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Magnetoresistance of icosahedral Al-Cu-Fe from 80 mK to 80 K

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Icosahedral $Al_{62.5}Cu_{25.5}Fe_{12}$ and $Al_{62.5}Cu_{25}Fe_{12.5}$ samples with resistivities at 4 K of about 4500 and 10 000 $\mu\Omega$ cm, respectively, have been studied. The magnetoresistance $\Delta\rho(B)/\rho$ was measured between 80 mK and 80 K up to 12 T at high temperatures. The results were well described by quantum corrections. The Coulomb interaction parameter F_σ was independently obtained from the low-temperature $\Delta\rho(T)/\rho$, and was found to be in agreement with results from $\Delta\rho(B)/\rho$. The sign change of $d\rho/dT$ with increasing resistivity could be ascribed to an increasing F_σ. The inelastic-scattering time $\tau_{\rm ie}$ saturated at low temperatures with a larger value for the high-resistivity sample. At 80 K, $\tau_{\rm ie}$ was about 10^{-13} s. Hence elastic scattering is intense and quasicrystals are electronically disordered materials. It is pointed out that these quasicrystals may be considered as model materials for the application of three-dimensional quantum-correction theories.

The large magnetoresistance in icosahedral alloys[1,2] is one of several prominent anomalies in electronic properties of these materials. The overall behavior of this phenomenon is understood in terms of quantum interference (QI) effects. However, although detailed analyses have given reasonable descriptions of the observed data, these results may be surprisingly different in details, even for materials of similar composition.[3-5]

The weak-localization (WL) contribution[6] to the magnetoresistance is described by the temperature-dependent inelastic-scattering time $\tau_{\rm ie}(T)$ and the constant spin-orbit scattering time $\tau_{\rm so}$. In the diffusion channel[6] of electron-electron interactions (EEI's), the Coulomb interaction parameter F_σ must be determined. In addition, the resistivity ρ, the diffusion constant D, and the effective electron Landé factor g^* are needed. ρ and D can be taken from other measurements or are fitted to $\Delta\rho(B)/\rho$, and g^* is usually taken to be 2 but is sometimes used as a free parameter.

Recent results illustrate different approaches to these fitting problems for icosahedral Al-Cu-Fe. In Ref. 3, ρ was fitted, in addition to the parameters mentioned above. A constant D was assumed for all samples, which is doubtful, since D varies with varying quantum corrections. An exponent for $\tau_{\rm ie}(T)$ of about $T^{-1.5}$ was obtained for 1.5–40 K and fields up to 9 T. This result was significantly different from an analysis of $\rho(T)$. In Ref. 5 both ρ and D were used as fitting parameters. F_σ was taken from $\rho(T)$ and the Hall effect. To achieve good fits, $\tau_{\rm ie}(T)$ was parametrized, thus forsaking independent information on this important parameter. Reasonable fits to $\Delta\rho(B)/\rho$ were obtained from 4–40 K up to 8 T. However, the fitted D increased with increasing ρ, when instead the opposite trend would be expected. In Ref. 4 all EEI contributions were neglected, which is likely unjustified. g^* was used as a fitting parameter. The observed $\Delta\rho(B)/\rho$ was described moderately well by WL with the improbable value of $g^*=130$.

These results illustrate a flexibility with many parameters in the theory, which is a problem in work on QI effects. In this paper magnetoresistance measurements of two Al-Cu-Fe quasicrystals of different resistivities are described. We aim at more stringent tests of theories by covering a larger range of temperatures and fields than hitherto used. Results are presented from 80 mK to 80 K in fields up to 12 T at high temperatures. Excellent fits were obtained over the full measurement range. The consistency of the fitting procedures is further supported by comparisons with $\Delta\rho(T)$ at low temperatures.

Samples were single-phase icosahedral $Al_{62.5}Cu_{25}Fe_{12.5}$ and $Al_{62.5}Cu_{25.5}Fe_{12}$ which were melt spun and annealed as described previously.[7,8] All peaks in x-ray diffraction were narrow and could be indexed as pure icosahedral phase. Electrical contacts were made to thin samples of surface area of a few mm^2 with silver paint stabilized by epoxy.

Measurements were made at low temperatures in a dilution refrigerator equipped with a superconducting solenoid to 7 T, and above 4 K in a temperature-stabilized inset of a flowing-gas cryostat with a superconducting magnet to 12 T. The small magnetoresistance of carbon thermometers at low temperatures was corrected for.

In the fitting procedures we used expressions for the EEI diffusion channel by Lee and Ramakrishnan,[6] for the WL contribution the Fukuyama-Hoshino results,[9] and for the EEI contribution in the Cooper channel the results of Altshuler *et al.*[10]

It is a delicate question for reliable fitting procedures to handle ρ, D, and the effective g^* factor. Using ρ as a free

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parameter, one can avoid the nontrivial problem to determine the resistivity of small pieces of quasicrystalline samples. However, all quantum contributions to $\Delta\rho/\rho$ are proportional to ρ, and this approach essentially adjusts the magnitude to fit observations. Thus one instead leaves open the question of whether QI can adequately describe observations. For amorphous metals this is an issue, since $\Delta\rho/\rho$ is often observed to be too large for theory.

We relied on measurements of ρ averaged over several samples from the same batch.[7] The estimated accuracy was about 15%. The average results for ρ at 4.2 K were 4.5 mΩ cm for $Al_{62.5}Cu_{25.5}Fe_{12}$ and 10 mΩ cm for $Al_{62.5}Cu_{25}Fe_{12.5}$.

The diffusion constant D was obtained from these ρ values and published results for the specific heat of Al-Cu-Fe quasicrystals with[11] 12 at. % Fe and[12] 12.5 at. % Fe. This gave $D = 0.25$ cm^2/s for $Al_{62.5}Cu_{25.5}Fe_{12}$ and $D = 0.13$ cm^2/s for $Al_{62.5}Cu_{25}Fe_{12.5}$. The samples of Refs. 12 and 13 do not have exactly the same composition as our samples. However, ρ in Al-Cu-Fe quasicrystals is largely governed by the Fe content[13] with values of about 10 000 $\mu\Omega$ cm around 12.5 at. % Fe and 4500 $\mu\Omega$ cm for 12 at. % Fe. Small differences in Al and Cu concentrations may therefore not be serious. This is supported by the similar specific-heat results for[2] $Al_{63}Cu_{25}Fe_{12}$ and[11] $Al_{63.5}Cu_{24.5}Fe_{12}$.

Although some variations in g^* cannot be excluded, a freely varying g^* can again adjust the amplitude of the calculated $\Delta\rho/\rho$. We took $g^* = 2$ in all cases.

In fit (i) to $\Delta\rho(B)/\rho$ these values of ρ, D, and g^* were used and the parameters were τ_{so}, F_σ, and $\tau_{ie}(T)$. The Cooper-channel contribution was neglected. For each sample at each measuring temperature, the best $\tau_{ie}(T)$ was determined by minimizing deviations between observations and calculations for a given (τ_{so}, F_σ). A net of points (τ_{so}, F_σ) was scanned, and the overall rms minimum gave the result for τ_{so}, F_σ, and $\tau_{ie}(T)$.

The measured magnetoresistance is shown by the symbols in Fig. 1 for two Al-Cu-Fe samples. The results of the fit described above are shown by the curves. The constants are given in Table I and $\tau_{ie}(T)$ is displayed in Fig. 2.

Below 4.2 K there were only small variations in F_σ when it was allowed to vary freely. All curves were calculated for a constant value, given in Table I. At 10 K the best fit was obtained for 0.62 and F_σ was negligible for $T \geq 40$ K. It is expected that F_σ vanishes at higher temperatures[14] but details of this temperature dependence are not known.

τ_{ie} is seen in Fig. 2 to saturate around 150–400 ps at low temperatures. Saturation of $\tau_{ie}(T)$ has been observed before[15,16] but not hitherto in quasicrystals. The origin is controversial.[15–17] Our results were obtained from observations and QI theories without any assumption about saturation and τ_{ie} was allowed to vary freely at each temperature.

Between 1.5 and 30 K, $\tau_{ie}(T)$ is similar to previous results.[3,5] The trend in Fig. 2 for $\tau_{ie}(T)$ of the high-resistivity sample is continued to 80 K. Average exponents p in $\tau_{ie}(T) = \tau_0 T^{-p}$ are in the range 1.5–1.8, consistent with the result that τ_{ie}^{-1} for electron-electron interactions in disordered materials should be the sum of two terms with $p = \frac{3}{2}$ and 2, respectively.[18] This average value of p in i-Al-Cu-Fe is different from metallic Si-B

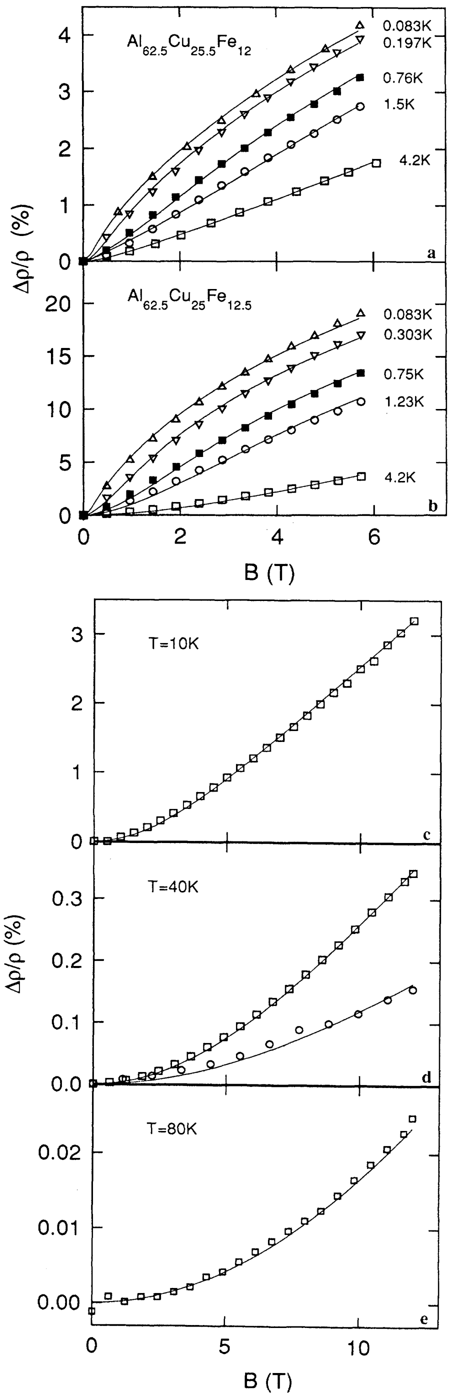

FIG. 1. The magnetoresistance for icosahedral Al-Cu-Fe samples. (a) $Al_{62.5}Cu_{25.5}Fe_{12}$ for $T \leq 4.2$ K, (b) $Al_{62.5}Cu_{25}Fe_{12.5}$ for $T \leq 4.2$ K, (c) at 10 K for $Al_{62.5}Cu_{25}Fe_{12.5}$, (d) at 40 K for $Al_{62.5}Cu_{25}Fe_{12.5}$ (squares) and $Al_{62.5}Cu_{25.5}Fe_{12}$ (circles), and (e) at 80 K for $Al_{62.5}Cu_{25}Fe_{12.5}$. The curves are results from fit (i) described in the text.

TABLE I. Sample properties and results of fits (i) and (iii).

Sample	$Al_{62.5}Cu_{25.5}Fe_{12}$	$Al_{62.5}Cu_{25}Fe_{12.5}$
$\rho(4.2\ K)$ ($\mu\Omega$ cm)	4500	10 000
$\rho(4.2\ K)/\ \rho(293\ K)$	1.61	2.15
	Fit (i)	
D (cm^2/s)	0.25	0.13
τ_{so} (ps)	0.19	1.05
F_σ [from $\Delta\rho(B)$]	0.72	1.16
F_σ [from $\Delta\rho(T)$]	0.71	1.22
	Fit (iii)	
D (cm^2/s)	0.22	0.14
τ_{so}(ps)	0.14	1.13
F_σ	0.72	1.23

close to the metal-insulator transition,[19] where $\tau_{ie}(T)$ was found to vary as T^{-1} from 10 to 0.3 K.

The calculations are the least sensitive to variations in τ_{so} and the error in τ_{so} is therefore the largest, probably a factor 2–3. There is a somewhat stronger variation of τ_{so} between the two samples with a value larger by a factor of 5 for $Al_{62.5}Cu_{25}Fe_{12.5}$. Some variation in τ_{so} between samples is expected due to varying Fermi surface properties. When τ_{so} can be varied by doping with a heavy element in an amorphous metal, a decrease of τ_{so} was found to be correlated with a decrease in F_σ.[20] The trend in Table I may suggest a similar relation also in quasicrystals.

Results for the temperature dependence of the resistivity below 4 K are shown in Fig. 3. For both samples a straight line in $\Delta\rho(T)$ vs $\sqrt{T}$ describes the data well. This result suggests that EEI effects dominate $\Delta\rho(T)$. F_σ was evaluated from the slope of these lines,[6] and was found to be in excellent agreement with values obtained from $\Delta\rho(B)$.

For $Al_{62.5}Cu_{25.5}Fe_{12}$ it can be seen from Fig. 2 that $\tau_{ie}(T)$ is constant up to 1.5 K, i.e., over almost the whole temperature range where $\Delta\rho(T)$ is analyzed in Fig. 3. There is no WL contribution to $\Delta\rho(T)$ in that region. Thus for this sample, the results for F_σ and $\tau_{ie}(T)$ from $\Delta\rho(B)$ and for F_σ from $\Delta\rho(T)$ are all compatible, which gives a high degree of consistency to our analyses. For $Al_{62.5}Cu_{25}Fe_{12.5}$ a WL contribution to $\Delta\rho(T)$ in Fig. 3 cannot be as rigorously excluded, since $\tau_{ie}(T)$ saturates only at about 400 mK. However, if one describes $\tau_{ie}(T)$ by an exponent p, this exponent would depend on temperature in the high-temperature end in Fig. 3, but systematic deviations in a plot of $\Delta\rho(T)$ vs $\sqrt{T}$ are not observed. Therefore a WL contribution would seem unlikely also for this sample. If there is a WL contribution to $\Delta\rho(T)$ of this sample, the resulting F_σ in Table I from $\Delta\rho(T)$ should be regarded as an upper limit.

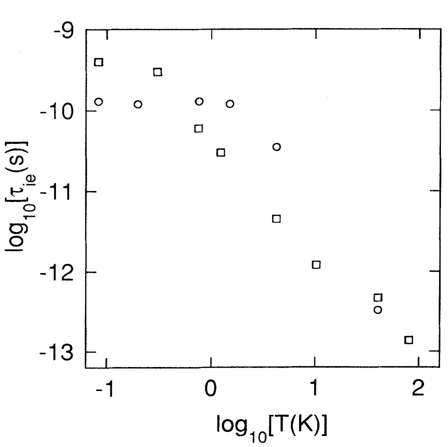

FIG. 2. $\log_{10}\tau_{ie}(T)$ vs $\log_{10}T$ for $Al_{62.5}Cu_{25}Fe_{12.5}$ (squares) and $Al_{62.5}Cu_{25.5}Fe_{12}$ (circles). For both samples and each temperature, $\tau_{ie}(T)$ was calculated by allowing it to vary freely.

The EEI contribution to $d\rho/dT$ becomes positive for $F_\sigma > \frac{8}{9}$.[6] The results for F_σ therefore suggest that $d\rho/dT$ changes sign with increasing ρ in Al-Cu-Fe quasicrystals due to an increasing screening parameter F_σ. With Thomas-Fermi screening $F_\sigma \leq 0.93$. The large value for $Al_{62.5}Cu_{25}Fe_{12.5}$ indicates that screening in quasicrystals cannot be treated in this approximation. Some alternative analyses were made to check the consistency of our results.

(ii) The Cooper channel[10] $\Delta\rho(B)/\rho$ was estimated. The largest correction at 80 mK and 6 T was 4% of the observed $\Delta\rho/\rho$. When these terms were included in a fit to all data, changes in F_σ and $\tau_{ie}(T)$ were found to be insignificant and τ_{so} changed by 10–20%. The Cooper-channel contribution is thus negligible also at our lower temperatures.

(iii) Values of D in fit (i) were checked as follows: D and F_σ were allowed to vary freely in a fit to $\Delta\rho(B)/\rho$, with the constraint that their variations were restricted by the relation between[6] them obtained from ascribing the straight lines in Fig. 3 to EEI. It is seen in Table I that there is good agreement with the results from fit (i). This result demonstrates that our fitting procedures con-

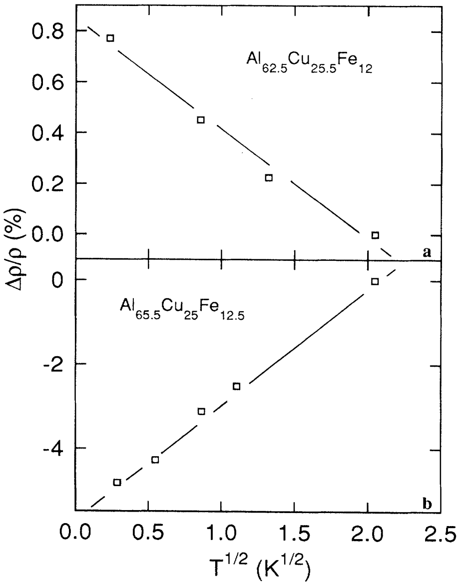

FIG. 3. $\Delta\rho/\rho$ vs $\sqrt{T}$ for (a) $Al_{62.5}Cu_{25.5}Fe_{12}$ and (b) $Al_{62.5}Cu_{25}Fe_{12.5}$. The straight lines are the best fits giving the values of $F_\sigma(\Delta\rho)$ in Table I.

verge to a similar overall minimum point $(\tau_{\text{ie}}(T),\tau_{\text{so}},F_\sigma,D)$, and are not likely to arrest at local minima in the multiparameter space of the variables.

(iv) QI theories give corrections for the conductivity $\Delta\sigma$ rather than $\Delta\rho$. When $\Delta\sigma$ is large it is not obvious that $-\Delta\sigma\approx\Delta\rho/\rho_0^2$ can replace $-\Delta\sigma=\sigma(0,T)-\sigma(B,T)=\Delta\rho/(\rho_0\rho)$. This point has seldom been considered in published work. We calculated $\Delta\sigma(B)$ below 4 K for WL and diffusion-channel EEI. Only for $Al_{62.5}Cu_{25}Fe_{12.5}$ was there a marginal improvement of the quality of the fits and small changes in the fitted parameters, the most important being a decrease of F_σ to 0.99 from 1.16 in Table I and a larger saturation value of 800 ps for $\tau_{\text{ie}}(T)$.

In spite of excellent fits obtained for $\Delta\rho(B)/\rho$, one must ask if there are problems that have not been properly addressed. Two such questions are (a) is $g^*\neq 2$ and (b) has any QI effect in the input parameters been neglected?

Two observations suggest that these objections are not significant in the present case: (a) F_σ was determined from $\Delta\rho(B)/\rho$, with $g^*=2$ and independently from $\Delta\rho(T)/\rho$, where g^* does not enter. The agreement between these two results gives some justification for our assumption about g^*. (b) QI effects in the input parameters that were not corrected for would give a large error when temperature is varied over a larger range. However, deviations between data and calculations are random for $Al_{62.5}Cu_{25}Fe_{12.5}$ at both 40 and 80 K in Fig. 1, and the trend for $\tau_{\text{ie}}(T)$ from lower temperatures is smoothly continued to 80 K in Fig. 2.

Summarizing our results, we note that the measurements of $\Delta\rho(B)/\rho$ cover a large variation of temperatures and fields. For $Al_{62.5}Cu_{25}Fe_{12.5}$ a range of a factor of 1000 in temperature is covered, over which $\Delta\rho(B)/\rho$ at 6 T varies by a factor of 4000. Our fits are almost equally good over this whole range of B and T, with no observable deterioration at low temperatures and high fields where deviations are otherwise often observed.

This point is remarkable, and in contrast to results in three-dimensional amorphous metals, where deviations between observations and calculations in analyses covering a substantial temperature region are regularly observed.[11,21] These difficulties have led to questions about the validity of QI theories.

Observation of a WL contribution to $\Delta\rho(B)$ at 80 K demonstrates that the elastic-scattering time τ must be much shorter than τ_{ie} at that temperature, i.e., $\tau\ll 0.1$ ps. Quasicrystals are therefore electronically disordered materials. Clearly theories for unconventional transport in quasicrystals must take the magnetoresistance into account.

The consistent results for F_σ from $\Delta\rho(B)$ and $\Delta\rho(T)$ are also noteworthy. In amorphous metals discrepancies between differently determined F_σ are well known.[15,22] On the other hand, good agreement was observed between results from $\Delta\rho(T)$ and $\Delta\rho(B)$ in metallic Si-B over a range of boron concentrations.[19]

Our fits of $\Delta\rho(B)/\rho$ for Al-Cu-Fe quasicrystals provide for a verification of WL and diffusion-channel EEI theories in considerable detail. The results also empirically justify that up to 20% in $\Delta\rho(B)/\rho$, interference terms between QI in WL and diffusion-channel EEI are insignificant. As shown by the examples of our successful fits, *i*-Al-Cu-Fe may in fact serve as a model material to demonstrate the precision of these quantum corrections in three dimensions.

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