Weak localization and enhanced electron-electron interaction in amorphous $Ca_{70}(Mg,Al)_{30}$

A. Sahnoune and J. O. Strom-Olsen
Department of Physics, McGill University, 3600 University Street, Montreal, Quebec, Canada H3A 2T8
(Received 25 October 1988)

We present measurements and analysis of low-temperature resistance and magnetoresistance in amorphous $Ca_{70}(Mg,Al)_{30}$. The data are described within error over the full range of field and temperature by the expressions due to weak localization and enhanced electron-electron interaction, the first time such close agreement has been found in a three-dimensional system. The results show that the quality of the fit is independent of the disorder parameter $(k_F l_e)^{-1}$ over the range 0.05–0.4.

I. INTRODUCTION

In highly disordered conductors, intense elastic scattering causes interference effects on the electron wave function which give rise to the quantum corrections to conductivity (QCC) known as weak localization[1] (WL) and enhanced electron-electron interaction[2] (EEI). A large number of theoretical papers have been published on the subject[3] and the various expressions for electrical conductivity have been extensively tested experimentally, particularly in two dimensions.[4] In three dimensions QCC theories have probably found their widest application to the electrical resistance of amorphous metals at low temperatures, principally magnetoresistance,[5] but also, to a lesser extent, temperature dependence.[6] Qualitatively, there is no doubt that the QCC expressions describe the observations, but when experiment and theory are compared in detail quite substantial discrepancies emerge.[5,7] The significance of these discrepancies has often been difficult to assess, because in many instances the systems examined have either been not very well characterized (e.g., they contained an indeterminate level of magnetic impurities, or had an open *d* band), or they have suffered from conflicting contributions due to superconductivity or ferromagnetism. Probably the best test hitherto has been through the work of Richter, Baxter, and Strom-Olsen,[8] who presented the results of a series of measurements on ultrahigh purity (i.e., magnetic contaminant typically <0.5 ppm) Mg-Cu alloys, doped with various levels of Ag or Au. These alloys are known to be free-electron-like,[9,10] show neither superconducting nor magnetic order, and have a disorder parameter $(k_F l_e)^{-1}$ small enough (about 0.03) for one to expect the QCC theories to be accurate. Furthermore, the alloys are so well characterized from independent measurements that the only two unknown parameters that appear in the theoretical expressions are the electron wave-function dephasing time, τ_ϕ, and the spin-orbit scattering time, $\tau_{s.o.}$, and $\tau_{s.o.}$ suffers the additional constraint of being temperature independent. Under these fairly stringent conditions it was found that at low magnetic fields (where contributions from WL are dominant) QCC theories describe the data to within error, but that at higher fields discrepancies appear which become more pronounced as the field increases. It was not possible to determine from these results where the theoretical expressions failed, but there was a slight trend for the discrepancies to increase as the spin-orbit scattering rate $\tau_{s.o.}^{-1}$ increased when Ag or Au was substituted for Cu. The range covered was from moderately weak to strong spin-orbit scattering ($\tau_{s.o.}^{-1} \sim 10^{12}$–$10^{14}$ s^{-1} as compared with a typical dephasing rate $\tau_\phi^{-1} \sim 10^{10}$ s^{-1} at 4.2 K).

In this article we present results on the magnetic field and temperature dependence of resistivity in $Ca_{70}(Mg,Al)_{30}$ amorphous alloys. Although similar to Mg-Cu, in that they are simple *s,p* metals,[11,12] they differ in two important respects. First, the level of spin-orbit scattering is an order of magnitude smaller, which allows QCC theories to be tested in the very weak spin-orbit scattering limit. Second, as Al replaces Mg, the elastic mean free path, l_e, is reduced by an order of magnitude, while other electronic parameters remain constant. Thus, in an otherwise constant system the effect of changing the disorder parameter $(k_F l_e)^{-1}$ from much less than unity to less than but of the order unity may be examined.

II. EXPERIMENTAL METHODS

Alloys were made by melt spinning precast buttons of ~ 1 g onto a Cu wheel with a tangential speed from 40 to 45 m s^{-1} under 15 kPa of high-purity helium. The buttons were made by induction melting the appropriate amount of Ca (99.999% purity from Rare Metallic Co., Japan), Mg (99.95% purity from Alfa Products), and Al (99.9999% purity from McKay Inc.) in a high-purity graphite crucible under high-purity argon (99.998% purity). Prior to melting, the Mg was quartz distilled[13] to remove any transition-metal impurities. The melt-spun ribbons were examined by x-ray diffraction and then stored under liquid nitrogen. No diffraction lines due to any crystal phase were observed, except that for some samples various oxides of Ca (CaO_4, CaO_2) were present on the surface. These oxides could be removed by polishing and have no effect on our results.

The values of resistivity, and its temperature coefficient at room temperature, are in excellent agreement with those measured by Mizutani *et al.*[11] Of the remaining parameters in Table I, $N(E_F)$ is calculated from the measured electronic specific heat[11] and the diffusivity from $N(E_F)$ and ρ.

Magnetic impurity content (in particular, Mn, which is the only contaminant of any of the starting elements

TABLE I. Physical parameters of $Ca_{70}(Mg,Al)_{30}$ amorphous alloys. Error: d, ±2%; ρ, ±5%; [Mn], ±10%.

Alloy	d (g/cm^3)	ρ ($\mu\Omega$ cm)	$(k_F l_e)^{-1}$	D (10^{-4} m^2/s)	$N(E_F)$ (states/eV atom)	F	T_F (K)	[Mn] (ppm)
$Ca_{70}Mg_{30}$	1.58	48	0.05	7.9	0.61	0.53	60440	3.4
$Ca_{70}Mg_{22.5}Al_{7.5}$	1.65	80	0.09	4.4	0.64	0.53	63460	3.8
$Ca_{70}Mg_{15}Al_{15}$	1.70	122	0.13	3.0	0.60	0.52	66090	3.5
$Ca_{70}Mg_{7.5}Al_{22.5}$	1.76	195	0.20	2.0	0.55	0.52	68770	4
$Ca_{70}Al_{30}$	1.78	342	0.35	1.25	0.49	0.52	70740	4

which may be expected to carry a moment in the alloys[14]) was measured by neutron activation. The results are shown in Table I, along with other pertinent parameters for the system. The density was measured using Archimedes's principle using toluene as the working fluid; the resistivity was determined from the measured resistance of a known length (typically 50 cm), the cross-sectional area being determined from the measured mass and density.

Resistance changes, either as a function of magnetic field or temperature, were measured by a four-terminal ac bridge[15] able to detect changes of less than 1 part in 10^5 in a 1-Ω resistor (the samples varied in resistance from 0.4 to 0.8 Ω). The temperature was varied from 1.2 to 20 K either by pumped helium or by a servo-controlled heater; in either case stability was 1% or better. A magnetic field of up to 4.3 T was provided by a superconducting solenoid. All data were stored on a microcomputer and processed on a SUN minicomputer.

III. RESULTS AND DISCUSSIONS

Figure 1 shows the magnetoresistance as a function of temperature in three of the $Ca_{70}(Mg,Al)_{30}$ alloys: $Ca_{70}Mg_{30}$, $Ca_{70}Mg_{15}Al_{15}$, and $Ca_{70}Al_{30}$. In all cases the points are the experimental data and the solid line the fit to the theoretical expressions, as discussed below. The features of the curves are in agreement with what is expected from QCC for alloys with weak spin-orbit scattering, namely a small positive magnetoresistance at low fields that changes sign and becomes negative at high fields. When the temperature is raised, the magnetoresistance diminishes as the phase-coherence time shortens due to electron-phonon scattering.

The data are fitted to contributions from WL and EEI, expressions for which are well known and are given in the Appendix. We follow a fitting procedure similar to that given in Richter, Baxter, and Strom-Olsen.[8] At low fields ($B/T < 0.4$ T/K) the contribution to $\Delta\rho/\rho$ from EEI is less than about 10^{-6} and may be neglected. Thus we begin by restricting the fitting to this range and use only Eq. (A1) to describe the data. In the fitting there are only two free parameters, B_ϕ and $B_{s.o.}$ (which are related to τ_ϕ and $\tau_{s.o.}$); furthermore, $B_{s.o.}$ must be independent of temperature. Thus each family of curves in Fig. 1 is fitted at low field to a common value of $B_{s.o.}$ and one value of B_ϕ at each temperature. At higher fields the contribution from EEI, which is positive, must be included, as is shown by the fact that the theoretical expression for WL fitted at low fields lies below the experimental data when it is extended to high fields, the separation between the two being about 30% at 4 T and 1.5 K. No new parameters occur in the expression for EEI and its contribution may thus be considered a zero-parameter fit. The result of including EEI gives an essentially perfect fit to the data within error over the full field range in four of the five alloys and a very good fit in $Ca_{70}Al_{30}$ as shown in Fig. 1(c). So far, as we know, this is the first three-dimensional system for which such excellent agreement has been found. Comparing these results with those from Cu-Mg alloys, two points emerge. The first is that the only essential difference between $Ca_{70}Mg_{30}$ and $Mg_{70}Cu_{30}$, with or without doping by Ag or Au, is the level of spin-orbit scattering, $\tau_{s.o.}$ being about 10^{-11} s in the $Ca_{70}Mg_{30}$ (see Table II for the various scattering times and lengths) and anywhere from 3×10^{-12} to 3×10^{-13} s in the Mg-Cu alloys. Thus we are led to the empirical conclusion that QCC expressions as currently given are only valid in the limit of very weak spin-orbit scattering, namely when $\tau_{s.o.}^{-1} < \tau_\phi^{-1} \ll \tau_e^{-1}$.

The second significant point is that the perturbation parameter $(k_F l_e)^{-1}$ varies from 0.05 in $Ca_{70}Mg_{30}$ to 0.4 in $Ca_{70}Al_{30}$. Since expressions (A1), (A2), and (A3) are only the first terms in a perturbation expansion, one need not expect them to be quantitatively accurate for $Ca_{70}Al_{30}$. Our results, however, suggest that the higher terms are not significant and they lend support to the argument of Morgan, Howson, and Saub[16] that expressions (A1), (A2), and (A3) may be useful even when $(k_F l_e)^{-1} \sim 1$.

The behavior of the dephasing time with temperature is shown in Fig. 2. All samples show essentially the same values for τ_ϕ, which may be described by the expression

$$1/\tau_\phi = 1/\tau_\phi^0 + AT^n , \quad (1)$$

as was the case in Mg-Cu and Mg-Zn.[8] A best fit to the data yields $n = 3.0 \pm 0.5$, which is quite consistent with dephasing due to electron-phonon scattering. At low temperatures τ_ϕ saturates to τ_ϕ^0, whose value is similar to that seen in Mg-Cu and Mg-Zn. Richter, Baxter, and Strom-Olsen[8] were able to exclude extraneous causes of dephasing, such as spin scattering by residual magnetic impurities, and hence proposed that τ_ϕ^0 may have some intrinsic origin, perhaps zero-point motion.[17–20] The present samples, however, contain about 4 ppm Mn. If we assume Mn to carry a moment of $2.2\mu_B$ and to have an exchange[21] of 0.25 eV with the conduction electrons,

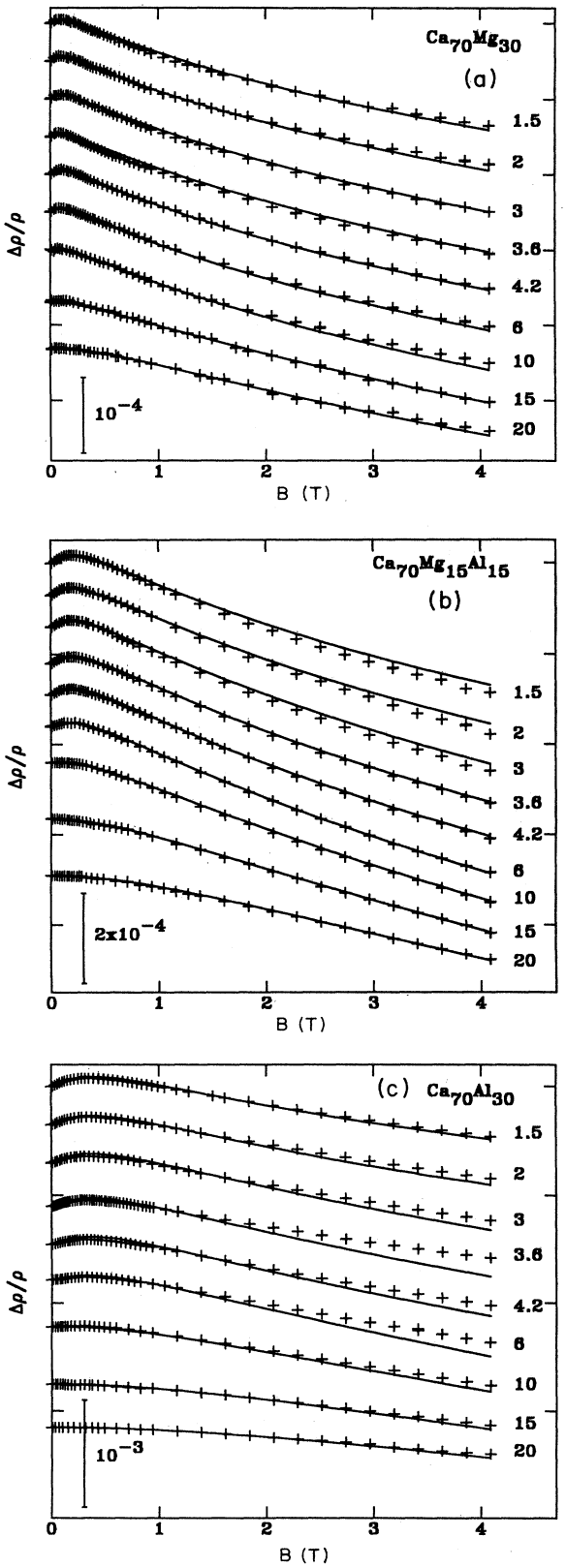

FIG. 1. Magnetoresistance of (a) $Ca_{70}Mg_{30}$, (b) $Ca_{70}Mg_{15}Al_{15}$, and (c) $Ca_{70}Al_{30}$. The + symbols are the experimental data and the solid line a fit as described in the text. Temperatures are indicated in the figure.

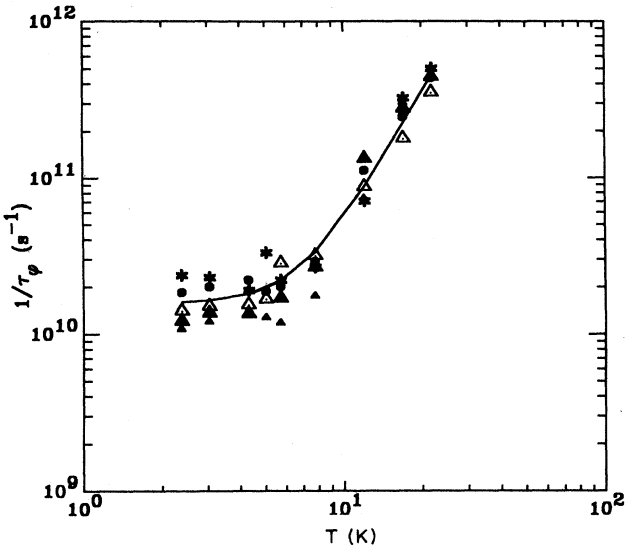

FIG. 2. Dephasing rate $1/\tau_\phi$ as a function of temperature in amorphous $Ca_{70}(Mg,Al)_{30}$. The solid line is a fit to the data using Eq. (1). $Ca_{70}Mg_{30}$ (*), $Ca_{70}Mg_{22.5}Al_{7.5}$ (▲), $Ca_{70}Mg_{15}Al_{15}$ (●), $Ca_{70}Mg_{7.5}Al_{22.5}$ (▲), and $Ca_{70}Al_{30}$ (△).

then spin scattering contributes a time of about 0.7×10^{-10} s to τ_ϕ^0, which is essentially the observed value. Thus, in the present alloys magnetic impurities prevent us from drawing any conclusion for an intrinsic limiting value for τ_ϕ^0.

The behavior of $\tau_{s.o.}$ with composition is given in Table II. There is very little change from $Ca_{70}Mg_{30}$ to $Ca_{70}Al_{30}$; the only point of interest is that $\tau_{s.o.}^{-1}$ actually decreases slightly, whereas one would perhaps expect it to increase, given that Al has an atomic number higher by 1. Perhaps this merely indicates that a hydrogenic orbit picture (which would give $\tau_{s.o.}^{-1}\sim Z^8$) is inadequate for describing the spin-orbit scattering of conduction electrons.

The temperature dependence of resistivity is also given by Eqs. (A1), (A2), and (A3). It turns out that, in contrast to magnetoresistance, EEI dominates the temperature dependence and that the contribution from WL may be neglected. In this case we expect[2]

$$-\frac{\Delta\rho}{\rho_0}=\frac{0.915e^2\rho}{4\pi^2\hbar}\left[\frac{4}{3}-\tfrac{3}{2}F^*-\frac{2}{\ln(T_F/T)}\right]\left[\frac{k_BT}{D\hbar}\right]^{1/2}, \tag{2}$$

where we have followed Hickey, Greig, and Howson[6] in replacing the screening parameter F by $F^*=F-\lambda$, λ being the electron-phonon mass-enhancement parameter. In the case of $Ca_{70}Al_{30}$ this may be estimated from thermopower measurements[22] to be 0.30, so that F^* becomes 0.22. The remaining parameters in (2) are known, so that the temperature dependence may now be calculated with no free parameters and compared with measurement, as in Fig. 3. As for magnetoresistance, the agreement between theory and experiment is essentially perfect, the

TABLE II. Characteristic scattering times and lengths of $Ca_{70}(Mg,Al)_{30}$ amorphous alloys. Error: τ_ϕ^0 and $\tau_{s.o.}$, ±10%.

Alloy	τ_e (10^{-15} s)	τ_ϕ^0 (10^{-10} s)	$\tau_{s.o.}$ (10^{-11} s)	l_e ($v_F\tau_e$ Å)	L_ϕ^0 $[(3D\tau_\phi^0)^{1/2}$ μm]	$L_{s.o.}$ $[(3D\tau_{s.o.})^{1/2}$ μm]
$Ca_{70}Mg_{30}$	1.4	0.6	0.7	18.4	0.4	0.13
$Ca_{70}Mg_{22.5}Al_{7.5}$	0.7	1.0	0.7	10.6	0.38	0.097
$Ca_{70}Mg_{15}Al_{15}$	0.5	0.6	1.0	6.7	0.24	0.093
$Ca_{70}Mg_{7.5}Al_{22.5}$	0.3	0.8	1.04	4.0	0.24	0.08
$Ca_{70}Al_{30}$	0.15	0.9	1.25	2.3	0.20	0.07

first time to our knowledge that such agreement has been observed.

IV. CONCLUSIONS

From the results in this paper and in Mg-Cu alloys,[8] we conclude that QCC theories are quantitatively accurate in bulk disordered metals when the spin-orbit scattering is very weak, but that even a moderate level of spin-orbit scattering is sufficient to produce significant discrepancies. We find that varying the disorder parameter from $\ll 1$ to $\lesssim 1$ has no significant effect on the fitting. Finally, using values of parameters determined from magnetoresistance, we found that QCC theories give a zero-parameter fit to the temperature dependence of the resistivity.

ACKNOWLEDGMENTS

This research was supported by the Natural Sciences and Engineering Research Council of Canada, The Fonds pour la Formation des Chercheurs et à l'aide à la Recherche of the Province of Quebec, the Ministère d'Enseignement et de la Recherche Scientifique of Algeria, and the graduate faculty of McGill University.

APPENDIX

1. Weak localization

According to Fukuyama and Hoshino, the magnetoresistance due to localization of the conduction electrons, in the presence of spin-orbit and inelastic scattering and including the splitting of the spin subbands, is given by[23]

$$\left[\frac{\Delta\rho}{\rho}\right]_{WL}=\rho\frac{e^2}{2\pi^2\hbar}\left[\frac{eB}{\hbar}\right]^{1/2}\left\{\frac{1}{2(1-\gamma)^{1/2}}\left[f_3\left[\frac{B}{B_-}\right]-f_3\left[\frac{B}{B_+}\right]\right]-f_3\left[\frac{B}{B_2}\right]\right.$$
$$\left.-\left[\frac{4B_{s.o.}}{3B}\right]^{1/2}\left[\frac{1}{(1-\gamma)^{1/2}}(t_+^{1/2}-t_-^{1/2})+t^{1/2}-(t+1)^{1/2}\right]\right\}, \quad \text{(A1)}$$

where

$$t=\frac{3(B_i+2B_s)}{4B_{s.o.}},$$
$$t_\pm=t+\tfrac{1}{2}[1\pm(1-\gamma)^{1/2}],$$
$$B_\pm=B_i+2B_s+\frac{2(B_{s.o.}-B_s)}{3}[1\pm(1-\gamma)^{1/2}],$$
$$B_2=B_i+\tfrac{2}{3}B_s+\tfrac{4}{3}B_{s.o.},$$

with

$$\gamma=\left[\frac{3g^*\mu_B B}{8eD(B_{s.o.}-B_s)}\right]^2.$$

g^* is the effective g factor and has a value of 2 in free-electron metals, and D is the diffusion constant.

The characteristic fields are related to the electron-scattering times by the relation $B_x=\hbar/4eD\tau_x$, where τ_x is the inelastic scattering time τ_i, spin-orbit scattering time $\tau_{s.o.}$, and magnetic impurity scattering time τ_s.

The function $f_3(x)$ in Eq. (A1) has been derived by Kawabata[24] and is given by

$$f_3(x)=\sum_0^\infty[2(n+1+1/x)^{1/2}-2(n+1/x)^{1/2}$$
$$-(n+\tfrac{1}{2}+1/x)^{-1/2}].$$

2. Electron-electron interaction

The magnetoresistance due to enhanced electron-electron interaction is a combination of two terms: one known as the diffusion-channel term and the other as the Cooper-channel term. The diffusion-channel (DC) contri-

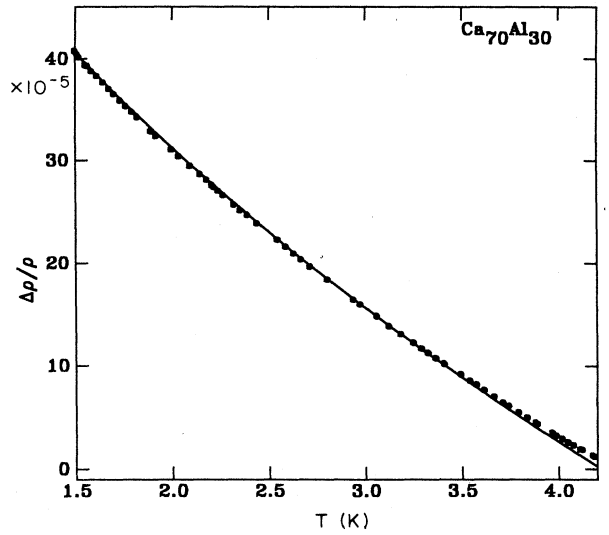

FIG. 3. Low-temperature resistivity of $Ca_{70}Al_{30}$. The points are the experimental data and the solid line a fit as described in the text [Eq. (2)].

bution has been calculated by Lee and Ramakrishnan and is given by[25]

$$\left[\frac{\Delta\rho}{\rho}\right]_{\rm DC}=\rho\frac{e^2}{2\pi^2\hbar}\left[\frac{eB}{\hbar}\right]^{1/2}\frac{\tilde{F}_\sigma}{2}\left[\frac{k_BT}{2eDB}\right]^{1/2}g_3\left[\frac{g\mu_BB}{k_BT}\right], \tag{A2}$$

where

$$\tilde{F}_\sigma=-\frac{32}{3F}\left[1+\frac{3F}{4}-\left[1+\frac{F}{2}\right]^{3/2}\right],$$

$$f=\frac{\int d\Omega\, V[q=2k_F\sin(\theta/2)]}{\int d\Omega\, V(q=0)},$$

$$g_3(x)=\int_0^\infty d\omega\left[\frac{d^2}{d\omega^2}[\omega N(\omega)]\right][(\omega+x)^{1/2}+|\omega-x|^{1/2}-2\omega^{1/2}].$$

F is the average over the solid angle Ω of the statistically screened Coulomb potential. For the Ca-Mg-Al alloys studied here we find, using Thomas-Fermi screening theory,[26] $F\approx 0.52-0.53$.

According to Isawa and Fukuyama, the magnetoresistance arising from the Cooper channel (CC), in the case of nonsuperconductors, is given by[27]

$$\left[\frac{\Delta\rho}{\rho}\right]_{\rm CC}=\rho\frac{e^2}{2\pi^2\hbar}\left[\frac{eB}{\hbar}\right]^{1/2}\times\frac{3\pi^2}{2}\left[\frac{k_BT}{4eDB}\right]^2 g(T,B)\Phi_F(B,T), \tag{A3}$$

where

$$\Phi_F(B,T)=-\sum_{k=0}^{\infty}\left[\zeta\left[\frac{5}{2},\frac{1}{2}+\frac{k+\gamma}{k\gamma}\right]-\frac{2}{3}\left[\frac{\gamma h}{k+\gamma}\right]^{3/2}\right],$$

$$\gamma=\frac{2eDB_i}{\pi k_BT},\quad h=\frac{B}{B_i}.$$

ζ is the generalized Riemann zeta function and

$$g(T,B)=\frac{1}{\frac{2}{F}+\ln\left[\frac{1.13T_F}{T^*}\right]},$$

with

$$T^*=\max\left\{T,\frac{4eDB}{k_B}\right\}.$$

T_F is the Fermi temperature.

[1]E. Abrahams, P. W. Anderson, D. C. Licciardello, and T. V. Ramakrishnan, Phys. Rev. Lett. **42**, 673 (1979).
[2]B. L. Alt'shuler and A. G. Aronov, in *Electron-Electron Interaction in Disordered Systems,* edited by A. L. Efros and M. Pollak (North-Holland, Amsterdam, 1985), Vol. 1.
[3]P. A. Lee and T. V. Ramakrishnan, Rev. Mod. Phys. **57**, 287 (1985).
[4]G. Bergmann, Phys. Rep. **107**, 1 (1984).
[5]J. Bieri, A. Fert, G. Creuzet, and A. Schuhl, J. Phys. F **16**, 2099 (1986).
[6]B. G. Hickey, D. Greig, and M. A. Howson, J. Phys. F **16**, L13 (1986).
[7]J. C. Ousset, H. Rakoto, J. M. Broto, V. Dupuis, and S. Askenazy, Phys. Rev. B **33**, 2799 (1986).
[8]R. Richter, D. V. Baxter, and J. O. Strom-Olsen, Phys. Rev. B **38**, 10421 (1988).
[9]J. Hafner, J. Non-Cryst. Solids **69**, 325 (1985).
[10]T. Matsuda and U. Mizutani, J. Phys. F **12**, 1877 (1982).
[11]U. Mizutani, M. Sasaura, Y. Yamada, and T. Matsuda, J. Phys. F **17**, 667 (1987).
[12]J. Hafner and A. Philipp, J. Phys. F **14**, 1685 (1984).
[13]G. Ravel, J. L. Pastol, J. C. Rouchaud, and R. Fromageau, Metall. Trans. **9B**, 665 (1978).
[14]L. A. Moberly, R. Roshko, and O. G. Symko, Phys. Rev. B **25**, 4695 (1982).
[15]R. W. Cochrane, B. J. Kastner, and W. B. Muir, J. Phys. E **14**, 425 (1982).
[16]G. J. Morgan, M. A. Howson, and K. Saub, J. Phys. F **13**, 2157 (1985).
[17]N. Kumar, D. V. Baxter, R. Richter, and J. O. Strom-Olsen, Phys. Rev. Lett. **59**, 1853 (1987).
[18]J. Rammer, A. L. Shelankov, and A. Schmid, Phys. Rev. Lett.

60, 1985 (1988).
[19]G. Bergmann, Phys. Rev. Lett. **60**, 1986 (1988).
[20]N. Kumar, D. V. Baxter, R. Richter, and J. O. Strom-Olsen, Phys. Rev. Lett. **60**, 1987 (1988).
[21]M. T. Béal-Monod and R. A. Weiner, Phys. Rev. **170**, 552 (1968).
[22]J. Erwin, R. Delgado, A. Armbuster, D. G. Naugle, D. P. Love, F. C. Wang, C. L. Tsai, and T. O. Callaway, Phys. Lett. **100A**, 97 (1984).
[23]H. Fukuyama and K. Hoshino, J. Phys. Soc. Jpn. **50**, 2131 (1981).
[24]A. Kawabata, Solid State Commun. **34**, 431 (1980).
[25]P. A. Lee and T. V. Ramakrishnan, Phys. Rev. B **26**, 4009 (1982).
[26]N. W. Ashcroft and N. D. Mermin, *Solid State Physics* (Holt, Rinehart and Winston, New York, 1976).
[27]Y. Isawa and H. Fukuyama, J. Phys. Soc. Jpn. **53**, 1415 (1984).