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Magnetic and electrical properties of the single-grained Al–Cu–Fe icosahedral phase

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Abstract. Magnetization of a single-grained quasi-crystal of nominal composition 66.32 at.% Al–20.41 at.% Cu–13.27 at.% Fe was investigated in the temperature range from 2 to 250 K and for magnetic fields below 5 T along fivefold, threefold and twofold axes. No anisotropy was found, which is consistent with the symmetry consideration based on the symmetry of the diffraction spots. Electrical conductivity along the twofold axis showed a minimum at around 20 K and a positive longitudinal magnetoresistance at low temperatures. The conductivity is discussed on the basis of the weak-localization theory.

1. Introduction

Anisotropies in icosahedral quasi-crystals are of particular interest, because the icosahedral symmetry of the diffraction spots implies a new class of symmetry in solids: *an orientational order without long-range translational symmetry*. Several physical properties of single-grained quasi-crystals have been investigated, including the electrical transport properties [1–5]. Some anisotropy has been reported for the electrical properties [2, 3] of decagonal phases.

On the other hand, the symmetry of the icosahedral phase is so high that no anisotropy is expected for second-rank tensors such as the electrical conductivity, if we assume that the tensor has the same symmetry as the intensity of the diffraction spots. In fact, no anisotropy has been reported for the icosahedral phase as yet [1, 4, 5]. The assumption is reasonable but may not be logically inevitable because the symmetry of the diffraction spots does not necessarily correspond to the symmetry in real space [6], although *local* point symmetry [6] exists. More accumulation of symmetry studies may be beneficial.

Anisotropy studies on magnetic properties seem to be still lacking at present. The magnetic susceptibility is a useful experimental quantity to check the anisotropy of small samples, because the direction of the sample can be freely varied and the comparison of the absolute values is meaningful with a lack of such disadvantages as geometrical problems, e.g. length and cross section, unavoidable in electrical resistance measurements.

A single-grained Al–Cu–Fe quasi-crystal of millimetre dimensions was prepared by Ishimasa and Mori [7] and was shown to have a good-quality icosahedral symmetry of F type.

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The weak-localization (WL) theory and the electron–electron interaction theory [8–11] in disordered systems have been stated to have peculiar properties in the electrical conductivity of the polygrained icosahedral phases [12–15], in which the electron–electron interaction effects have been pointed out as being very important at low temperatures. The theories were shown to be useful in deducing important parameters of the transport process from experimental results [10, 11]. Some localization on the quasi-crystal was discussed [16]. The theories might, therefore, be regarded as approximations for the icosahedral phase even if the phase does not have a completely disordered nature but has the ordered nature characteristic of icosahedral symmetry. An experimental study on a single-grained Al–Cu–Fe quasi-crystal may be of much value, because spurious effects arising from bulk or surface crystallinity [12, 17] can be avoided by the use of a single-grained sample.

The present paper reports the magnetic susceptibility along the fivefold, threefold and twofold axes, and it reveals little anisotropy, which is consistent with the symmetry consideration based on the diffraction spots. The electrical conductivity and the magnetoresistance of the single-grained sample were also investigated and showed the WL effect.

2. Experimental procedure

The details of the sample preparation method have been reported in [7]. Materials of nominal composition 66.3 at.% Al–20.4 at.% Cu–13.3 at.% Fe were melted in a plasma jet furnace. The alloy was powdered using an agate mortar. It was annealed at 1143 K for 12 h with a coexisting liquid phase and then slowly cooled to 1095 K with a cooling rate of 4 K h^{-1}. It was successively annealed at the above temperature for 24–50 h, and single quasi-crystals were formed. A single-domain sample was cut from the ingot with a specified direction by the use of the x-ray Laue method. The fivefold Laue pattern is shown in figure 1(*c*). The resulting sample had millimetre dimensions and was 33 mg in mass. The chemical composition of the sample was analysed to be 62.0 at.% Al–22.8 at.% Cu–15.2 at.% Fe using an electron probe x-ray microanalyser with the ZAF (atomic number, absorption and fluorescence) correction. The spots in the electron diffraction patterns shown in figures 1(*a*) and 1(*b*) are very sharp and they are located at the exact positions expected from icosahedral symmetry. The correlation length of this sample was estimated to be greater than 200 nm from the width of the x-ray diffraction peaks. Therefore it is expected that intrinsic phenomena of long-range icosahedral order could be investigated using this sample.

The susceptibility was measured by use of a magnetic balance with a sensitivity of 0.1 μg in a magnetic field H of below 5 T in the temperature region between 2 and 300 K. The sample was fixed in a quartz cell with Apiezon N grease within a few degrees accuracy in the direction. The net magnetization of the sample was obtained by subtracting the magnetic force of the grease in the same cell from the total force. The magnetic force of the cell was measured between 2 and 300 K. The force of the sample was about 15% of the total force.

The electrical resistivity was measured by use of the standard DC four-probe method with a measuring current of 25 mA under a magnetic field of below 8 T in the temperature region between 2 and 220 K. The sample was about 5 mm long with a 0.4 mm × 0.4 mm square section. Gold wires of 50 μm diameter were silver pasted as electrodes. The distance between the two voltage electrodes was short (1.7 mm),

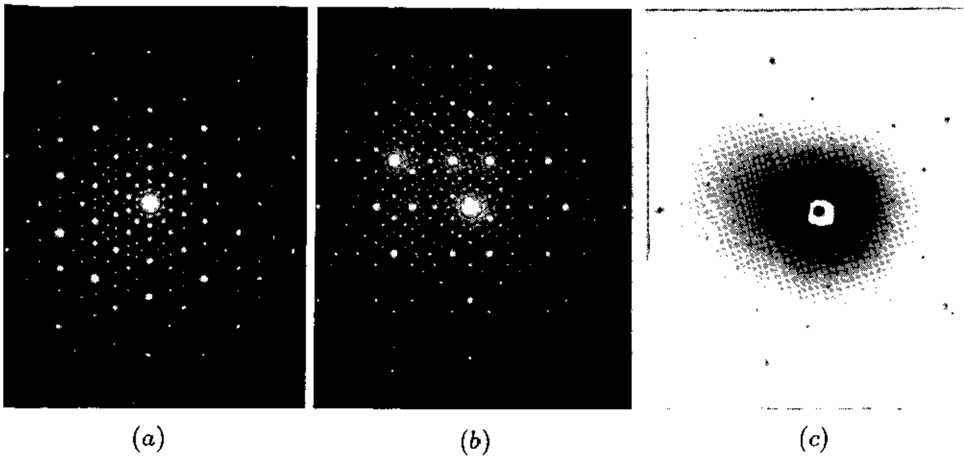

Figure 1. Electron diffraction patterns along (*a*) fivefold and (*b*) twofold axes. (*c*) The x-ray Laue pattern along the fivefold axis.

and the absolute value of the resistance was determined to within an accuracy of only a few tens of per cent, although the relative value concerning the temperature and field dependences was determined to within an accuracy of 0.01%.

3. Magnetic properties

The magnetization σ per gram is shown as a function of the magnetic field H along threefold and fivefold axes in figure 2(*a*). The σ versus H curves are shown along twofold and fivefold axes in figure 2(*b*). The curves along different axes are in agreement with each other. Figures 3(*a*) and 3(*b*) show the magnetic susceptibilities χ measured at the magnetic field of 5 T as a function of temperature T with H parallel to the threefold and fivefold axes and with H parallel to the twofold and fivefold axes, respectively. They are also in agreement with each other. The single-grained quasi-crystal sample shows, therefore, little anisotropy in χ.

We can understand the absence of anisotropy in χ on the basis of the Neumann principle [18], if we assume that χ has the same symmetry as the intensity of the diffraction spots. Let us take a symmetry operation U compatible with the icosahedral point group, and the operation of U should give no change in the susceptibility tensor χ; $U\chi U^{-1} = \chi$. The equality means that χ and U commute. U compatible with the icosahedral group is three dimensional [18] (irreducible), and Schur's lemma gives that χ is a constant matrix. The same conclusion can also be drawn from direct operations on χ of the fivefold rotation along the z axis and the twofold rotation canted by 31.72° from the z axis in the y–z plane. The second-rank susceptibility tensor χ should have, therefore, no anisotropy. The symmetry assumption based on the intensity of the diffraction spots is, therefore, considered to be reasonable, although it may not be logically inevitable.

The insets in figures 3(*a*) and 3(*b*) show σ as a function of $T^{1/2}$, which does not indicate a linear relationship characteristic of the electron–electron interaction effect

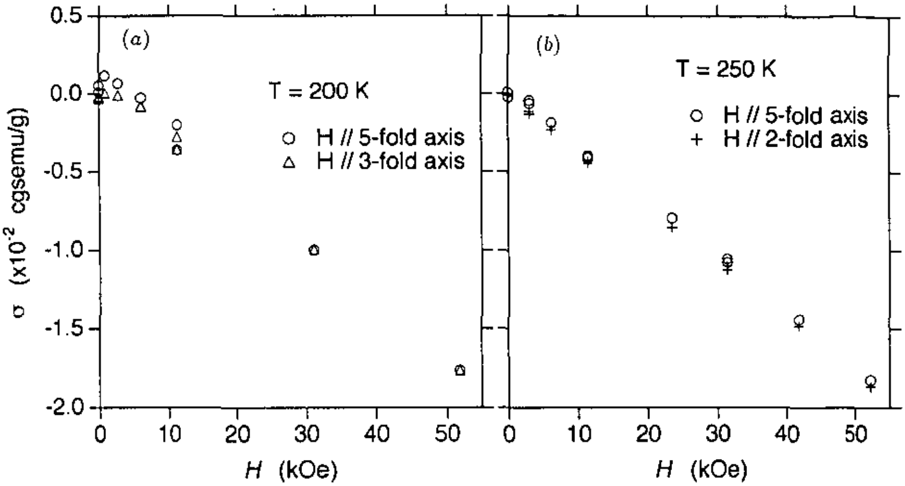

Figure 2. (a) Magnetization curves with H parallel to the threefold and fivefold axes at 200 K. (b) Magnetization curves with H parallel to the twofold and fivefold axes at 250 K.

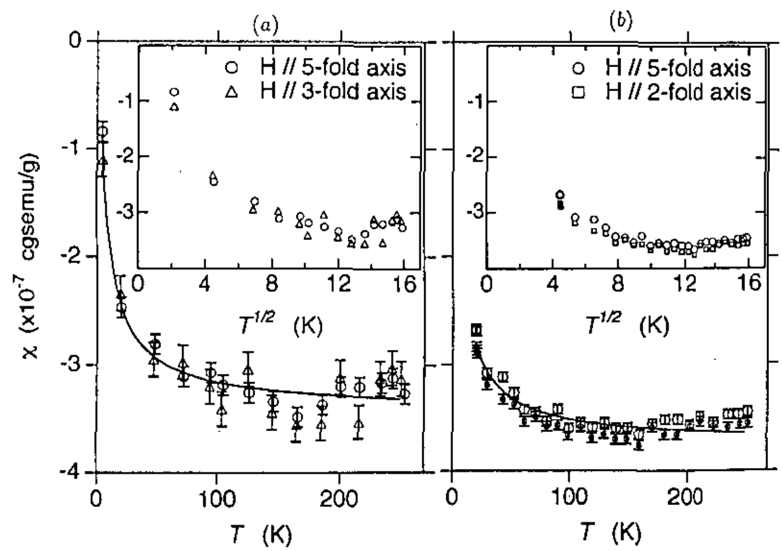

Figure 3. (a) Magnetic susceptibility measured at a magnetic field of 5 T as a function of temperature with H parallel to the threefold and fivefold axes. (b) Magnetic susceptibility measured at a magnetic field of 5 T as a function of temperature with H parallel to the twofold and fivefold axes. The Curie–Weiss fit is shown by the full curves. The error bars show the random error. The data for (a) and (b) were taken in different runs and are difficult to compare directly because of a slight error in the cell subtraction process.

in a disordered system [8–10, 13]. The temperature dependence is rather well fitted to the Curie–Weiss law as shown by the full curves in figure 3. The discrepancy between

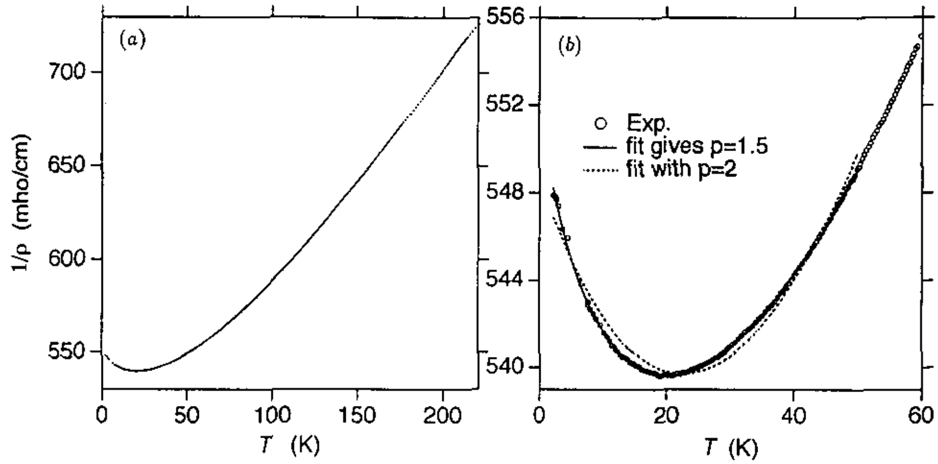

Figure 4. (*a*) Electrical conductivity along the twofold axis as a function of temperature T. (*b*) A fit (——) to the WL theory with spin–orbit and spin–Zeeman effects [20] below 50 K resulting from $p = 1.5$, together with a fit (– – –) with fixed $p = 2$.

the data and fitted curves at high temperatures may be due to the T^2-term [19]. It might be possible that the Curie–Weiss behaviour overcomes the $T^{1/2}$ variation.

4. Electrical conductivity

The conductivity $1/\rho$ along the twofold axis is shown as a function of T in figure 4(*a*). The temperature dependence is similar to that of a polygrained sample [13]. Klein *et al* [13] showed that the one-electron WL theory affords a plausible explanation of the $1/\rho$ versus T characteristic in taking account of the spin–orbit interaction, although the electron–electron interaction effect in a disordered system [8, 9] was interpreted as causing a significant deviation from the one-electron WL theory below 15 K. The full curve in figure 4(*b*) shows a fit below 50 K to the formula of the WL theory with the spin–orbit effect [20]:

$$1/\rho = 1/\rho_0 + A[3(1 + t^p)^{1/2} - t^{p/2}] \tag{1}$$

where $1/\rho_0 = ne^2\tau/m$ is a temperature-independent term of the conductivity, $t = T/T_{so}$, $A = (3^{1/2}e^2/2\pi^2\hbar v_F\tau)(\tau/\tau_{so})^{1/2}$, τ is the elastic relaxation time, τ_{so} is the spin–orbit lifetime, and the parameter T_{so} is defined as $(4c_\tau/\tau_{so})^{1/p}$, where the inelastic lifetime τ_ϵ is assumed to vary as $\tau_\epsilon = c_\tau T^{-p}$. The fit is good and gives the four adjustable parameters as $p = 1.48 \simeq 1.5$, $T_{so} = 83.1$ K, $A = 75.3\ \Omega^{-1}\ \text{cm}^{-1}$ and $1/\rho_0 = 327\ \Omega^{-1}\ \text{cm}^{-1}$. The value of p is close to 1.5 corresponding to the index of electron–electron scattering [11]. A fit with fixed $p = 2$, a mostly accepted value [10], was tried and is shown by the broken curve in figure 4(*b*), which gives poor agreement with experiment. The electron–electron interaction contribution has a tendency to decrease the curvature at low temperatures in relatively low resistivity samples [13, 15], which is unfavourable for a better fit. It is to be noted that the data points in figure 4(*b*) show no deviation from the one-electron WL theory with $p = 1.5$.

The longitudinal magnetoresistance along the twofold axis is shown as a function of H in figure 5(*a*). The result is similar to that for the polygrained sample [13], although the magnetoresistance minimum [13] was not found at 30 K. Klein *et al* [13] argued that the WL theory [20] can also explain qualitatively the magnetoresistance behaviour. The dotted curves in figure 5(*b*) show the quantitative fit based on the formula of the magnetoresistance given by the theory with the spin–orbit and spin–Zeeman effects [20] below 50 K:

$$\Delta[1/\rho(H,T)]/A = h^{1/2}F[(1+t)/h] + 0.5[h/(1-\gamma)][F(t_+/h) - F(t_-/h)] - [1/(1-\gamma)^{1/2}](t_-^{1/2} - t_+^{1/2}) + t^{1/2} - (1+t)^{1/2} \quad (2)$$

where $h = ev_F^2\tau\tau_{so}H/3c\hbar$, $t_\pm = t + 0.5[1 \pm (1-\gamma)^{1/2}]$ and $\gamma = (g^*\mu_B H\tau_{so}/2)^2$. Four parameters ($p = 1.48$) were used. Only one parameter ($g^*\tau_{so}$) was, therefore, adjustable, because an apparently adjustable parameter for the normalization of H is given by the already-determined parameter A and the normal magnetoresistance $\Delta(1/\rho)_n = -(1/\rho_0)(eH\tau/mc)^2$ [21] is negligible on account of the small diffusion constant $D = 0.3$ cm^2 s^{-1} [13] ($\tau = 3Dm/2E_F \simeq 10^{-16}$ s). The fit gives $g^*\tau_{so}$ as 2×10^{-12} s. The fit may be considered fairly good in view of the fact that there is only one adjustable parameter. The fit is good to a similar degree in the case of polygrained Al–Cu–Ru icosahedral alloy after careful removal of surface impurity phases [12]. The values of $g^*\tau_{so}$, A and D [13] give $g^* \simeq 23$, and such enhancement [22] of g^* has been known in semimetals such as Bi which shows a pseudo-gap [23] at E_F as in the case of the Al–Cu–Fe quasicrystal [19, 24].

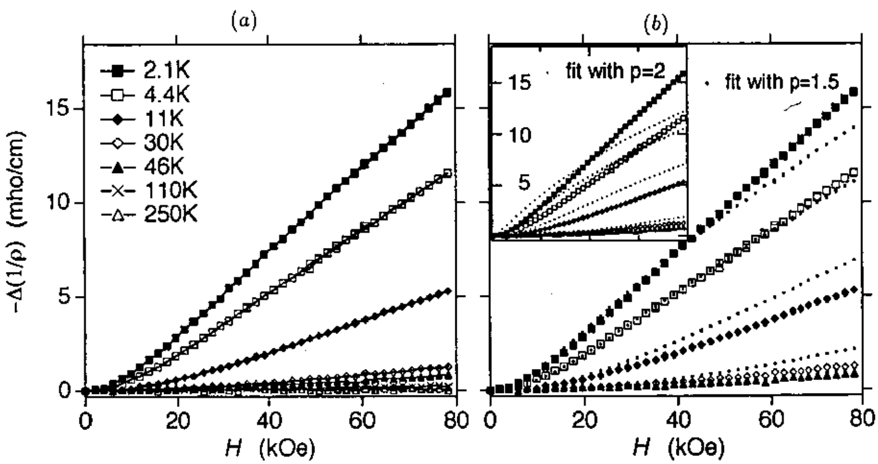

Figure 5. (*a*) Electrical conductivity change $-\Delta(1/\rho)$ along the twofold axis as a function of magnetic field H. (*b*) Experimental $-\Delta(1/\rho)$ versus H below 50 K, together with the fit (······) to the WL theory with $p = 1.5$. The inset shows the same experimental data, together with the fit (······) with $p = 2$.

The inset in figure 5(*b*) shows fits with fixed $p = 2$ as dotted curves, which again give poor agreement with experiment. The value of the power p is a controversial problem in disordered systems [10, 11, 25, 26]. The present fitting scheme apparently supports $p = 1.5$, corresponding to the index of the electron–electron scattering [11], although it may be appropriate to mention the possibility that paramagnetic scattering [9, 27] affects the lifetime in view of a slight (but possibly not negligible in the scattering process) magnetic moment [19] as shown in figure 3. Additionally it might be worth noting that the x-ray diffuse scattering intensity around the diffraction spots might suggest an $\omega \sim q^2$ dispersion relation for longitudinal acoustic phonons [28], which corresponds to $p = 1.5$ for the electron–phonon scattering in a crude approximation $\alpha^2(\omega) =$ constant in the Eliashberg function $\alpha^2(\omega)F(\omega)$ [10].

The temperature and the magnetic field dependences of $1/\rho$ are, therefore, concluded to be indicative of the one-electron WL effects in the single-grained Al–Cu–Fe icosahedral phase, but positive evidence of the electron–electron interaction effects was not found in the present experiment.

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