for an antiferromagnet or spin-glass reduce the pair breaking. However, we find that the introduction of dynamics (time correlations) always reduces the pair breaking for all types of magnetic ordering. In particular, for the spin-glass case we found that the time correlations give rise to a "re-entrant" phenomena.

I. INTRODUCTION

The interest in systems which exhibit both superconductivity and magnetic ordering has intensified as a result of recent work in superconducting ternary alloys.[1-3] The destruction of superconductivity at the onset of ferromagnetic order and the coexistence of superconductivity and antiferromagnetic order found experimentally has raised many interesting questions. There are now several systems which show the coexistence of magnetism and superconductivity.[4] This wealth of new data has inspired several recent theories[5] which propose to explain the above phenomena. Theoretically, we can distinguish two regions of interest. One is the region above the concentration-dependent magnetic critical temperature T_M. Far above T_M it is valid to assume that the magnetic impurities are noninteracting. Abrikosov and Gorkov[6] (AG) have shown that the superconducting transition temperature T_c decreases linearly as a function of the concentration x at small concentrations of magnetic impurities and vanishes at a critical impurity concentration x_{AG}. However, near the magnetic transition, the assumption of noninteracting magnetic impurities breaks down. Modifications of the AG theory due to spatial spin correlations of both the ferro- and antiferromagnetic type have been considered. Toxen *et al.*[7] generalized the AG theory to include spin correlations of antiferromagnetic type. Rainer,[8] Entel and Klose,[9] Sakurai,[10] and Machida and Youngner[11] have also developed this idea to discuss the effect of spin correlations on the superconducting properties. The second region of interest is $T < T_M$ where the interactions between magnetic impurities are strong and spontaneous magnetic ordering sets in. This case is more difficult to handle because there exists an additional exchange field acting on the conduction-electron spins,[12] and the conduction electrons are scattered by the excitation of the spin system.[13] These additional mechanisms lead to pair breaking in the superconducting state. For ferromagnetic ordering, it is difficult to modify the AG theory in this region to account for the magnetic interactions because of these additional mechanisms.[12,13] However, for an antiferromagnet or spin-glass, where the average internal magnetic fields are small, the motion of the conduction electrons should not be affected.

In the previous extensions[7-11] of the AG theory to include spin fluctuations, only elastic scattering of the conduction electrons by the local magnetic moments was considered. Several authors have discussed the influence of spatial correlations on the pair breaking. Here we have included the time correlations as well. Dynamics have been taken into account by Maekawa and Tachiki,[14] Entel and Klose,[9] and Machida and Youngner[11] but only in the limit when the frequency of spin motion is large compared with the transition temperature. This approximation leads to a result for T_c similar to that obtained by Berk and Schrieffer[15] for nearly ferromagnetic alloys. To the best of our knowledge, there has been no development which includes the dynamics when the frequency is comparable to the transition temperature. In this paper, we discuss the influence of *time-dependent* and spatial-dependent correlations on the pair breaking. For the ferromagnetic case, we discuss only the case $T > T_M$, while we consider all temperatures for a spin-glass. We find, in agreement with previous workers that spatial correlations of the ferromagnetic type have the tendency to enhance the pair breaking effect, while the spatial correlations in an antiferromagnet or spin-glass reduce the pair breaking. However we find that the introduction of dynamics (time correlations)

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reduces the pair breaking for all types of magnetic ordering.

In Sec. II, we present a method for incorporating the time correlations (dynamics) into the spin-flip scattering time τ. This gives an approximate way of introducing the effects of dynamics on the pair breaking. While we realize that it is only an approximation, it should be considered a first step in a complete solution of the problem. We also describe the results for the ferromagnetic case in this section. In Sec. III, we discuss the spin-glass case. Finally in Sec. IV we present some conclusions and further comparison of our results with experiments.

II. SPATIAL AND TIME CORRELATIONS-FERROMAGNETIC CASE

We assume that the system is described by the Hamiltonian

$$H = H_{\rm BCS} - \frac{I}{2} \sum_{i,k,k',\sigma,\sigma'} e^{i(\vec{k}-\vec{k}')\cdot\vec{R}_i} [\vec{S}_i \cdot (C^\dagger_{k\sigma} \vec{\sigma}_{\sigma\sigma'} C_{k'\sigma'})] - \frac{1}{2} \sum_{ij} J_{ij} \vec{S}_i \cdot \vec{S}_j \ , \tag{2.1}$$

where $H_{\rm BCS}$ is the BCS Hamiltonian for the system without magnetic impurities, $\vec{\sigma} \equiv (\sigma^x, \sigma^y, \sigma^z)$ are the three Pauli matrices and S_i is the magnetic spin located at R_i, and I is the s-f exchange term. The second term describes the interactions between the electrons and the localized spins of the rare-earth or transition-metal ions while the third term describes the interactions between the localized spins. This coupling leads to a magnetic ordering at a temperature T_M. For simplicity we assume that the magnetic state is determined only by the third term. We assume a simple relation between T_M and the concentration of magnetic impurities x to obtain the phase diagrams shown below.

Abrikosov and Gorkov[6] first showed that in the Born approximation, T_c is given by

$$\ln \frac{T_c}{T_{c0}} = \psi(\tfrac{1}{2}) - \psi\left(\frac{1}{2} + \frac{1}{2\pi T_c \tau}\right) \ , \tag{2.2}$$

where T_{c0} is the transition temperature of the system without magnetic impurities, $\psi(z)$ is the digamma function, and τ is the spin-flip scattering time. In the absence of spin correlations, τ in the Born approximation is given by

$$1/\tau_{\rm AG} = 2\pi N(E_F) x (\tfrac{1}{2} I)^2 S(S+1) \ , \tag{2.3}$$

where $N(E_F)$ is the density of states at the Fermi energy and x is the concentration of magnetic impurities. This result was obtained by AG for noninteracting impurities. When spatial and time correlations are taken into account, the result for τ can be generalized within the Born approximation. Following the Appendix of Greene and Kohn[16] and Grest *et al.*[17] we have

$$\frac{1}{\tau} = \frac{1}{\tau_{\rm AG}} \frac{1}{2k_F^2 S(S+1)} \times \int^{2k_F} q dq \int_{-\infty}^{+\infty} d\omega g(q,\omega) \frac{\beta\omega}{e^{\beta\omega}-1} \ , \tag{2.4}$$

where

$$g(q,\omega) = S(q,\omega) - S_{\rm Bragg}(q,\omega) \tag{2.5}$$

and

$$S(q,\omega) = \frac{1}{2\pi N} \int_{-\infty}^{+\infty} dt e^{-i\omega t} \sum_{ij} \langle S_i(t) S_j(0) \rangle e^{i\vec{q}\cdot\vec{R}_{ij}} \ , \tag{2.6}$$

$$S_{\rm Bragg}(q,\omega) = \frac{1}{2\pi N} \int_{-\infty}^{+\infty} dt e^{-i\omega t} \sum_{ij} \langle S_i \rangle \langle S_j \rangle e^{i\vec{q}\cdot\vec{R}_{ij}} \ . \tag{2.7}$$

This result for $1/\tau$ is equal to $1/\tau_{\rm AG}$ in the limit $S(q,\omega) \sim \delta(\omega)\delta(q)$. In performing the q integral in Eq. (2.4) we introduce a cutoff q_c at the small momentum transfer in order to take into account the lifetime effect of one electron due to impurities and defects on critical scattering. If the ω dependence of $g(q,\omega)$ is purely elastic, then we obtain the expected behavior of $1/\tau$ [see for example, Eq. (5), Ref. 7] in which only spatial spin correlations are taken into account. We express $g(q,\omega)$ in a more convenient form using[18]

$$g(q,\omega) = \frac{k_B T}{g^2 \mu_B^2} \chi(q) \frac{\beta\omega}{e^{\beta\omega}-1} F(q,\omega) \ , \tag{2.8}$$

where $\chi(q)$ is the q-dependent susceptibility and $F(q,\omega)$ is the spectral weight function. Since $F(q,\omega)$ is not known in general we will use the hydrodynamic form

$$F(q,\omega) = \frac{1}{\pi} \frac{Dq^2}{(Dq^2)^2 + \omega^2} \ , \tag{2.9}$$

where D is the spin-diffusion constant. While this equation is not valid for large ω and q we find that the major contribution to $1/\tau$ comes from small ω and expect this form for $F(q,\omega)$ to give reasonable results. It also agrees with the form for $F(q,\omega)$ found in the high temperature, low-q limit both from microscopic theories[19] and experimentally.[20]

Our approximation is to put the q and ω dependence into the spin-flip scattering time τ, assuming that Eq. (2.2) is valid. We can then derive the phase diagrams for the cases of interest. This manner of introducing the dynamics to the problem can be con-

sidered as a first step in a complete calculation and can be used as both a comparison and first indication. A complete solution of the problem involves generalizing the results developed by Maki[21] to include inelastic scattering. This gives rise to a set of coupled integral equations. Nass *et al.*[22] have solved these equations for a superconducting spin-glass at low temperatures. Results for the dependence of T_c on the concentration of magnetic impurities have not been calculated.

Using Eqs. (2.2) to (2.9), we can derive the phase diagram for the ferromagnetic case. From Eqs. (2.4), (2.8), and (2.9) we can calculate $1/\tau$ versus temperature. For the ferromagnetic case in Eq. (2.8) we assume the Orstein-Zernike form for $\chi(q)$:

$$\chi(q) = \frac{S(S+1)}{T - T_M + a^2 q^2} \quad . \tag{2.10}$$

We also assume that the spin-diffusion constant D takes its scaling form[19]

$$D = D_0 (T/T_M - 1 + a^2 q^2)^{1/4} \quad . \tag{2.11}$$

We tried several different forms for D, including D constant and find that the results for $1/\tau$ and the phase diagrams are essentially insensitive to the T and q dependence of D. In our calculations we have normalized the energies by kT_{c0} and the wave vectors with $q_{BZ} = 2k_F$; where q_{BZ} is the radius of the Brillouin zone and k_F is the Fermi wave vector. We take $q_c = q_{BZ}/50$. Then the only parameters remaining are x_{AG}, $B = D_0 q_{BZ}^2 / kT_{c0}$, and $C = a^2 q_{BZ}^2 / T_{c0}$. The parameter x_{AG} is the concentration for which T_c/T_{c0} goes to zero in the AG theory. If $B \to 0$ for $C \neq 0$ then we reproduce the previous results which treat the spatial correlations.[7–11]

For a ferromagnet, we have calculated $1/\tau$ including only spatial correlations ($B \to 0$) and with time dependent correlations ($B \neq 0$). The results are plotted in Fig. 1. For all the cases considered, we obtain the known AG elastic limit for very high temperatures. As B is increased, time correlations become more important and $1/\tau$ decreases, showing that the introduction of dynamical effects reduces the pair breaking. This will usually increase the region of coexistence if any exist. We do not plot $1/\tau$ below T_M, since we do not know a form for $\chi(q)$ and $F(q,\omega)$ for $T < T_M$. Below T_M in the ferromagnetic case, there are large internal magnetic fields which arise from the spontaneous magnetization that makes any simple modification of the AG theory incorrect. Using our result, Eq. (2.4), for $1/\tau$ and assuming a simple relation between T_M and x, we can calculate phase diagram for superconductivity and magnetic ordering. We choose a relation between T_M and x which corresponds to the experimental result for $(Lu_{1-x}Ho_x)Rh_4B_4$.[23] We then varied the two parameters B and C in an attempt to reproduce qualitatively

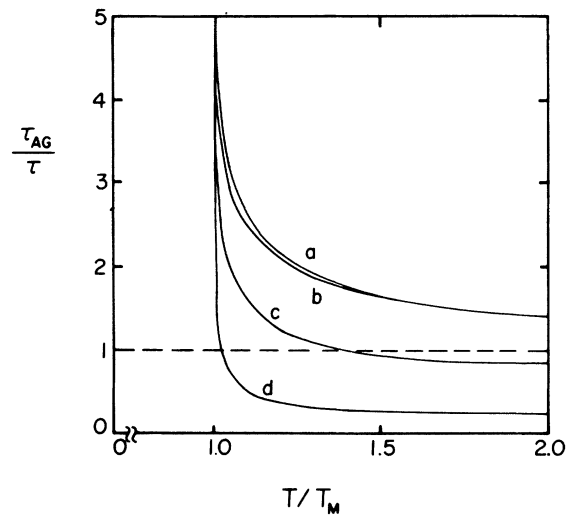

FIG. 1. Temperature dependence of the inverse spin-flip scattering time $1/\tau$ normalized with $1/\tau_{AG}$. The dashed line at $\tau_{AG}/\tau = 1$ is the AG result. Curves a to d are for $B = 0$, 0.1, 10, and 100. In all cases $C = 10$. For large T we obtain the AG result.

the experimental phase diagram for this system. The results are shown in Figs. 2 and 3. In Fig. 2, we take $C = 10$ and vary the value of B. Since we are interested in plotting T_c/T_{c0} versus x and not x/x_{AG}, we have to choose x_{AG}. To fit the experimental phase diagram for $(Lu_{1-x}Ho_x)Rh_4B_4$, we used $T_{c0} = 11.5$ K and $x_{AG} = 0.324$.[24] Note that as B increases, T_c increases. The T_c curve always terminates at the T_M curve, because of the introduction

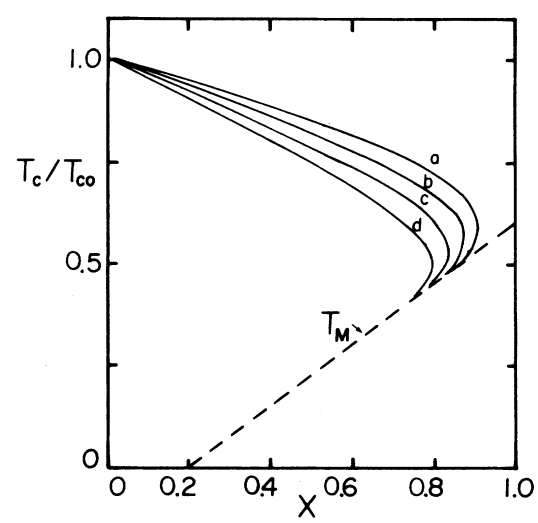

FIG. 2. Phase diagram for $(Lu_{1-x}Ho_x)Rh_4B_4$. Curves a to d show the superconducting transition temperature for $B = 10$, 5, 2, and 0. In all cases $C = 10$, $T_{c0} = 11.5$ K, and $x_{AG} = 0.324$.

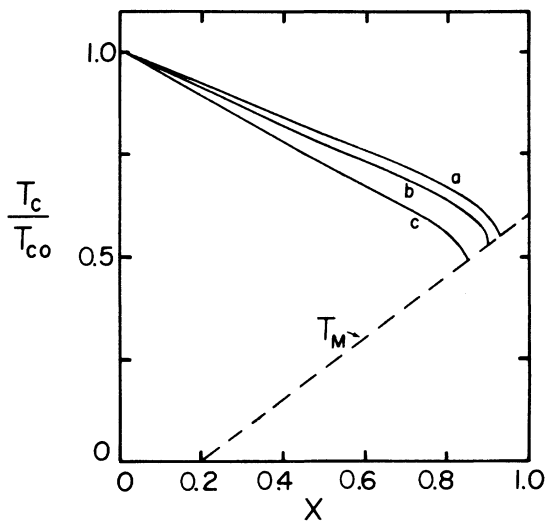

FIG. 3. Phase diagram for $(Lu_{1-x}Ho_x)Rh_4B_4$. Curves a to c show the superconducting transition temperature for $B=10$, 5, and 0. In all cases $C=100$, $T_{c0}=11.5$ K, and $x_{AG}=0.05$.

of the lower cutoff q_c for the q integral in Eq. (2.4) which takes into account the lifetime effect of one electron due to defect and impurity scattering.[10] Without this lower cutoff the curve for T_c continues down to $T=0$, following very close to the T_M curve. Then there would always be a smaller paramagnetic region between the superconductor and ferromagnetic states.[11] Since this is not seen experimentally we have introduced a long-wavelength cutoff in the q integration which eliminates this region. From Fig. 2 we see that all the T_c curves have a downward bend before crossing the T_M curve, which is not seen experimentally. In the experimental data (Ref. 23) we have that T_c decreases from its T_{c0} value almost linear with x without any bending down to the T_M curve. We obtain better argument with the experimental results if we decrease x_{AG}, which corresponds to a stronger s-f exchange interaction I. In Fig. 3, we take $C=100$ and $x_{AG}=0.05$ $[N(E_F)I^2/4T_{c0}=1.64]$. As seen from Fig. 3, the curves for T_c increase as we increase B. Note that we have not obtained results for $T_c < T_M$. If we neglect the large internal field, then the spontaneous magnetic ordering would reduce the pair breaking and enhance superconductivity. However, this large internal magnetic field suppresses superconductivity and will probably produce a first-order phase transition along or just below the T_M line. Within this model we cannot say how large the coexistence region is if it exists at all. We present the fit of the experimental phase diagram data for $(Lu_{1-x}Ho_x)Rh_4B_4$ as an example of all the pseudoternary compounds.[4] Qualitatively we obtain the same results for other compounds, like $(Y_{1-x}Gd_x)Rh_4B_4$,[25] $(Er_{1-x}Gd_x)Rh_4B_4$,[25] and $(Er_{1-x}Ho_x)Rh_4B_4$.[26] Note that the coexistence of superconductivity and long-range *ferromagnetic* order has not been seen in any of these materials.

III. SPIN-GLASS

In the spin-glass case, one must include the effects of disorder on $1/\tau$ and especially on $g(q,\omega)$. Grest *et al.*[17] have shown that for the spin-glass case

$$g^{SG}(q,\omega)=\frac{1}{2\pi N}\int_{-\infty}^{+\infty}dt e^{-i\omega t}\sum_{ij}e^{i\vec{q}\cdot\vec{R}_{ij}}\{[\langle S_i(t)S_j(0)\rangle]_c-[\langle S_i\rangle]_c[\langle S_j\rangle]_c\}\ , \tag{3.1}$$

where $[\ \ ]_c$ represents the configuration average. In a spin-glass $[\langle S_i\rangle]_c=0$, so that only the first term in Eq. (3.1) contributes to the scattering. We can express $1/\tau$ in terms of the spectral function $F(q,\omega)$ if we add and subtract in Eq. (3.1) the term $[\langle S_i(\infty)S_j(0)\rangle]\equiv\tilde{Q}_{ij}$ which is time independent. We find for a spin-glass that

$$\frac{\tau_{AG}}{\tau}=\frac{\tilde{Q}}{S(S+1)}+\frac{1}{2k_F^2S(S+1)}\int^{2k_F}qdq\int_{-\infty}^{+\infty}d\omega\frac{\beta^2\omega^2e^{-\beta\omega}}{(e^{-\beta\omega}-1)^2}k_BT\chi(q)F(q,\omega) \tag{3.2}$$

if we take $\tilde{Q}_{ij}=\tilde{Q}\delta_{ij}$ which occurs in mean-field theory. As above we have to assume a form for $F(q,\omega)$ and $\chi(q)$. For the $F(q,\omega)$, we follow the suggestion by Dzyaloshinskii and Volovik[27] which was applied to the resistivity by Fischer[28] and use the form given in Eq. (2.9). Since we know little about the dependence of D on q and T in a spin-glass, we take $D=D_0$, a constant. However the results are not dependent on this choice. We can use this form for $F(q,\omega)$ both above and below the transition temperature T_{SG} since even at low temperatures we do not have well-defined excitations. Most likely they are of the diffusive type. For $\chi(q)$, we consider a simple model which neglects the q dependence and take

$$\chi(q)=\begin{cases}\dfrac{S(S+1)}{k_BT}, & T\geq T_{SG}\\[2mm] \dfrac{S(S+1)}{k_BT_{SG}}, & T<T_{SG}\end{cases}\ . \tag{3.3}$$

Above T_{SG} we use the usual paramagnetic result, but below T_{SG} we take χ to be constant in agreement with the experimental results. The quantity $\tilde{Q}$ is like

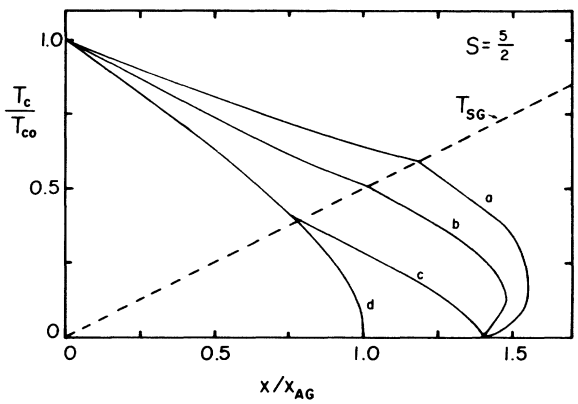

FIG. 4. Phase diagram for spin-glass with spin $S=\frac{5}{2}$. Curves a to c show T_c for $B=5$, 2, and 0.1. The curve d is the AG result.

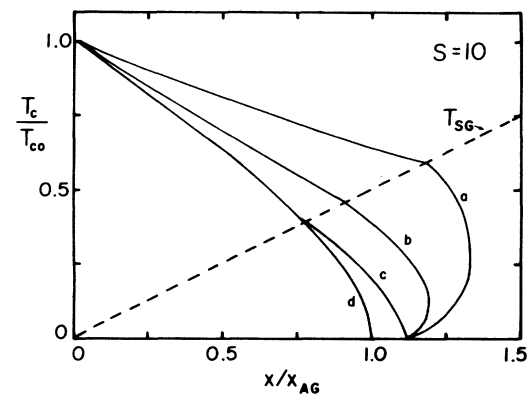

FIG. 5. Phase diagram for spin-glass with spin $S=10$. Curves a to c show T_c for $B=5$, 1, and 0. The curve d is the AG result.

an order parameter for the spin-glass, and we assume[22,28]

$$\tilde{Q}=S^2(1-T/T_{SG}) \quad . \tag{3.4}$$

The value of $\tilde{Q}$ at low temperature is difficult to determine because the usual spin-glass models are not ergodic. We have chosen the value of $\tilde{Q}$ at $T=0$ to give the correct zero temperature limit for $1/\tau$ as was found by Grest *et al.*[17] In that case $1/\tau$ was calculated directly for a simple model without the need to introduce phenomenological forms for $F(q,\omega)$ or $\chi(q)$. In the spin-glass case we consider the entire temperature region both above and below T_{SG}. We assume that below T_{SG} the coherence length is long enough that the effect of the internal magnetic field which are random in direction do not contribute to the pair breaking. We have calculated the phase diagram for a spin-glass for different S values. In Fig. 4, we show the case for $S=\frac{5}{2}$ with $T_{SG}=0.5x/x_{AG}$. Note that even when $B=0$ we have a larger region of coexistence than the usual AG theory would predict, due to the spatial correlations. As B increases, the region of coexistence increases and at the same time we have the *re-entrant* phenomena in the spin-glass in qualitative agreement with experiment.[29] The small kink at $T_{SG}=T_c$ is not real but an artifact of the assumed temperature dependence of $\tilde{Q}$. The same qualitative phase diagram is obtained for the case $S=10$, shown in Fig. 5. The only difference is that T_c goes to zero at a different critical concentration x_{cr}, which is given by $x_{cr}=[(S+1)/S]x_{AG}$. The curvature of T_c/T_{c0} for $T<T_M$ is seen in the experimental results for $(La_{1-x}Gd_x)Ru_2$.[27] We should point out that the Born approximation overestimates the value of x_{cr}. Keller and Benda[13] showed that this result is correct only for a large internal field. However this correction will only enhance the curvature in T_c/T_{c0} shown in Figs. 4 and 5, and does not change the essential conclusions for the spin values shown. This effect is important for $S=\frac{1}{2}$ or 1.

IV. CONCLUSIONS

We have investigated the influence of *time-dependent* and spatial-dependent correlations on the pair breaking, for ferromagnetic and spin-glass systems. We find that spatial correlations of the ferromagnetic type enhance the pair breaking effect, while the spatial correlations in an antiferromagnet or spin-glass reduce the pair breaking. At the same time we find that the introduction of dynamics (time correlations) always reduces the pair breaking for all types of magnetic ordering.

In the ferromagnetic case we fit the experimental phase diagram data for pseudoternary compound $(Lu_{1-x}Ho_x)Rh_4B_4$ and find good qualitative agreement with experiment. In the spin-glass case the introduction of the time correlations on the pair breaking gave as a result a re-entrant phenomena for the spin-glasses, in qualitative agreement with experiment. As we already mentioned in the text the way of introducing the effects of dynamics (time correlations) on the pair breaking is only an approximation. It should be considered a first step in a complete solution of the problem.

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