Electrical resistivity of icosahedral Mg-Al-Zn alloys

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The temperature and magnetic field dependence of the electrical resistivity has been measured on single-phase icosahedral $Mg_{32}(Al_{1-x}Zn_x)_{49}$ at $x=0.5$ and $x=0.69$. For temperatures between 4 and 20 K the observed behavior is found to agree quantitatively with the theories of quantum corrections to electron transport in disordered conductors. By fitting the magnetoresistance measurements to these theories, we can determine the resistivity in a way which is independent of the sample geometry, and we find that it shows a remarkably strong dependence on the alloy composition.

The explosion of interest in metastable alloys exhibiting long-range icosahedral order reflects the fact that this phase is apparently a truly new fundamental form of atomic order, intermediate between a crystal and a glass. Not surprisingly, most published work has been directed towards establishing the exact structure of the phase;[1-4] however, some results have also been presented which deal with its physical properties.[4-6]

To date most measurements of electrical properties in icosahedral phases have concentrated on Al-Mn or U-Pd-Si alloys,[4-6] systems where interpretation is complicated by the presence of open d bands and magnetic effects. The measurement of electrical resistivity in the Al-Mn system is often further impaired by irregular sample geometry and the presence of Al as a second phase, and as a result the resistivity of i-Al_4Mn has been quoted as anywhere from 150 to 800 $\mu\Omega$ cm.[5,6] Recently, Sokoloff[7] has shown that defects, and the strong s-d scattering of the Mn must be included in the model of an ideal icosahedral quasicrystal for the atomic structure of this phase in order to explain these large resistivities.

For this reason it is of considerable interest to investigate the resistivity of icosahedral phases which contain no transition elements, such as the recently reported system $Mg_{32}(Al_{1-x}Zn_x)_{49}$.[8] In this communication we report on resistivity measurements made on samples in this alloy system, with $x=0.5$ and $x=0.69$. We have been able to describe the temperature and field dependence of the resistivity at low temperatures by the theoretical expressions for quantum corrections to the conductivity caused by weak localization and enhanced electron-electron interaction. In so doing we can extract a value for the absolute electrical resistivity independent of the geometrical restrictions inherent in a direct four-terminal measurement.

The alloys were prepared by induction melting elemental constituents in a graphite crucible, under an argon atmosphere. The starting materials were Al and Zn 99.999%, Mg 99.95%. Weight loss during alloying was typically 3% (primarily loss of Zn) and the compositions quoted are the nominal ones. The icosahedral phase was prepared by melt spinning onto a copper wheel at a tangential velocity of 48 m/s. The resulting ribbons were typically 0.5–1.5 mm wide, 10 μm thick, and up to 50 mm long. They invariably possessed rough edges and surfaces, and usually holes or cracks as well. The presence of the icosahedral phase was confirmed by x-ray diffraction, and a typical diffraction pattern is shown in Fig. 1. The observed peaks are indexed according to the scheme used by Bancel *et al.*[2] We note that three of the minor peaks seen in Al-Mn quasicrystals are extinct in the alloys considered here [(111000), (111100), and (110010)]. Ribbons from the same batch were also examined (without further preparation) in a Phillips model EM-300 transmission electron microscope. Our microscopic studies revealed the same features discussed in Ref. 7. Based on our x-ray and microscopic measurements we estimate that any possible crystalline contamination represents no more than 5% of the sample. The microscopic studies also revealed no trace of a possible contaminant amorphous phase. Resistance and magnetoresistance were measured by a four-terminal ac technique to a precision of a few parts in 10^6. Further details will be given in a forthcoming article.[9]

The room-temperature magnetic susceptibilities were

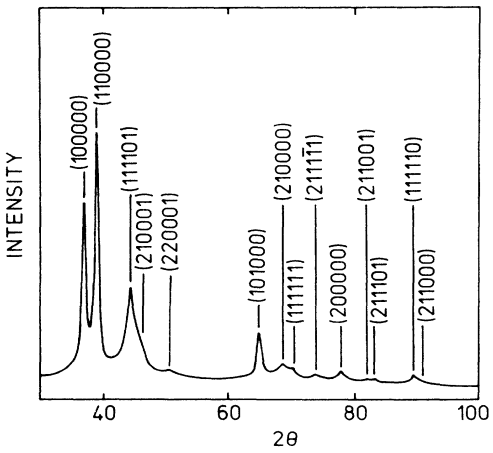

FIG. 1. X-ray diffraction pattern for $Mg_{32}(Al_{1-x}Zn_x)_{49}$ using Cu $K\alpha$ radiation. The indexing scheme used follows that of Ref. 2.

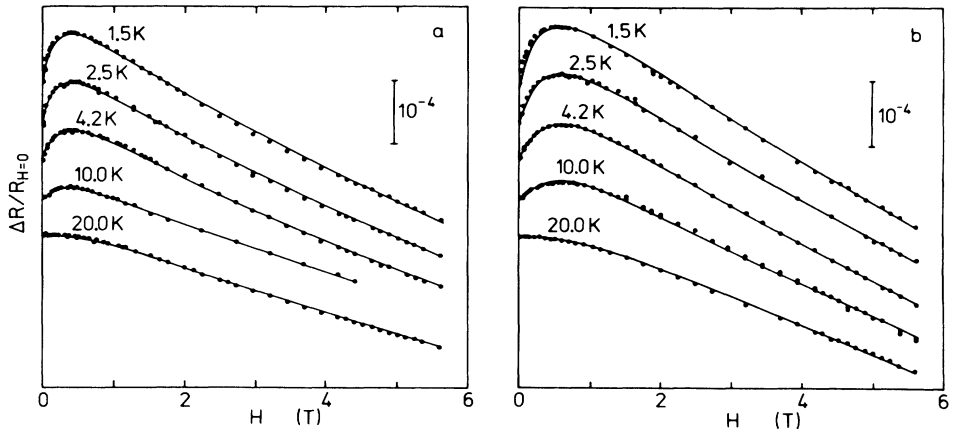

FIG. 2. Low-temperature magnetoresistance of $Mg_{32}(Al_{1-x}Zn_x)_{49}$ for (a) $x=0.5$; (b) $x=0.69$. The points are the measured data and the solid curve is the best fit to Eq. (1). The temperature for each curve is given on the left-hand side of the figure.

measured on an alternating gradient magnetometer to provide some additional information on the electronic structure of these alloys. Values of $3.8\pm0.4\times10^{-6}$ and $-3.4\pm0.2\times10^{-6}$ emu/mole were found for $x=0.5$ and $x=0.69$, respectively. Assuming appropriate values for the elemental core susceptibilities,[10] we obtain estimates for the valence susceptibilities of 8.8×10^{-6} emu/mole for $x=0.5$ and 2.4×10^{-6} emu/mole for $x=0.69$. Over a similar range of composition, the valence susceptibility of amorphous $Mg_{1-x}Zn_x$ changes from 8.7×10^{-6} emu/mole at $x=0.3$, to 7.1×10^{-6} emu/mole at $x=0.4$. Although uncertainty in the values for the core susceptibilities leads to considerable uncertainty in the values for the valence susceptibility (in both the glass and the icosahedral phase), we note that the icosahedral phase shows a significantly stronger compositional variation than does the glass. As with the glass, however, the size of the valence susceptibility of the icosahedral samples is consistent with a substantially *s-p* electron band structure.

Representative magnetoresistance measurements are shown in Fig. 2, along with calculated curves based on the theory of weak localization. According to this theory the relative change in sample resistance, as a function of field, is given by[11,12]

$$\frac{\delta R}{R}=\rho\frac{e^2}{2\pi^2\hbar}\left[\frac{eH}{\hbar}\right]^{1/2}\left\{(\tfrac{1}{2}+\beta)f_3\left[\frac{H}{H_i}\right]-\tfrac{3}{2}f_3\left[\frac{H}{H_2}\right]-\frac{1}{2\sqrt{1-\gamma}}\left[f_3\left[\frac{H}{H_+}\right]-f_3\left[\frac{H}{H_-}\right]\right]\right.$$
$$\left.-\left[\frac{4H_{so}}{3H}\right]^{1/2}\left[\frac{1}{\sqrt{1-\gamma}}[(t_+)^{1/2}-(t_-)^{1/2}]+\sqrt{t}-\sqrt{t+1}\right]\right\}, \quad (1)$$

where

$$f_3(1/h)=\sum_{n=0}^{n=\infty}[2(\sqrt{n+1+h}-\sqrt{n+h})-1/\sqrt{n+0.5+h}]$$

and

$$H_i=\hbar/4eD\tau_i,\quad H_{so}=\hbar/4eD\tau_{so},\quad H_2=H_i+\tfrac{4}{3}H_{so},$$
$$\gamma=\left[\frac{3g\mu_BH}{8eDH_{so}}\right]^2,\quad t=3H_i/4H_{so},\quad t_\pm=t+0.5(1\pm\sqrt{1-\gamma}),\quad H_\pm=H_i+\frac{2H_{so}}{3}(1\pm\sqrt{1-\gamma}).$$

In these expressions τ_i and τ_{so} are the inelastic and spin-orbit electron scattering times, respectively, D is the electron diffusivity, β is the Maki-Thomson parameter related to superconducting fluctuations, and ρ is the resistivity of the material.

Fitting Eq. (1) to the data yields three parameters: the inelastic field H_i, the spin-orbit field H_{so}, and the resistivity ρ.[13] Of these only H_i should vary significantly with temperature. However, to compensate for possible variations in bridge gain, the fit for each temperature was performed with both H_i and ρ allowed to vary. H_{so} on the other hand, was held as a constant value for all temperatures. The results from the fits are summarized in Table I. Above 4.2 K the agreement between experiment and

TABLE I. Parameters obtained from fits of Eq. (1) to normalized magnetoresistance measurements of i-$Mg_{32}(Al_{1-x}Zn_x)_{49}$ alloys. Errors quoted are the purely statistical errors from the fit.

	$x=0.69$		$x=0.5$	
	H_i (mT)	ρ ($\mu\Omega$ cm)	H_i (mT)	ρ ($\mu\Omega$ cm)
T (K)	$H_{so}=94$ mT		$H_{so}=61$ mT	
20.0	261±40	99±4	145±7	62±1
15.0	98±3	100±1	70±3	63±1
10.0	33±0.6	92±1	20±0.3	57±1
7.0	16.7±0.4	96±1		
4.2	14.6±0.2	106±1	4.0±0.1	75±1
3.6	10.4±0.2	107±1	2.8±0.1	76±1
2.5	8.7±0.2	110±1	2.2±0.1	82±1
1.5	6.6±0.2	118±1	1.0±0.1	88±1

theory is excellent and leads to a value of resistivity which is constant to within 10%. Below 4.2 K the overall size of the effect increases due to contributions from interaction effects not included in Eq. (1). Thus the fit returns an artifially large value for ρ, and at the same time the quality of the fit deteriorates.

The values of ρ obtained above 4.2 K are consistent with less precise measurements made from conventional four terminal dc measurements made at room temperature. Thus our best estimates for the resistivities at room temperature are 59±5 and 90±10 $\mu\Omega$ cm for $x=0.5$ and $x=0.69$, respectively (allowing for the decrease in sample resistance observed upon warming from 4.2 to 300 K, 8% for $x=0.69$, and 3% for $x=0.5$). The procedure used to obtain these values is supported by results obtained for amorphous $Mg_{70}Zn_{30}$, where the resistivity found from low-temperature magnetoresistance measurements was within 10% of the conventionally measured value (45 $\mu\Omega$ cm).[9] We note that the icosahedral phase has a higher resistivity, and shows a stronger compositional dependence than does the glass. These observations, along with the strong compositional dependence of the magnetic susceptibility, may reflect a strong energy dependence of the density of states similar to that seen in the $X\alpha$ calculations on icosahedral clusters of Al by McHenry *et al.*[14]

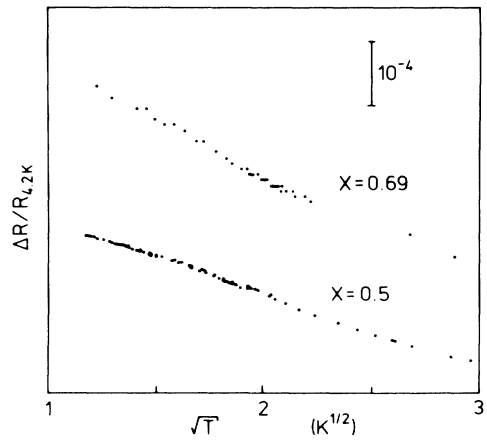

FIG. 3. Low-temperature resistance of $Mg_{32}(Al_{1-x}Zn_x)_{49}$ plotted as a function of $T^{1/2}$. Linear least-squares fit to these data between 1 and 4 K gives slopes of 1.1×10^{-4} for $x=0.5$ and 1.9×10^{-4} for $x=0.69$.

The other parameters listed in Table I are also reasonable. The spin-orbit field scales, within error, to the zinc concentration, as one would expect. Its value is also comparable to that found for amorphous $Mg_{70}Zn_{30}$.[9] The inelastic field, H_i also has a magnitude consistent with that which is seen in $Mg_{70}Zn_{30}$ and shows a temperature dependence somewhat greater than T^2 above 4.2 K, which again is similar to what is seen in other systems.[15]

The temperature dependence of the zero-field resistivity is shown in Fig. 3. The variation below 4 K is close to $T^{1/2}$, as seen in all highly disordered conductors,[16] in agreement with the temperature dependence predicted by the enhanced interaction theory. The magnitude of the $T^{1/2}$ slope is also similar to those seen in metallic glasses of comparable resistivity and so provides further evidence for the importance of quantum corrections due to strong elastic scattering in icosahedral alloys.

In conclusion, we have shown for the first time, consistent quantitative agreement between measurements of the temperature and field dependence of the resistivity of a quasicrystalline alloy, and the theories of quantum corrections to conduction in disordered alloys. Using these results we have been able to estimate the resistivity of such an alloy in a way which is independent of the sample geometry and imperfections which have plagued previous attempts to measure this quantity. The resisitivities so obtained, as well as the measured valence susceptibilities, show a much stronger compositional dependence than would be expected for a free electron system (such as glassy $Mg_{70}Zn_{30}$). Although more work is needed to fully understand these results, we note that they appear to be consistent with a non-free-electron-like density of states such as that seen in calculations of icosahedral clusters of Al.

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[13]In principle, other parameters appear as well: The diffusivity D (independent from its appearance in ρ), and the Maki-Thomson parameter β. In practice, D is unimportant so long as it exceeds 1 cm^2/s. Using the value of ρ found from the fits, we estimate D to be about 5 cm^2/s. β was set to zero since any attempt to vary it produced a small negative (i.e., unphysical) value for the optimal fit.
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