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Investigation on the synthesis, characterization and electronic behaviour of $Al_{65}Cu_{20+x}Ru_{15-x}$ ($x = 2, 1, 0$ and -1) quasi-crystalline alloys

N P Lalla, R S Tiwari and O N Srivastava
Department of Physics, Banaras Hindu University, Varanasi 221 005, India

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Abstract. The σ–T variation in the quasi-crystalline alloys $Al_{65}Cu_{20+x}Ru_{15-x}$ ($x = 2$, 1, 0 and -1) has been investigated in the temperature range 15 K $< T <$ 300 K (in some cases in the range 4.2–300 K) to make the conductivity versus temperature variation intelligible. The electronic behaviour of the stable quasi-crystal in most of the studies carried out so far has been explained in terms of the theories developed for disordered systems and presumed to hold good for quasi-crystals as well. In the present investigation we have analysed our σ–T data in terms of the power-law localization of electronic states, giving rise to the power-law dependence of conductivity on temperature: $\Delta\sigma \propto T^a$. The applicability of the power law has been demonstrated on the basis of a very good fitting of σ–T data in accordance with $\sigma(T) = \sigma(0) + CT^a$ in a wide temperature range, from 4.2 K through 15 K to 300 K. The exponent a of the power law has been found to decrease with decreasing Ru concentration. A plausible reason for the decrease in the exponent a has been outlined in terms of a decrease in the atomic potential with decreasing Ru concentration.

1. Introduction

The quest for the expected unusual physical properties, particularly for the electronic transport properties arising from the unusual atomic order of quasi-crystals has been persued ever since their discovery [1]. The investigation of the physical properties of quasi-crystalline (QC) systems and their correlation with structural characteristics with particular emphasis on quasi-periodicity is of obvious importance. However, comparatively few studies have been made in this regard. Some of the earlier reports [2–4] on the measurements of transport properties carried out on previously known quasi-crystals were not conclusive. The real effect of the quasi-periodicity was seriously masked by their native content of structural disorder, namely the phason disorder [5], and by the presence of secondary crystalline phases. Because of their thermodynamically unstable nature, it was not possible to eliminate completely these poisoning features by thermal annealing treatments. However, with the discovery [6, 7] of the thermodynamically stable QC systems, e.g. $Al_{65}Cu_{20}Fe_{15}$ and $Al_{65}Cu_{20}Ru_{15}$, it has become possible to obtain highly structurally ordered quasi-crystals without a contaminating crystalline phase and to analyse the effect of long-range quasi-periodic order as well as the icosahedral symmetry on the electronic transport behaviour. The first study on the electrical behaviour of Al–Cu–Ru reveals that the remarkable features of the QC order in these alloys is the anomalously [8] large electrical resistivity (e.g. 30 000 $\mu\Omega$ cm at 4 K in the case of Al–Cu–Ru) of these quasi-crystals. Mizutani *et al* [9, 10] have shown that the ordered version of the QC phase is more resistive compared with the disordered version of the same alloy—a feature contrary to the crystalline case.

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The reason for the occurrence of high electrical resistivities (about 6000 $\mu\Omega$ cm at room temperature for Al–Cu–Fe) in these alloys has been attributed [8] to the reduced density of states at the Fermi level. Recent electronic band-structure calculations [11, 12] of some crystalline approximants of realistic quasi-crystals have also revealed the existence of a pseudo-gap at the Fermi level. The occurrence of the pseudo-gap has been attributed to the strong scattering of electrons by the quasi-lattice. Some experimental evidence [13] of these pseudo-gaps has also been given. Low-temperature (below 20 K) electronic property measurements [8, 14–16] carried out on these stable QC alloys have invariably shown the presence of enhanced quantum interference effects (QIEs). The σ–T data have been well fitted on the basis of weal-localization (WL) effects (including strong spin–orbit scattering effects) even beyond the perturbative limit $(k_F l)^{-1} > 1$. The presence of strong electron–electron interaction (EEI) effects [15] as well as the possibility of a MI transition [14] have also been reported in the case of Al–Cu–Fe. The composition variation studies reveal the presence of a pseudo-gap at the Fermi level and unusual behaviour of the thermopower; the Hall conductivity has been explained in terms of the conductivity spectrum $\sigma(E)$ [17, 18]. An alternative approach for explaining the observed variation in conductivity with temperature of a stable QC-system is in terms of power-law-localized electronic states leading to a power-law variation in σ typified by $\sigma \propto T^a$ [19–22].

In the present paper, we report the study of the effect of a small concentration change in Ru corresponding to $x = 2$, 1, 0 and -1 for the $Al_{65}Cu_{20+x}Ru_{15-x}$ QC system, on its conductivity σ, and the σ–T variation in the temperature range 15–300 K and in some cases down to 4.2 K. It should be pointed out that Ru is a d transition element and hence is likely to influence the electronic behaviour of the Al–Cu–Ru QC system significantly. The variation in concentration of the transition element Ru is expected to affect the conductivity behaviour significantly. However, previous investigations do not embody the studies relating to the variation in Ru concentration. In this regard the present report appears to be the first of its type. Attempts have been made to understand the variation in a with Ru concentration and the σ–T variation in terms of power-law localization of electronic states presumably occurring because of quasi-periodicity.

2. Experimental details

Al–Cu–Ru alloys of composition $Al_{65}Cu_{20+x}Ru_{15-x}$ ($x = 2$, 1, 0 and -1) were prepared by RF induction melting in an argon atmosphere of high-purity (99.99%) elemental constituents in the desired stoichiometric concentrations. To ensure homogenization, each ingot was remelted four to five times. Weight loss on alloying was typically less than 1%. The so-formed ingot was cut into rectangular blocks (5 mm $\times$ 2.5 mm $\times$ 400 μm) and then annealed in a high vacuum (10^{-6}–10^{-7} Torr) in a sealed silica tube for 100–120 h. These annealed samples were then subjected to structural characterization and conductivity measurements. The structural characterization embodied powder x-ray diffraction and transmission electron microscopy (TEM) in selected-area diffraction (SAD), convergent-beam electron diffraction (CBED) and high-resolution electron microscopy (HREM) modes. For the TEM investigation, the sample was prepared by sprinkling the crushed sample specimen onto carbon-coated copper grids.

Figure 1 depicts the powder x-ray diffraction profiles of the annealed samples of different compositions of Al–Cu–Ru quasi-crystals. All the diffraction peaks of the different XRD profiles were successfully indexed [23] on the basis of the face-centred icosahedral (FCI) phase. The structural coherence as determined by the correlation length, measured using

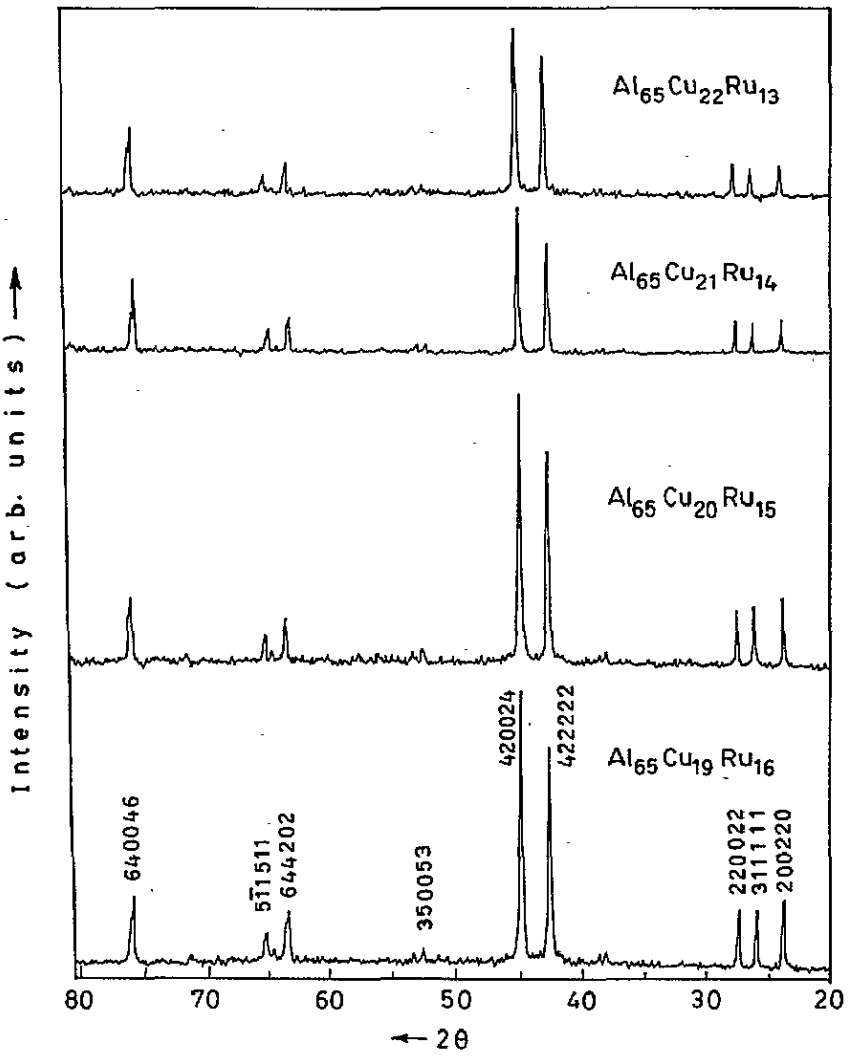


Figure 1. Powder x-ray diffraction profiles of bulk $Al_{65}Cu_{20+x}R_{15-x}$ ($x = 2$, 1, 0 and -1) QC alloys (FCI) prepared by slow cooling of the melt followed by annealing treatment for 100–120 h at 870 °C.

the FWHM of the x-ray diffraction peaks, was found to be better than about 1000 Å. To explore the possibility of the presence of any secondary crystalline phase [24] such as Al_2Cu_3 or Al_2Ru, the TEM investigations of different sample were carried out, in imaging as well as diffraction (SAD and CBED) modes. There was no discernible evidence of the presence of any crystalline phase. The SAD patterns in figure 2 typically represent the characteristic icosahedral symmetry ($m\bar{3}\bar{5}$) and the body-centred-type icosahedral reciprocal lattice observed in these alloys. To probe the local area as well as long-range structural order, the techniques of CBED (with 40 nm probe size) and HREM were employed (figure 3). The presence of the perfect pentagonal symmetry is vividly evident from the CBED pattern in figure 3(*a*). Furthermore unlike the case of the Al–Mn QC system, in the present case very sharp high-order Laue zone lines and associated fine structure clearly indicate the high

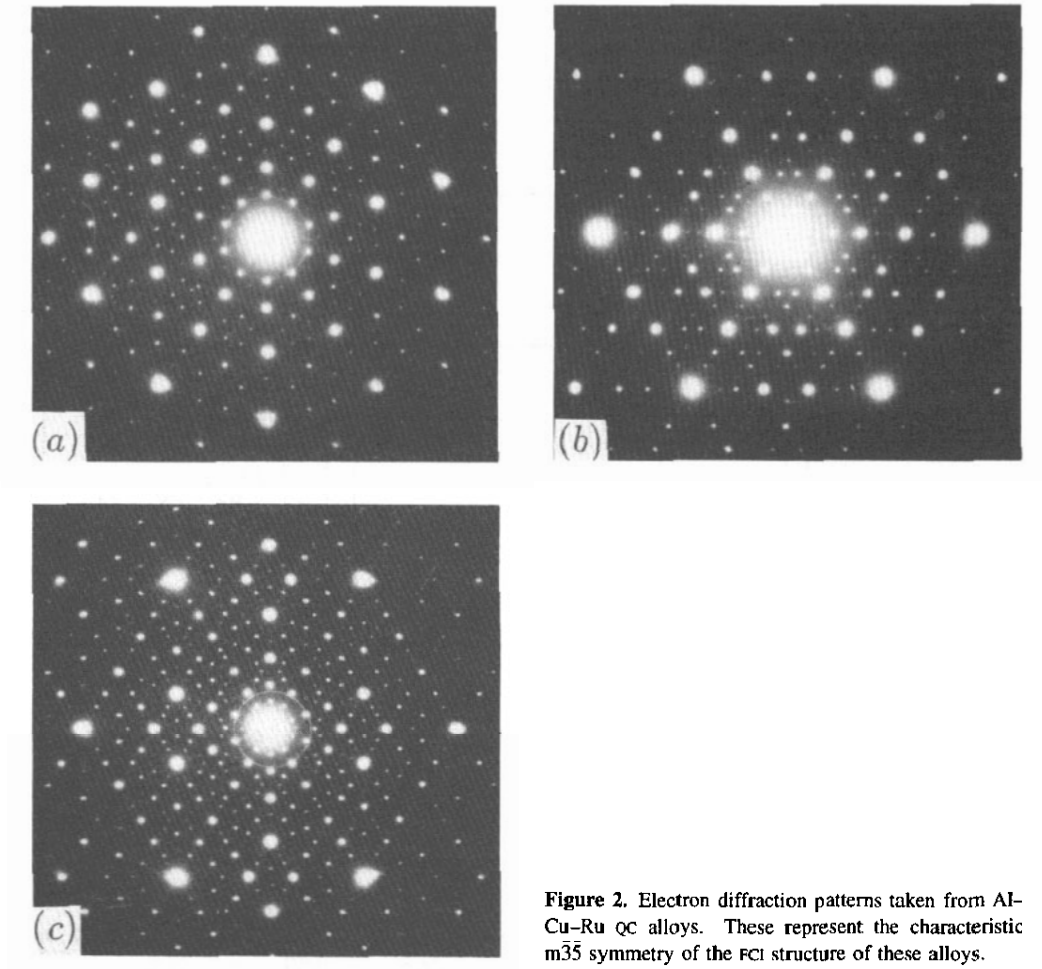


Figure 2. Electron diffraction patterns taken from Al–Cu–Ru QC alloys. These represent the characteristic $m\bar{3}\bar{5}$ symmetry of the FCI structure of these alloys.

structural quality of the samples. It is further evidenced by HREM, exhibiting nearly perfect quasi-periodic order (figure 3(*b*)). Thus the structural characterizations clearly indicate high purity as well as the high structural order of the present QC samples.

Electrical conductivity measurements of all the samples were carried out using the four-probe van der Pauw technique. Electrical contacts on the sample were made using silver glue. Electrical measurements were done employing a Keithley 220 constant-current source (2 mA, at 5.0 V compliances), a Keithley 181 nanovoltmeter, a Keithley 705 scanner and a Keithley 485 picoammeter. Data accumulation was done with an IBM 286-based computer. To achieve a low temperature (about 15 K), an APD helium closed-cycle chiller was used. Measurements at 4.2 K were made using liquid helium. Data were collected during both cooling as well as heating runs.

3. Results and discussion

The results of the resistivity ρ (or conductivity σ) measurements are listed in table 1. It presents room-temperature resistivity (or conductivity) and the ratios R of the resistivity at

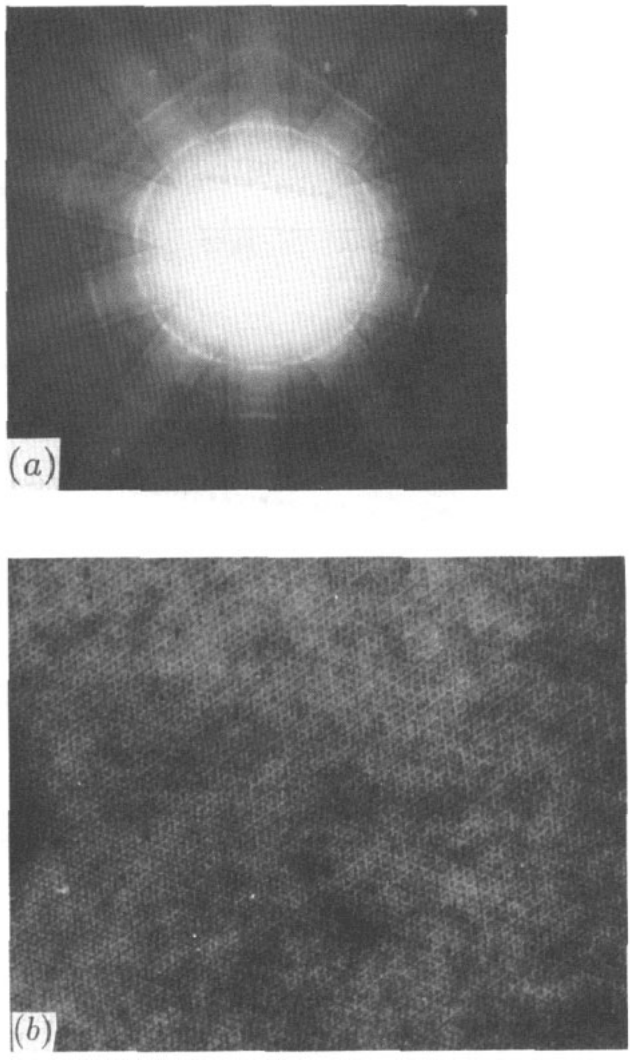


Figure 3. (*a*) CBED pattern taken along the fivefold zone axis of the icosahedral phase. It represents 5m symmetry. (*b*) High-resolution transmission electron micrograph of the FCI phase taken along the threefold symmetry axis.

15 K to that at 300 K for the $Al_{65}Cu_{20+x}Ru_{15-x}$ QC alloys. A systematic variation in the resistivities as well as the ratios R can be observed with the variation in the concentration of Ru and hence of Cu. It is observed that, as the Ru concentration increases, the resistivity as well as the ratio R both increases systematically. The observations of the $\sigma-T$ variations of different QC samples of various compositions in regard to Ru have been summarized in figure 4. It represents the variation in normalized conductivity with temperature. In order to confirm further the nature of the conductivity variation with temperature the nature of $\sigma-T$ in the temperature region below 15 K down to 4.2 K was also monitored. Representative examples of this variation are shown in the inset of figure 4. The salient features of the different $\sigma-T$ curves are as follows.

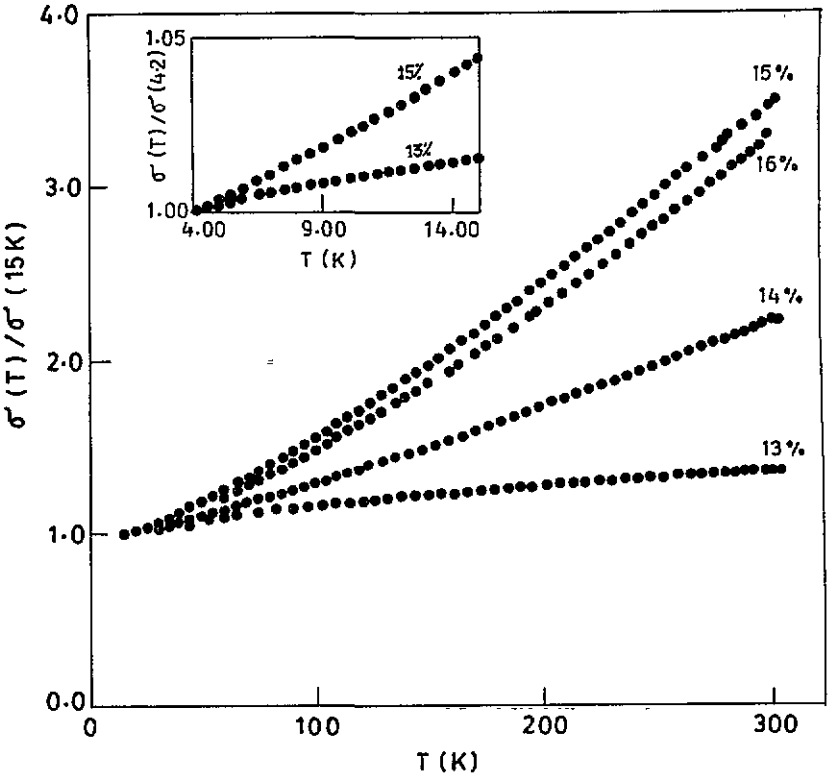


Figure 4. Curves of the normalized conductivity σ versus temperature T for $Al_{65}Cu_{20+x}Ru_{15-x}$ ($x = 2$, 1 0 and -1) QC alloys. The inset shows the normalized conductivity versus temperature variation for $x = 2$ and 0 in the temperature range 4.2–15 K.

Table 1. Results of resistivity measurements.

Composition	Resistivity (300 K) ($\mu\Omega$ cm)	$R = \sigma(300\text{ K})/\sigma(15\text{ K})$	Exponent a in $\Delta\sigma \propto T^a$
$Al_{65}Cu_{19}Ru_{16}$	15 100	3.4	1.35
$Al_{65}Cu_{20}Ru_{15}$	10 100	3.5	1.30
$Al_{65}Cu_{21}Ru_{14}$	6000	2.2	1.24
$Al_{65}Cu_{22}Ru_{13}$	2600	1.22	0.6

(a) One common feature of these curves is that the conductivity increases with increasing temperature (i.e. the σ–T slopes are positive).

(b) The overall slopes of different curves increases with increasing Ru content from 13 to 15 at.% and, at 16 at.%, there is a slight decrease in the net slope.

The nature of the σ–T curve as observed in the present investigation is reminiscent of the conductivity–temperature (σ–T) variation for disordered metallic systems. One significant observation of the σ–T variation is that the ratio $R = \sigma(300\text{ K})/\sigma(15\text{ K}) = 3.5$ for $Al_{65}Cu_{20}Ru_{15}$ which is apparently much larger than any corresponding estimate for disordered metallic systems. A typical value of R for the disordered metallic state is 0.05

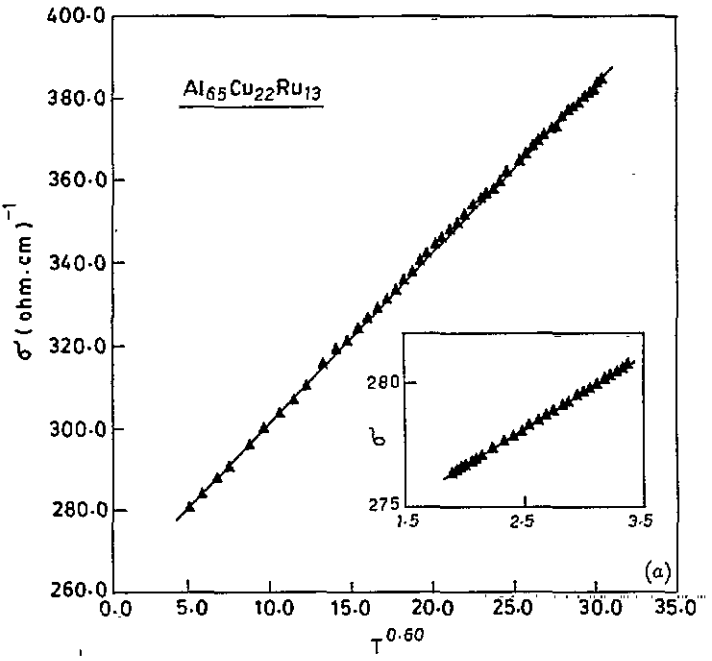
different for different alloy systems (see table 1). It may be pointed out that the variation in Ru concentration, which has been studied for the first time in the present investigation, leads to rather significant changes in room-temperature conductivity and also the ratio R. It may be pointed out that the electronic transport behaviour of stable icosahedral-quasi-crystals has not been successfully explained so far. In most cases [8, 14–16], attempts have been made to explore the σ–T behaviour in terms of QIEs which include the EEI effect and WL effect. Thus for example in the low-temperature region (0.5 K $< T <$ 5 K) the variation in σ with T has been shown to be in accordance with EEI, since the conductivity has been found to be proportional to T. At a comparatively higher temperature (10 K $< T <$ 30 K), the σ–T behaviour has been made intelligible in terms of the WL effect, where σ varies linearly with temperature. However, there are several crucial aspects which are suggestive of the fact that QIEs may not be operating for icosahedral QC alloys.

(i) For all stable icosahedral quasi-crystals, $(k_F l)^{-1} > 1$; it is usually 2. For these conditions, as is known [26–28], the QIEs are not expected to hold good.

(ii) Often the presumed EEI component of QIEs is found to be operative at comparatively high temperatures, about 77 K. For all known disordered metallic systems, the EEI has not been found to extend to such a high temperature. Generally it is found to be present below 20 K [29, 30].

(iii) Since the QIEs i.e. the EEI and WL, are perturbative effects, the change in conductivity should be much smaller than the Boltzmann constant [26]. However, for stable quasi-crystals such as Al–Cu–Ru and Al–Cu–Fe systems the change in conductivity corresponding to these effects turns out to be comparable with the Boltzmann conductivity (see R-values in table 1).

In the present investigation encompassing the σ–T variation of $Al_{65}Cu_{20+x}Ru_{15-x}$ (x = 2, 1, 0 and −1) QC alloys, it appears to be possible to explain the σ–T variation in terms of EEI effects extending up to about 100 K. Again at higher temperatures 100 K $< T <$ 300 K, $\sigma(T)$ varies linearly with T and this seems to suggest the presence of WL effects. However; in view of points (i)–(iii) mentioned above, it is clear that the presence of perturbative effects (QIEs) do not seem to be realistic for stable perfect icosahedral quasi-crystals. The electronic characteristics of these quasi-crystals appear to be beyond the regime of the perturbative effects and the conductivity variation with temperature cannot be explained on basis of concepts advanced for disordered metallic and insulating systems. At this point, we tried to explore alternative mechanisms capable of explaining the conductivity behaviour of the present QC alloys $Al_{65}Cu_{20+x}Ru_{15-x}$ (x = 2, 1, 0 and −1). In this regard the emphasis was laid on electronic characteristics derived in recent years specially for QC systems and not on those developed for disordered metallic and insulating systems which are presumed to hold good for QC systems as well. One such characteristic relates to the nature of electronic states which for the particular case of quasi-crystals has been shown [19–22] to be critical (neither extended not localized). If this is valid, i.e. based on critical electronic states, the conductivity is known to vary according to a simple power law $\sigma \propto T^a$ [19, 31]. We attempted to explore the applicability of this power law to the stable QC alloys $Al_{65}Cu_{20+x}Ru_{15-x}$ (x = 2, 1, 0 and −1) investigated in the present study. A power-law variation in conductivity, namely $\sigma(T) = \sigma(0) + CT^a$, with temperature for $Al_{65}Cu_{22}Ru_{13}$ is shown in figure 5(a). As can be seen from figure 5(a), the power law fits the experimentally observed σ–T variation for all Ru concentrations very well. Very strong evidence for the applicability of the power law in the present case is the fact that this gives quite a good fit throughout the investigated temperature region, 15 K $< T <$ 300 K. It may be pointed out that studies aimed at explaining the temperature variation in conductivity in a wide temperature range 15 K $< T <$ 300 K do not seem to have been carried out so far. Thus



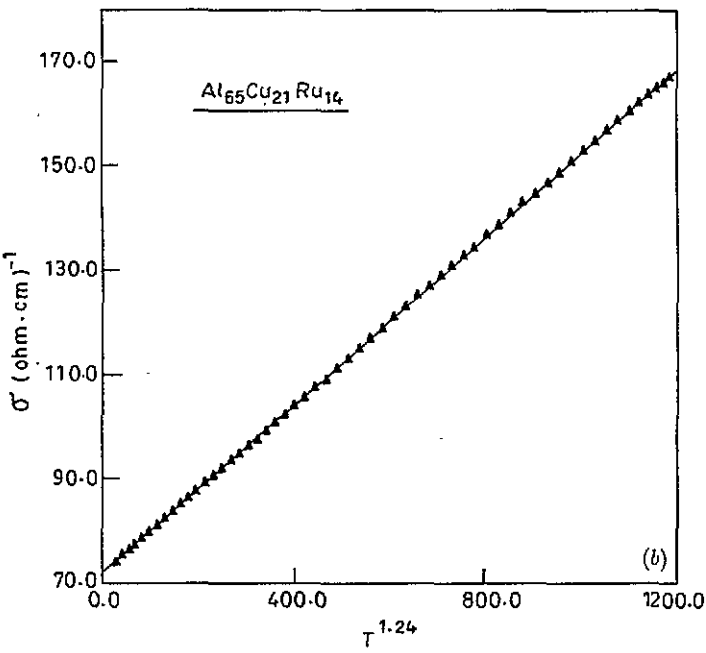


Figure 5. Curves representing the power-law variation in conductivity, $\sigma(T) = \sigma(0) + CT^n$, for (*a*) $Al_{65}Cu_{22}R_{13}$, (*b*) $Al_{65}Cu_{21}R_{14}$, (*c*) $Al_{65}Cu_{20}R_{15}$ and (*d*) $Al_{65}Cu_{22}R_{16}$ QC alloys. The insets in (*a*) and (*c*) represent the power-law variation in the temperature range 4.2–15 K.

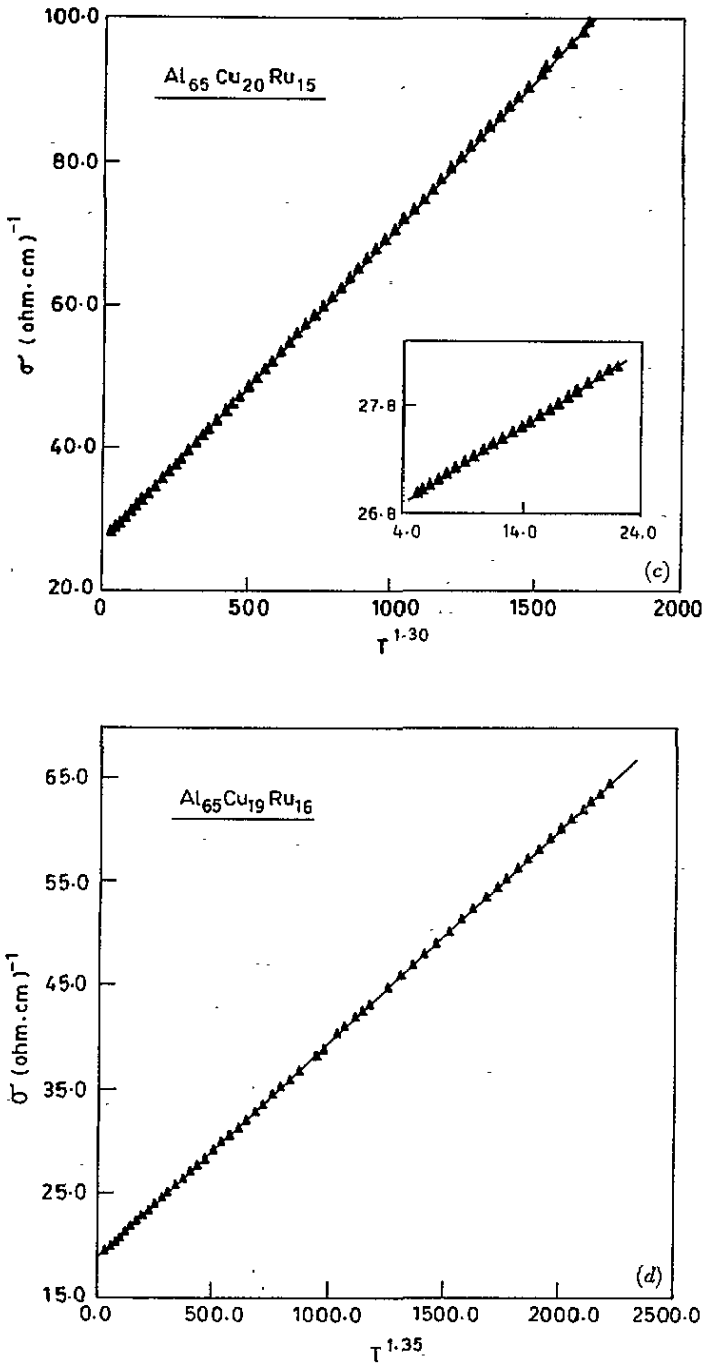


Figure 5. (Continued)

the present investigation embodying the validity of the power law from a low temperature (15 K) to a high temperature (300 K) appears to be one of the first such studies. The power-law fitting was obtained for Al–Cu–Ru alloys with other Ru concentrations as well

(see figures 5(*b*)–5(*d*)). As outlined earlier, the variation in conductivity with temperature in the lower-temperature region (4.2–15 K) was monitored. Representative examples of the power-law variation in conductivity with temperature, following broadly the same pattern (power) are shown in the insets of figures 5(*a*) and 5(*c*). This low-temperature (about 4.2 K) σ–T variation is in accordance with the same power law applicable in the temperature range 15–300 K. This testifies to the validity of the power law for the QC materials, reflecting the influence of quasi-periodicity on the conductivity. The present power-law fitting gives a conductivity variation of the type $\sigma(T) = \sigma(0) + CT^a$. From the σ–T curves in figure 5 it is obvious that $\sigma(0)$ is always finite, indicative of the fact that the $\sigma(T)$ variation does not follow a pure power law, i.e. $\sigma(T) = CT^a$, which is expected from the theory. The origin of the $\sigma(0)$ term appears to be as follows.

$\sigma(T) = CT^a$ is predicted for an ideal QC lattice which does not have any native disorder, but real experimental quasi-crystals may always have some degree of chemical disorder below the detection limit. When the complexity of the matching rule for the growth of 3D quasi-periodic structures is considered, the occurrence of a small degree of chemical disorder is quite expected. The presence of disorder will disrupt the real effect of quasi-periodicity and may act as channels for electronic conduction, giving rise to non-zero conductivity even at 0 K. Since in general at low temperatures the disorder is not affected by temperature, hence this term will nearly always be constant and present additively, giving rise to a conductivity variation of the type $\sigma(T) = \sigma(0) + CT^a$. The power law has been found to be different for different Ru concentrations. It is observed (see table 1) that, as the Ru concentration decreases from 16 to 13 at.%, a in $\sigma(T) = \sigma(0) + CT^a$ decreases from 1.35 to 0.6. As is known [3, 4, 19, 20, 31, 32], the power-law dependence of conductivity with temperature may arise owing to hopping of electrons between critical electronic wavefunctions, decaying as power law, given by $\psi \propto r^{-\beta}$ [19]. This gives the power-law dependence of conductivity on temperature as $\sigma \propto T^a$, $a = 2\beta/d$ where β is the power of decay of the wavefunction; d is the dimensionality of the system. It is generally taken that $\beta < d/2$ or $> d/2$ depending upon whether the wavefunction is predominantly delocalized or localized. Since the scattering potential for a QC system such as Al–Cu–Ru is high, the system is likely to be nearer to the localized situation (Anderson-like localization) and hence $\beta > d/2$, implying thereby that a may be greater than 1.0 [32]. In the present case the a-values giving the best possible law fits for most Ru concentrations (16, 15 and 14 at.%) are greater than unity. Another aspect of the power-law variation in conductivity with temperature in the present observation is the weakening of the exponent from 1.35 to 0.6, with decreasing Ru concentration. This appears to arise because with decreasing Ru concentration the s–d scattering would decrease, causing a net decrease in pseudopotential scattering of electrons. This will decrease the exponent of the power-law localization tendency [19] of the electronic wavefunctions, leading to a decrease in the exponent a. Further, not all the compositions may be compatible with the ideal QC structure; hence the variation in composition will lead to chemical disorder. This would affect the electronic behaviour. It is worth mentioning that, since the deviation of $\sigma(T)$ from linearlity is rather small, a quadratic polynomial $\alpha T^2 + \beta T + \gamma$ which requires the same number of fitting parameters as that for the power-law model could be considered. We attempted to fit the data to the quadratic polynomial as well and compared carefully the fitting error function $\sum[\sigma_f(T) - \sigma_{obs}(T)]^2$ (σ_f is the fitted value and σ_{obs} the observed value). The error function corresponding to the power law was invariably found to be less than that of the quadratic polynomial fitting. It was typically 0.33–0.20 times the value corresponding to quadratic polynomial fitting for different Al–Cu–Ru QC alloys. For

example in the case of $Al_{65}Cu_{20}Ru_{15}$ the error corresponding to power-law fitting is 1.36 and for quadratic polynomial fitting is 6.96.

The above analysis reveals that the power-law fitting is in better conformity with the observed results on the variation in conductivity σ on temperature T than is the quadratic polynomial fitting.

4. Conclusion

In summary it can be said that the electronic properties of stable quasi-crystals are of significant interest and at present not fully understood. In the present study, we have investigated the effect of change in Ru concentration; Ru is a transition metal which plays a significant role in stability and has a profound effect on the electronic behaviour of the $Al_{65}Cu_{20+x}Ru_{15-x}$ ($x = 2$, 1, 0 and -1) stable quasi-crystal. The σ–T variation exhibits an overall positive slope and the resistivity ratios $R = \sigma(300\ \mathrm{K})/\sigma(15\ \mathrm{K})$ are ambiguously large compared with those of the amorphous metals. In some cases, it has been found to be 3.5 (see table 1). In most work carried out so far the low-temperature σ–T variation has been explained on the basis of QIEs, which were originally developed for the disordered systems and have been presumed to hold good for stable quasi-crystals also. However, since these effects are of a perturbative type, their appearance does not seem to be realistic for stable quasi-crystals which have a disorder parameter $(k_F l)^{-1} > 1$. Moreover the resistivity ratios R of 3.5 cannot be explained on the basis of the QIEs. Keeping these facts in view we have attempted to explain the σ–T data for $Al_{65}Cu_{20+x}Ru_{15-x}$ based on the theories developed specifically for quasi-crystals, which predict a σ–T variation of the type $\sigma \propto T^a$, arising owing to power-law decay of the electronic wavefunctions. The genuineness of the power-law dependence of conductivity on temperature has been demonstrated on the basis of a $\sigma(T) = \sigma(0) + CT^a$ fitting of the σ–T data in a wide temperature range from 4.2 K through to 15 K to 300 K. The exponent of the temperature was found to depend on the Ru concentration. Thus it can be said that the net elecrtronic behaviour of the Al–Cu–Ru quasi-crystal lies beyond the regime of QIEs; rather the observed σ–T behaviour appears to be intrinsic to the quasi-periodic nature of its atomic order, giving rise to power-law-localized states. The Ru concentration has a profound effect on its conductivity. The exponent of the temperature in $\sigma(T) = \sigma(0) + CT^a$ decreases with decreasing Ru concentration. This appears to arise owing to the decrease in the exponent of the wavefunction decay with the decrease in the strength of atomic potential in the Ru-deficient Al–Cu–Ru QC alloys.

Acknowledgments

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