Attenuation of magnetic interactions in amorphous metals

S. J. Poon
Department of Applied Physics, Stanford University, Stanford, California 94305
(Received 16 April 1979)

In an amorphous superconductor diluted with magnetic impurities, the range of indirect exchange interaction can be determined from upper-critical-field and density-of-states measurements. It is found that the range of magnetic interaction in some amorphous *d*-band metals is larger than the free-electron transport mean free path. The present result has important implications on the occurrence of various magnetic phases in amorphous alloys.

The Ruderman-Kittel-Kasuya-Yosida (RKKY)[1] interaction is known to play a significant role in determining the magnetic properties of alloys. de Gennes[2] extended the RKKY calculation to disordered alloys where the mean free path is finite. He showed that the RKKY interaction would then be attenuated according to an exponential law $\exp(-r/l_0)$ where l_0 is approximately the mean free path. Heeger *et al.*[3] and more recently Smith[4] explained their NMR and magnetization results on *CuAl*Mn and *AgAu*Mn alloys, respectively, using the de Gennes theory. So far these have been the only quantitative test of the RKKY theory in disordered alloys. A direct application of this theory to magnetic metglasses would limit the range of interaction to the first nearest neighbors (FNN) when the transport mean free path l_{tr} (from nearly-free-electron calculation $\sim 1-2$ Å) is taken as the relevant length. In fact, the unusual magnetic properties, such as the frequent occurrence of spin-glass and mictomagnetic phases in amorphous rare-earth alloys have been interpreted by various authors using a FNN interaction model which includes fluctuation in FNN.[5] To avoid complications encountered in concentrated alloys, one should investigate the range of magnetic interaction in amorphous dilute alloys from which the validity of the popular interpretation in concentrated alloys can be tested.

In this article, I show that the range of RKKY interaction l_0 in amorphous superconductors diluted with magnetic impurities can be determined from upper-critical-field $H_{c2}(T)$ and density-of-states $N(0)$ measurements. I first establish a rigorous relationship between the kernel $K(\vec{q})$ in the linearized Ginzburg-Landau equation and the RKKY magnetic susceptibility $\chi(\vec{q})$. In the dirty limit, the electron correlation length determined from superconductive measurements would yield the correct attenuation of the interaction. The present study is motivated by the experimental results on amorphous $La_{80}Au_{20}$ alloys containing less than 1-at.% Gd,[6] as shown in Fig. 1. The scaling relation $M/c = g(T/c, H/c)$ obtained indicated that the magnetic interaction varies as $1/r^3$ and thus $l_0 >> l_{tr} \sim 2$ Å. Moreover, the spin-glass temperatures which are determined by the strength of RKKY interaction are comparable to those obtained in crystalline alloys (e.g., La, LaB_6, $LaAl_2$) containing small amount of Gd. This also casts doubt on the validity of the FNN interaction model.

The derivation of the kernel $K(\vec{q})$ follows closely to that of de Gennes.[7] $K(\vec{q})$ is found to be

$$K(\vec{q}) \propto \sum_{E,n} \left(\frac{Vk_BT}{\hbar}\right)\left(\mathrm{Im}\,\frac{1}{E+i|\omega_n|}\right) \times \int_0^\infty dt\, e^{-2|\omega_n|t/\hbar}\,\mathrm{Re}F(\vec{q},E,t) \quad , \tag{1}$$

where $\omega_n = (2n+1)\pi k_BT$ is the Matsubara frequency, the correlation function

$$F(\vec{q},E,t) = \langle E|e^{-i\vec{q}\cdot\vec{r}(t)}e^{i\vec{q}\cdot\vec{r}}|E\rangle \quad .$$

V is the interaction potential, and E is measured from

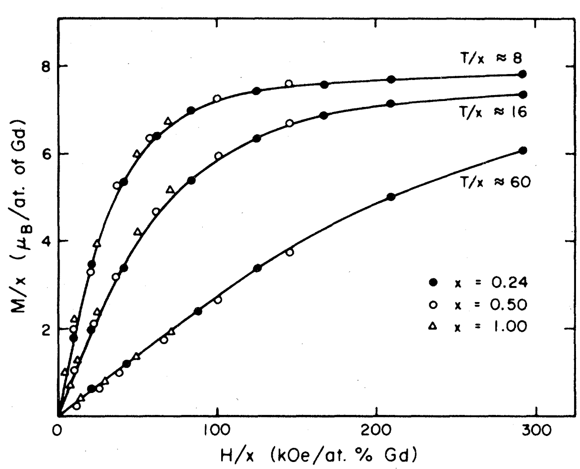

FIG. 1. Reduced magnetization M/x as a function of reduced magnetic field H/x at three different temperature T/x (from Ref. 6).

the Fermi level. Simplifying

$$K(q) \propto \sum_n \frac{N(0)\,Vk_BT}{\hbar}\int_0^\infty dt\, e^{-2|\omega_n|t/\hbar}\,\mathrm{Re}F(\vec{q},0,t) \ . \tag{2}$$

The RKKY magnetic susceptibility function $\chi_c(\vec{q})$ is in general given by

$$\chi_c(\vec{q}) \propto J^2 \sum_{E\leq 0,E'} \left[\frac{\langle E|e^{i\vec{q}\cdot\vec{S}}|E'\rangle\langle E'|e^{-i\vec{q}\cdot\vec{T}}|E\rangle}{E-E'}\right]_c , \tag{3}$$

where $[\ \ ]_c$ denotes configurational average and J is the coupling between localized moment and conduction electrons.

Next we introduce a generalized susceptibility function $\chi_c(\vec{q},\omega_n)$ simply by adding or subtracting an imaginary part $i|\omega_n|$ to the energy E. In the time-dependent picture, it is just like introducing a relaxation process to the electronic states. $\chi_c(\vec{q},\omega_n)$ derived from Eq. (3) has the simple form

$$\chi_c(\vec{q},\omega_n) \propto \frac{iJ^2}{\hbar}\sum_{E\leq 0}\int_0^\infty dt\, e^{-2|\omega_n|t/\hbar}F(\vec{q},E,t) \ . \tag{4}$$

The magnetic susceptibility function $\chi(\vec{q})$ is given by $\lim_{T\to 0}\mathrm{Re}\chi_c(\vec{q},\omega_n)$. One can see from Eqs. (2) and (4) that both $K(\vec{q})$ and $\chi(\vec{q})$ are given by the same correlation function $F(\vec{q},E,t)$ which depends on the same electron correlation length in disordered alloys. One can relate Eqs. (2) and (4) to obtain

$$K(\vec{q}) \propto \sum_n \frac{Vk_BT}{J^2}\mathrm{Im}\left(\frac{\partial\chi_c(\vec{q},\omega_n)}{\partial E_F}\right)_{E_F=0} . \tag{5}$$

Without using the intuitive form of $F(\vec{q},0,t)$ given in Ref. 7, one can employ the self-consistent method of Ref. 2 to evaluate the susceptibility function $\chi_c(\vec{q},\omega_n)$ and thus $K(\vec{q})$ from Eq. (5). In the clean limit, the nearly-free-electron (NFE) treatment gives

$$K(\vec{q}) \propto \mathrm{Im}\sum_n \frac{N(0)\,Vk_BT}{\hbar v_Fq} \times \ln\left|\frac{\hbar^2q^2+2\hbar mv_Fq+2mi|\omega_n|}{\hbar^2q^2-2\hbar mv_Fq+2mi|\omega_n|}\right| \ . \tag{6a}$$

The kernel $K(R)$ can be obtained by Fourier transforming $K(\vec{q})$

$$K(R) \propto \sum_n \frac{N(0)\,Vk_BT}{\hbar R^2}\sinh\left(\frac{2|\omega_n|}{\hbar v_F}+\mathcal{O}\right)R \ , \tag{6b}$$

where

$$\mathcal{O} \sim i\left(\frac{|\omega_n|}{\hbar v_F}\right)^2\left(\frac{\hbar}{mv_F}\right) << i\frac{\omega_n}{\hbar v_F} \ ,$$

for $|\omega_n| << mv_F^2$ and v_F is the band Fermi velocity. Using a similar approach the temperature-dependent RKKY theory yields an interaction range $\sim\hbar v_F/k_BT$ at finite temperature. Close to the critical temperature, T_c, it is sufficient to retain only the q^2 term in $K(q)-K(0)$ so that Eq. (6a) gives

$$K(q)-K(0) \propto N(0)\,Vk_BT_c\hbar^2v_F^2q^2\sum_n\frac{1}{|\omega_n|^3} \ . \tag{7}$$

In disordered alloys, we obtain

$$K(q) \propto \frac{N(0)\,Vk_BT}{\hbar}\sum_n \frac{\mathrm{Im}k_n(q)\left(1-\frac{v_F}{2l_c}\mathrm{Im}k_n(q)\right)-\frac{v_F}{2l_c}[\mathrm{Re}k_n(q)]^2}{\left(1-\frac{v_F}{2l_c}\mathrm{Im}k_n(q)\right)^2+\left(\frac{v_F}{2l_c}\mathrm{Re}k_n(q)\right)^2} , \tag{8}$$

where

$$k_n(q) = \frac{1}{2v_Fq}\ln\frac{\hbar^2q^2+2\hbar mv_Fq+2mi|\omega_n|+\frac{mi\hbar v_F}{l_c}}{\hbar^2q^2-2\hbar mv_Fq+2mi|\omega_n|+\frac{mi\hbar v_F}{l_c}}$$

and the electron correlation length l_c (the distance beyond which the wave functions lose their phase correlation) comes from $\chi_0(\vec{q})$. Expanding $K(q)-K(0)$ up to q^2 term for $T \simeq T_c$, the result is

$$K(q)-K(0) \propto N(0)\,Vk_BT_c\hbar^2v_F^2q^2 \times\sum_n\left(1+\frac{\hbar v_F}{2l_c|\omega_n|}\right)^{-1}\frac{1}{|\omega_n|^3} \ . \tag{9}$$

It should be reminded that in deriving the results (6a) and (8), the NFE approximation is assumed. However, for small q, these results are valid for any isotropic Fermi surface as long as v_F is interpreted as the band velocity and l_c is the electron correlation length. Equations (7) and (9) are the same as those obtained by Gor'kov.[8] The gradient of the upper critical field near T_c obtained from $K(\vec{q})$ is given by

$$\left(\frac{dH_{c2}}{dT}\right)_{T_c} = 12ck_B(1+\lambda)/\pi ev_Fl_c \ ,$$

where the electron-phonon renormalization factor $1+\lambda$ has been included.[9] Except in the NFE approximation, there is no *a priori* reason to relate l_c to the free-electron transport mean free path l_{tr}. On the other hand, using the Kubo-Greenwood formalism, it had been shown that the electrical conductivity in disorder alloys was given by a complicated function of l_c and band-structure parameters.[10]

The magnetic susceptibility for an isotropic Fermi surface can be approximated as

$$\chi(R) \approx \chi_0(R)\eta(l_c)e^{-R/l_c} \quad , \qquad (10)$$

where $\chi_0(R)$ is the band susceptibility in the clean limit and $\eta(l_c)$ is an enhancement factor obtained from the self-consistent treatment of $\chi(R)$. Fähnle[11] found significant band-structure effect on $\chi_0(R)$ over several atomic spacings. In a non-self-consistent treatment, $\eta(l_c)=1$. However even in the NFE case, $\eta(l_c)$ can only be obtained by numerical integration of the $\chi(\vec{q})$ function. For our present values of l_c, I obtain $\eta(l_c)$ by numerical interpolation of the results of Ref. 2 with $k_F \approx 1$ Å^{-1}.

The range of RKKY interaction l_0 is defined by $\chi(l_0)/\chi_0(l_0)=e^{-1}$ which gives $l_0=l_c[1+\ln\eta(l_c)]$. The correlation length l_c is determined from $H_{c2}(T)$ and $N(0)$ measurements. We introduce a parameter l_f such that

$$\left(\frac{dH_{c2}}{dT}\right)_{T_c}^{-1}(1+\lambda) \propto v_{Ff}l_f = v_F l_c \quad ,$$

where the subscript f denotes NFE results by counting all the valence electrons. The band Fermi velocity $v_F \propto S_F/N(0)$ where S_F is the spherical-Fermi-surface area.[12] With additional parameters defined by $\delta = S_{Ff}/S_F$ and $\alpha = N(0)/N_f(0)$, one obtains $V_{Ff}/V_F=\alpha\delta$. Approximately $\delta = S_{Ff}/S_F=(z_v/z_{\text{eff}})^{2/3} \geqslant 1$ where z_v and z_{eff} denote the number of valence and effective electrons, respectively. Therefore, $l_c=\alpha\delta l_f$ and a lower limit on l_c is reached when $\delta=1$. The values of α, l_f, and the lower limits of l_c which also give the minimum range of the RKKY interaction l_0 for three well studied amorphous superconductors, are listed in Table I. One can see that l_0 is at least three times the usually quoted transport mean free path.

Although the discussion presented here is based on amorphous superconductors, the conclusion is expected to be applicable to other amorphous alloys. Thus the present study indicates that the attenuation of RKKY interaction in some d-band amorphous metals is effective *only beyond the second nearest neighbors.* This certainly has significant implications on the nature of magnetic interaction and the occurrence of various magnetic phases in amorphous metals, depending on alloy concentration. So far, we have not separated the conduction electrons into s and d states. If the conduction band happens to be d-band dominated, then whether the RKKY interaction plays an important role in determining the magnetic structure would depend on the coupling $J_{d\mu}$ (μ is the local moment). The latter is usually found to be small in crystalline alloys. However recent results from microwave magnetic resonance experiments on amorphous Gd-Al alloys[13] indicates that $J_{d\mu}$ is quite large. In concentrated alloys, if l_0 extends to second or third nearest neighbors, one would not expect a drastic difference in the interaction range between amorphous and crystalline alloys. Then, in place of the attenuation picture, one should really focus on the strength of interaction J^2 and the structural short-range order (clusters containing atoms up to second or third coordination shell) which is sufficient for determining the band structure and thus the susceptibility $\chi_0(R)$ in both cases. A cluster-type band structure and susceptibility calculation would help to clarify the picture. Recently, a magnetic cluster description of spin glasses in amorphous alloys without invoking a very short interaction range was presented.[14] In dilute alloys containing less than 1-at.% magnetic solutes, the situation could be quite different. It can be shown that even by including the band-structure effect, as in Ref. 11, the RKKY interaction recovers its $1/r^3$ dependence at large distance. Then l_0 certainly plays an important role. If we take the value $\delta \approx 0.5$ so that $l_0 \approx 12$ Å in

TABLE I. Upper-critical-field gradient, density of states at the Fermi level, the parameter α, and lengths l_f, l_c, l_0, and l_{tr} as discussed in text.

Alloys	$\left(\frac{dH_{c2}}{dT}\right)_{T_c}$ (kOe/K)	$N(0)$ (spin states/eV atom)	α	l_f (Å)	l_c (Å)	l_0 (Å)	l_{tr} (Å)
$La_{80}Au_{20}$	22[a]	0.8[b]	2.76	1.62	4.47	6.17	1.9
$La_{80}Ga_{20}$	22.5[b]	0.7[b]	2.30	1.68	3.86	5.62	1.7
$(Mo_{0.8}Ru_{0.2})_{80}P_{20}$	24.5[c]	0.93[c]	4.00	1.00	4.00	5.74	1.6

[a]W. L. Johnson, S. J. Poon, and P. Duwez, Phys. Rev. B 11, 150 (1975).
[b]W. H. Shull, D. G. Naugle, S. J. Poon, and W. L. Johnson, Phys. Rev. B 18, 3263 (1978).
[c]W. L. Johnson, S. J. Poon, J. Durand, and P. Duwez, Phys. Rev. B 18, 206 (1978).

$La_{80}Au_{20}$ alloys, then the attenuation effect is important for alloys containing less than ~0.5-at.% Gd. This result is in marginal agreement with experimental observation.[6] The evidence of magnetic interaction in dilute alloys is also shown by recent studies[14] of T_c suppression ΔT_c in amorphous superconductors carrying localized moments (up to ~1 at.%). The reduced spin-flip scattering is manifested by the reduced ΔT_c with respect to the Abrikosov-Gor'kov theory.

Finally one should note that the value of transport mean free path l_{tr} depends on the particular set of electronic states one uses to interpret the conductivity data. For example, $l_{tr} \sim 1-2$ Å is the result of a NFE calculation by counting the appropriate number of conduction electrons in the modified Ziman theory.[15] The success of this approach relies on the fact that the hypothetical electronic states are used in a self-consistent manner with a structure factor obtained from x-ray plane-wave diffraction. However, the value of l_{tr} obtained is irrelevant to l_c in the susceptibility calculation. For comparison we have included the values of l_{tr} in Table I.

In conclusion, it is shown that a direct relationship exists between the kernel in the superconductivity theory and magnetic susceptibility. Thus upper-critical-field measurement should yield the same value of electron correlation length as in magnetic susceptibility. In amorphous *d*-band alloys, a reliable value of this correlation length can only be obtained by knowing also the data on the density of states. Also from available data on $H_{c2}(T)$ and $N(0)$ it is found that the range of RKKY interaction is larger than the value conventionally quoted in the literature.

ACKNOWLEDGMENT

The author thanks Dr. A. P. Malozemoff for useful discussions on Ref. 2. This work was supported by the Air Force Office of Scientific Research Contract No. F49620-78-C-0009.

---

[1]For a review, see C. Kittel, Solid State Phys. 22, 1 (1968).

[2]P. G. de Gennes, J. Phys. Rad. 23, 630 (1962). The magnetic susceptibility is not simply given by $\chi(R)=\chi_0(R)e^{-r/l_0}$ where $\chi_0(R)$ is the free-electron result, as cautioned by the author. Such $\chi(R)$ does not satisfy the global conservation of susceptibility. However, the exponential factor gives a rather good qualitative description. Also one should note the errors in Eq. (38), where a factor of $\frac{1}{2}$ is missing in S', and S'' should be

$$\frac{1}{2}\ln\frac{(2q'-p)^2+4q''^2}{(2q'+p)^2+4q''^2} .$$

When $q''=0$ in the clean limit, S'' is reduced to the usual free-electron result.

[3]A. J. Heeger, A. P. Klein,and P. Tu, Phys. Rev. Lett. 17, 803 (1966).

[4]F. W. Smith, Solid State Commun. 25, 341 (1978).

[5]See papers in *Amorphous Magnetism II,* edited by R. A. Levy and R. Hasegawa (Plenum, New York, 1977); in Proceedings of the 24th Conference on Magnetism and Magnetic Materials; Michael W. Klein, J. Phys. F 7, 1699 (1977); T. Kaneyoshi, J. Phys. Soc. Jpn. 45, 94 (1978).

[6]S. J. Poon and J. Durand, Solid State Commun. 21, 793 (1977); Phys. Rev. B 18, 6253 (1978). The strength of interaction determined from high-field magnetization is about two orders of magnitude higher than what one would expect from dipolar interaction, meanwhile no crystal-field effect has been observed in LaGd alloys.

[7]P. G. de Gennes, *Superconductivity in Metals and Alloys* (Benjamin, New York, 1966).

[8]L. P.Gor'kov, Zh. Eksp. Teor. Fiz. 37, 833, 1407 (1959) [Sov. Phys. JETP 10, 593, 998 (1960)].

[9]G. Bergmann, Phys. Lett. C 27, 159 (1976).

[10]A. B. Chen, G. Weiss, and A. Sher, Phys. Rev. B 5, 2897 (1972).

[11]M. Fähnle, J. Mag. Mater. 10, 9 (1979).

[12]Smearing of Fermi surface in *k* space is expected, but it should be limited by the exclusion principle and also the sum rule that integration over phase space gives the total number of electrons.

[13]J. P. Jamet and A. P. Malozemoff, Phys. Rev. B 18, 75 (1978).

[14]S. J. Poon and J. Durand, Solid State Commun. 21, 999 (1977); Magnetization results on these alloys do not show clustering of magnetic impurities.

[15]L. V. Meisel and P. J. Cote, Phys. Rev. B 17, 4652 (1978).