Specific-heat and transport properties of alloys exhibiting quasicrystalline and crystalline order

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(Received 25 April 1988)

A systematic study of specific-heat and transport properties of the icosahedral (*I*) and Frank-Kasper (FK) phases of Al-Cu-Li and Al-Cu-Mg alloys was performed. The intrinsic resistivities of some samples were reproducibly derived with use of a method independent of sample microstructure and geometry, based only on superconducting properties. Al-Cu-Li icosahedral alloys were obtained both in thin ribbon and bulk forms. When preparation method and composition were the same, both *I*- and FK-phase Al-Cu-Li showed similar electronic, vibrational, and transport properties. The electronic density of states for Al-Cu-Mg alloys is close to that of the free-electron model while that of Al-Cu-Li alloys is a factor of at least 3 less. Effects of cooling rate and local atomic ordering on the electronic and vibrational properties were observed.

INTRODUCTION

Since the initial discovery of the icosaheral (*I*) phase in a rapidly solidified Al-Mn alloy,[1] there has been a great deal of effort to investigate the properties of the quasicrystalline (qc) phase with the majority of work focusing on the structural nature of these materials. With improved sample quality, various techniques have been used to probe the electronic properties of the *I* phase, which include conductivity,[2] specific heat,[3,4] soft-x-ray emission,[5] and NMR[6] just to name a few. Earlier studies were centered on systems containing transition elements with unfilled *d* bands whose transport properties are dominated by *s-d* scattering. Recent studies[3,7] have concentrated on systems containing only simple metals so that contact with the nearly-free-electron model can be made.

In this paper we present the results of specific-heat, transport, and superconducting measurements on the icosahedral (*I*) and Frank-Kasper (FK) phases of Al-Cu-Mg and Al-Cu-Li alloys. These alloys are selected for several reasons: (1) both the *I* and FK phase of a given composition can be obtained, allowing direct comparison of the effects of crystalline and quasicrystalline order; (2) extensive structural studies by x-ray and electron diffraction, and stability measurements by calorimetry, have been performed to characterize these systems and identify impurity phases, if present; (3) the *I* phase of Al-Cu-Li can be formed by different techniques and over a compositional range which allows observations of effects due to cooling rates and possible local atomic odering on electronic properties; (4) while the *I* phase has been found to be a stable phase in the Al-Cu-Li system, in the Al-Cu-Mg system it is known to be metastable, and of particular interest is whether this stability is reflected in the electronic and vibrational properties of these systems.

EXPERIMENTAL PROCEDURES

Al-Cu-(Li,Mg) alloys were prepared by melting the Al-Cu ingot at an appropriate composition with Li (99.8% purity) or Mg (99.99% purity) pieces in a graphite crucible using an induction furnace in an argon atmosphere. Thin ribbons (1–2 mm wide and 15–40 μm thick, produced by different wheel speeds) were obtained by melt spinning on a rotating 8-in.-diam copper wheel in an inert atmosphere. Structures of samples, which were powdered before x-ray studies, were examined by a Siemen's x-ray diffractometer with Cu $K\alpha$ radiation. Investigations of several *I*- and FK-phase samples were reported earlier.[8]

Electrical-resistivity measurements were performed from 0.35 K to room temperature using the standard four-terminal technique. Measurements were performed on ribbon samples of *I*-phase $Al_{52.4}Cu_{12.6}Mg_{35}$, FK-phase $Al_{52.4}Cu_{12.6}Mg_{35}$ annealed from *I*-phase $Al_{56.1}Cu_{10.2}Li_{33.7}$, and *I*-phase $Al_{55.6}Cu_{11.1}Li_{33.3}$, and on bulk samples of *I*-phase $Al_{60}Cu_{10}Li_{30}$, FK-phase $Al_{56.1}Cu_{10.2}Li_{33.7}$, FK-phase $Al_{55.6}Cu_{11.1}Li_{33.3}$, and FK-phase $Al_{52.4}Cu_{12.6}Mg_{35}$. In the case of FK-phase $Al_{55.6}Cu_{11.1}Li_{33.3}$, measurements were performed on single crystals (millimeter size in all dimensions) extracted from the cast ingot. All compositions for alloys listed are nominal. The actual compositions of the different phases will be discussed. Upper-critical-field measurements were done in longitudinal fields. Experimental details have been presented previously.[7] The specific heat was measured in the temperature range 0.7–10 K using the thermal-relaxation method. The sample holder consisted of a sapphire substrate onto which a thin film of nichrome was evaporated as a heater and an unencapsulated Ge thermometer was attached using General Electric GE-7031 varnish. The sample was mounted on the other side with Apiezon N grease. The substrate was suspended from a copper block by four 3-mil Pt-10-at. % Rh wires which served as electrical leads and also a thermal link. The bare Ge thermometer was calibrated during each run against a calibrated Ge thermometer. The heat capacities of the substrate and N grease were measured separately and subtracted from the value measured with sample. The precision and accuracy of the calorimeter were tested by measuring the specific heat of a 50-mg Cu sample of 99.999% purity which had been annealed at

TABLE I. Specific-heat and transport results for single-phase superconducting samples. Measurements were performed on melt-spun samples. ρ_{meas} are values directly measured and ρ_{calc} are calculated using Eq. (6).

Nominal composition	γ (ergs/g K^2)	Θ_D (K)	T_c (K)	ΔT_c (mK)	λ	$N(0)$ (states/eV atom)	$\left[\frac{dH_{c2}}{dT}\right]_{T_c}$ (kG/K)	ρ_{meas}(4.2 K) ($\mu\Omega$ cm)	ρ_{calc}(4.2 K) ($\mu\Omega$ cm)
FK-phase $Al_{52.4}Cu_{12.6}Mg_{35}$	365±20	342±10	0.81	63	0.34	0.34	−3.3	45	55
I-phase $Al_{52.4}Cu_{12.6}Mg_{35}$	360±20	295±10	0.72	230	0.33	0.33	−2.7	68	65
I-phase $Al_{56.1}Cu_{10.2}Li_{33.7}$	140±15	346±10	1.49	90	0.45	0.11	−1.7	~500–1000	95

650 °C for 5 h and etched in dilute nitric acid before mounting. The measured heat capacity was in excellent agreement with accepted values.[9]

Samples measured in bulk form, ~150 mg, were mounted on the substrate with ~1 mg of N grease for thermal contact. Ribbon samples, ~60 mg, were gently broken and mixed with ~12 mg of N grease before mounting. Internal relaxation times for all samples were less than that of the calorimeter for all temperatures measured, allowing simple reduction of data. The addenda contribution to the total heat capacity was smallest for bulk samples (15–25 %) and largest for ribbon samples (30–85 %), which accounts for the increased scatter in the data for latter samples.

ANALYSIS OF SPECIFIC-HEAT DATA

For samples which were determined by x-ray diffraction to have no detectable second phase, specific-heat data were analyzed in a straightforward manner. The specific heat in the normal state was assumed to be the sum of a linear electronic term and a cubic phonon term,

$$C_n=\gamma T+\beta T^3 , \tag{1}$$

while that of the superconducting state was represented by

$$C_s=\beta T^3+\eta \exp\left[-\frac{1.76T_c}{T}\right]F\left[\frac{T}{T_c}\right] , \tag{2}$$

where β, the phonon term is the same above and below T_c. The second term in (2) is just a convenient form for the superconducting electronic contribution based on the Mühlschlegel calculation,[10] where $F(T/T_c)$ is a multiplicative factor to account for the deviation of the exponential from the Mühlschlegel result and is expressed as a fourth-order polynominal for ease of computation. The coefficient of the exponential term, η, is equal to γT_c for BCS superconductors, but was allowed to vary to optimize the fit. The above expressions were broadened assuming a Gaussian distribution in T_c to account for the width of the observed transitions. Samples which were analyzed by this method include the I and FK phases of $Al_{52.4}Cu_{12.6}Mg_{35}$ and I-phase $Al_{56.1}Cu_{10.2}Li_{33.7}$. The results of these analyses are summarized in Table I.

All remaining samples had varying amounts of second phase (2–15 wt. %), which x-ray diffraction revealed to be essentially fcc Al [the composition of the second phase has also been carefully studied by others[11,12] using transmission electron microscopy (TEM) and scanning electron microscopy (SEM) and found to be fcc Al]. The specific-heat data for these samples were analyzed using a simple expression based on a two-phase system composed of a normal and a superconducting phase, which gives

$$C=[(1-x)\gamma_1+x\gamma_2]T+\beta T^3 , \tag{3}$$

for $T>T_c$, and

$$C=(1-x)\gamma_1+\beta T^3+x\gamma_2 T_c \exp\left[-\frac{1.76T_c}{T}\right]F\left[\frac{T}{T_c}\right] , \tag{4}$$

for $T<T_c$, where

$$\beta=(1-x)\beta_1+x\beta_2 , \tag{5}$$

x is the weight fraction of second phase, β_1 the phonon term for the primary phase, β_2 the phonon term for the secondary phase, γ_1 the electronic term for the primary

TABLE II. Specific-heat results of I and FK phases derived from measurements on alloys with nominal compositions. Estimated compositions of I and FK phases are listed. See text for discussion of resistivity results. Measurements were performed on bulk samples except for melt-spun I-phase $Al_{55.5}Cu_{11.1}Li_{33.3}$.

Nominal composition	Second phase of Al (wt. %)	Phase composition	γ (ergs/g K^2)	Θ_D (K)	$N^*(0)$ (states/eV atom)	ρ_{meas}(4.2 K) ($\mu\Omega$ cm)
$Al_{56.1}Cu_{10.2}Li_{33.7}$	5.1	FK-phase $Al_{54.2}Cu_{10.6}Li_{35.2}$	152±10	485±10	0.15	180[a]
$Al_{55.6}Cu_{11.1}Li_{33.3}$	2.9	FK-phase $Al_{54.4}Cu_{11.4}Li_{34.2}$	152±10	500±10	0.15	200
$Al_{60}Cu_{10}Li_{30}$	13.5	I-phase $Al_{54.5}Cu_{11.4}Li_{34.1}$	127±8	485±10	0.13	175
$Al_{55.6}Cu_{11.1}Li_{33.3}$	1.7	I-phase $Al_{54.8}Cu_{11.3}Li_{33.9}$	70±10	435±15	0.07	~800

[a]Similar values for FK-phase single crystals.

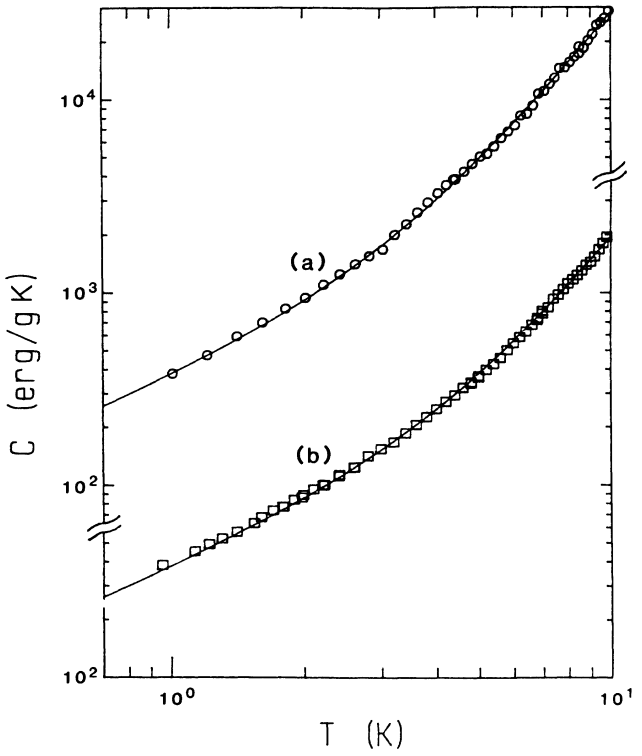

FIG. 1. Specific heat of (a) I-phase $Al_{52.4}Cu_{12.6}Mg_{35}$ and (b) FK-phase $Al_{52.4}Cu_{12.6}Mg_{35}$.

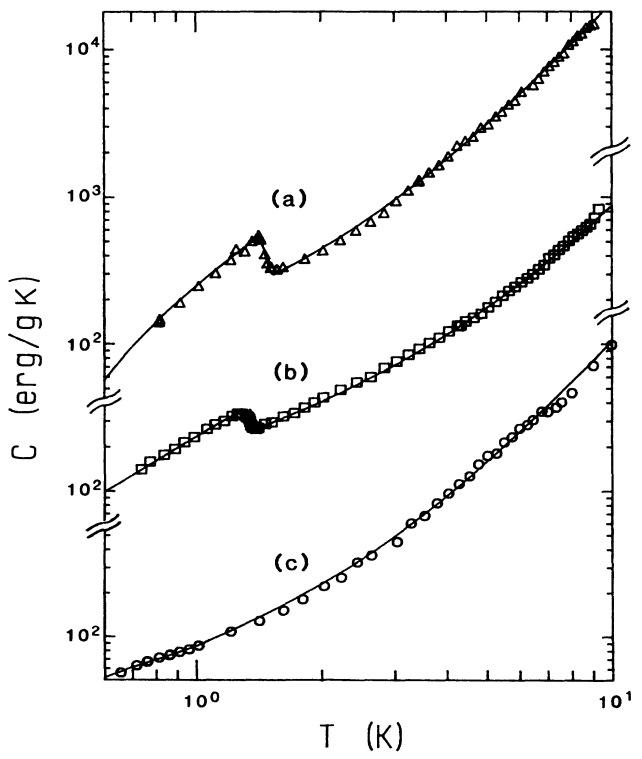

FIG. 2. Specific heat of (a) I-phase $Al_{56.1}Cu_{10.2}Li_{33.7}$, (b) I-phase $Al_{60}Cu_{10}Li_{30}$, and (c) I-phase $Al_{55.5}Cu_{11.1}Li_{33.3}$.

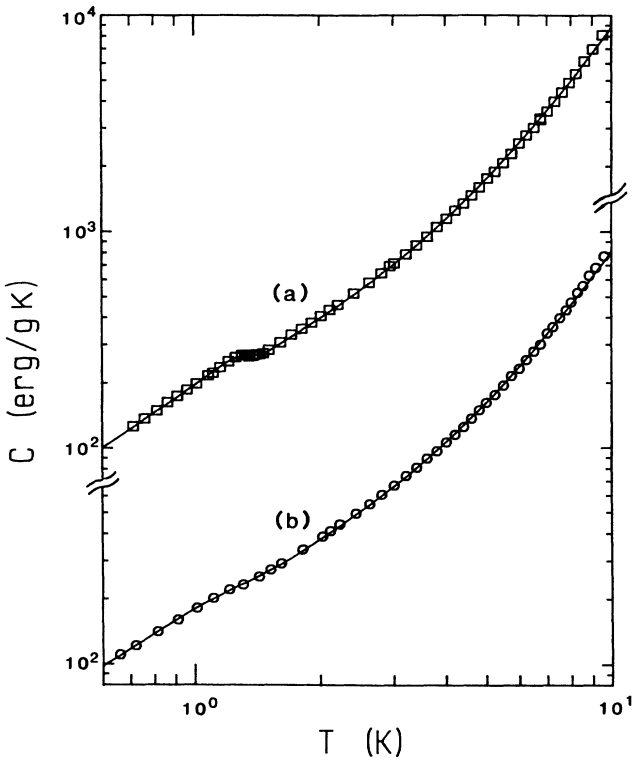

FIG. 3. Specific heat of (a) FK-phase $Al_{56.1}Cu_{10.2}Li_{33.7}$ and (b) FK-phase $Al_{55.5}Cu_{11.1}Li_{33.3}$.

phase, γ_2 the electronic term for the secondary phase, and T_c the superconducting temperature of the secondary phase. The total electronic term, γ_{tot}, and the phonon term β were determined from the linear region of the (C/T)-versus-T^2 plot [Eq. (3)] above the superconducting transition. γ_2 and β_2 were assumed to be that of pure fcc Al (500 ergs/g K^2 and 6460 ergs/g K^4, respectively). The data were then fitted with the constraint that $\gamma_1 = (\gamma_{\text{tot}} - x\gamma_2)/(1-x)$, and γ_1, x, T_c, and ΔT_c were then varied using a nonlinear least-squares-fitting routine to optimize the fit [Eq. (4)].

The renormalized density of states $N^*(0)$ can be calculated from

$$N^*(0) = (1+\lambda)N(0) = 0.4224\gamma_1 , \tag{6}$$

where γ_1 is expressed per mole. For superconductors, where it is possible to determine the electron-phonon–coupling parameters λ from the McMillan equation, one can obtain the bare density of states $N(0)$. The results are given in Table II and the fits are described by the solid lines in Figs. 1–3.

RESULTS

Specific heat

The specific-heat data for the single-phase Al-Cu-Mg alloys are shown only for the region above T_c since the superconducting transitions were not fully resolved due to thermometry limitations. The electronic density of

states (proportional to γ) for the I and FK phases are found to be similar and close to that predicted by the free-electron model. T_c, determined by resistivity, is slightly higher in the I phase (0.81 K) than in the FK phase (0.73 K). This increase in T_c is most likely due to softening of phonon modes in the I phase, which is reflected in the reduced Debye temperature and leads to an enhancement of the electron-phonon–coupling parameter λ, as determined by Θ_D, T_c, and μ^* (with the renormalized Coulomb repulsion parameter set equal to a typical value of 0.13) using McMillan's equation.

The specific-heat data for rapidly quenched single-phase I-phase $Al_{56.1}Cu_{10.2}Li_{33.7}$, Fig. 2(a), shows a well-defined jump at a temperature in good agreement with T_c determined by resistivity measurements (1.49 K), followed by an exponential decrease. The fit, assuming bulk superconductivity, is in good agreement with the data, and the jump in the electronic heat capacity ($\Delta C/\gamma T_c$) of 1.5 is quite close to the BCS value of 1.43. The density of states is found to be quite small, approximately one-third of the free-electron value. The Debye temperature of I-phase $Al_{56.1}Cu_{10.2}Li_{33.7}$ was found to be 350 K. While comparable to Θ_D found in the Al-Cu-Mg alloys, it is significantly lower than that found in any other Al-Cu-Li sample. For the as-cast $Al_{60}Cu_{10}Li_{30}$, x-ray diffraction showed the sample to be predominantly I-phase with approximately 10–15 wt. % second-phase fcc Al. The data are shown in Fig. 2(b). The jump in the electronic heat capacity at ~ 1.34 K was $\Delta C/\gamma T_c = 0.4$ and a substantial linear term was detected below T_c, clearly indicating that only part of the sample was superconducting. The jump in the heat capacity could be accounted for by 13.5 wt. % Al, and the solid line in Fig. 2(b) is the result of the Gaussian-broadened fit using Eqs. (3)–(5). This result suggests that the I-phase crystals within the sample have a composition of approximately $Al_{55}Cu_{11}Li_{34}$, in fair agreement with that determined independently by different techniques.[11,12] Actually, alloys of this composition also contain a trace amount (~ 2 wt. %) of Al_2CuLi Tl phase as revealed by our x-ray data and also according to previous studies.[11,10] The presence of this additional phase, however, does not affect the results reported here. In view of these findings, an almost pure I phase of $Al_{55.6}Cu_{11.1}Li_{33.3}$ was prepared by melt spinning, and the result of the specific-heat measurement is shown in Fig. 2(c). The superconducting transition has almost entirely disappeared with only a small bump in the heat-capacity data around 0.9 K to indicate a minute superconducting phase (also manifested in only a 60% drop in resistivity at the same temperature) which could be accounted for by just 2 wt. % Al, conclusively showing that the I phase of this composition (approximately $Al_{55}Cu_{11}Li_{34}$) is not superconducting. Our result is thus in disagreement with an earlier report on superconductivity in nominally $Al_{60}Cu_{10}Li_{30}$ as-cast alloy (Ref. 13). Θ_D of the rapidly quenched I-phase $Al_{55.6}Cu_{11.1}Li_{33.3}$ was found to be 435 K, while that of the I phase in the as-cast sample is 485 K.

The specific heat of the FK phase was measured for two samples with initial compositions of $Al_{56.1}Cu_{10.2}Li_{33.7}$ and $Al_{55.6}Cu_{11.1}Li_{33.3}$ which x-ray diffraction revealed contained trace amounts of second-phase fcc Al. The results are shown in Fig. 3, along with the fits obtained assuming 5% and 3% second-phase Al, respectively. Subtracting the amount of fcc Al from the initial composition gives an estimated composition of $Al_{54.5}Cu_{11}Li_{34.5}$ for the FK phase in both samples, which agrees well with results obtained in Ref. 12. The renormalized densities of states derived for both samples are identical and quite similar to that derived for the bulk I-phase sample with comparable Θ_D.

Transport

The upper-critical-field data are shown in Fig. 4 for the three bulk superconductors measured. In the dirty limit (see, for example, Wong *et al.* in Ref. 7),

$$N^*(0) = 9.46\times 10^{-7}\frac{M}{\rho d}\left[-\frac{dH_{c2}}{dT}\right]_{T=T_c}, \tag{7}$$

where $N^*(0)$, the renormalized density of states, is in states/eV atom; M, the molar weight, in g; ρ, the resistivity, in Ω cm; d, the density, in g/cm^3; and dH_{c2}/dT is in kG/K. From specific-heat measurements it is possible to obtain $N^*(0)$, and if the initial critical-field gradient is known, then it should be possible to derive the resistivity ρ using the above relationship, independent of sample geometry or presence of microcracks that make the

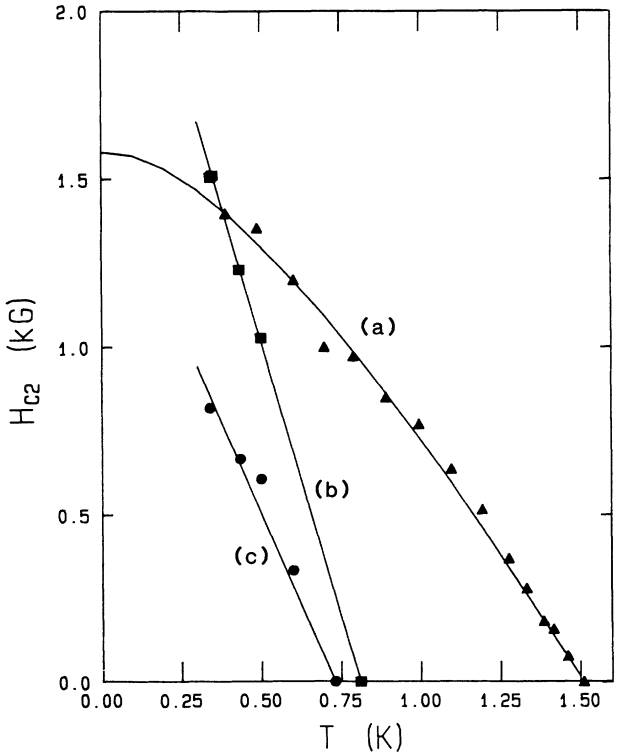

FIG. 4. Upper-critical-field data of (a) I-phase $Al_{56.1}Cu_{10.2}Li_{33.7}$, (b) I-phase $Al_{52.4}C_{12.6}Mg_{35}$, and (c) FK-phase $Al_{52.4}Cu_{12.6}Mg_{35}$. Curve (a) is obtained from the conventional theory of H_{c2} (see Ref. 7, Wong *et al.*). Lines drawn for curves (b) and (c) are guides to the eye.

determination difficult by standard methods. Values of ρ determined by this method can be compared with those obtained by direct measurement using the four-terminal technique. The results of these analysis and resistivity values are listed in Tables I and II.

DISCUSSION

In the Al-Cu-Mg alloys, both *I* and FK phases, were found to be superconducting with a similar density of states (DOS), close to the free-electron value. The Debye temperature of the rapidly quenched *I* phase (295 K) was found to be 25% lower than that of the as-cast FK phase (342 K), a difference which may be largely due to different preparation techniques rather than structural differences, as wll be discussed later in the Al-Cu-Li system, where it is possible to obtain both *I* and FK phases by conventional casting.

In the Al-Cu-Li alloys it was found that composition and preparation methods could greatly affect the electronic, vibrational, and superconducting properties. For alloy phases attainable by conventional casting (estimated compositions of FK-phase $Al_{54.5}Cu_{11}Li_{34.5}$ and *I*-phase $Al_{54.5}Cu_{11.4}Li_{34.1}$), the renormalized density of states (0.13–0.16 states/eV atom) were all similar ($\sim$30% of the free-electron value) with all Θ_D the same, $\sim$490 K. Icosahedral samples prepared by rapid quenching (estimated compositions of *I*-phase $Al_{54.8}Cu_{11.3}Li_{33.9}$ and *I*-phase $Al_{56.1}Cu_{10.2}Li_{33.7}$) showed a large variation in the renormalized density of states (0.07 and 0.16 states/ev atom, respectively) and consistently lower Θ_D as compared with the as-cast samples (435 and 346 K, respectively). *I*-phase $Al_{56.1}Cu_{10.2}Li_{33.7}$, with the largest $N^*(0)$ and lowest Θ_D of any *I* phase of Al-Cu-Li measured, was found to be suerpconducting, while all other samples of Al-Cu-Li remained normal down to 0.35 K. The increased Al content and reduced Θ_D of *I*-phase $Al_{56.1}Cu_{10.2}Li_{33.7}$ are both favorable to superconductivity and are the most probable reasons for the observed superconductivity in this sample. Other workers[12] have observed structural variations in these alloys under different preparation conditions which have been attributed to phason strains, and departure from ideal icosahedral symmetry with increased quenching rates. This is consistent with our observation of decreasing Θ_D, indicating increasing disorder, with higher cooling rates.

The substantial reduction (a factor of 3–6) of the renormalized DOS of the Al-Cu-Li alloys compared with the isostructural Al-Cu-Mg alloys has also been spectroscopically confirmed by soft-x-ray–emission measurements.[5] The transition DOS profile obtained by this method suggests that the bonding in the Al-Cu-Li alloys possessed significant covalent character, possibly accounting for the low DOS. The increased Θ_D (greater than 40%) in the *I*-phase $Al_{55}Cu_{11}Li_{34}$ alloys as compared to the Al-Cu-Mg alloys suggest an increase in the interatomic bonding of these alloys. The high Θ_D values, together with the low DOS, is likely to account for the unusual stability of the *I*-phase crystals in the as-cast $Al_{60}Cu_{10}Li_{30}$ alloy.

Resistivity values derived from Eq. (6) for melt-spun *I*- and FK-phase $Al_{52.4}Cu_{12.6}Mg_{35}$ ribbons are in good agreement with those obtained from direct measurements. The electronic mean free path l_e determined for these phases is $\sim$10 Å. Four-point measurements on bulk *I*- and FK-phase Al-Cu-Li alloys yield room-temperature ρ values of $\sim$220 $\mu\Omega$ cm, which decrease to $\sim$180 $\mu\Omega$ cm at 4.2 K. The variation in resistivity values among bulk samples with different microstructures (e.g., orientation of *I*-phase dendrites with respect to the direction of current flow, and grain–to–sample-size ratio[13]) and amount of Al phase is small. This observation can be explained qualitatively. Measurement of the critical field of the superconducting Al interdendritic regions in $Al_{60}Cu_{10}Li_{30}$ yields a rather high ρ value of ≥ 10 $\mu\Omega$ cm at 4.2 K for the Al phase. This, together with the rather restricted geometry of interdendritic regions, does not results in a reduction of the resistivity of the matrix due to the presence of thin Al layers. Indeed, similar ρ values were also obtained for the $\sim$2-mm single crystals of FK-phase $Al_{55}Cu_{11}Li_{34}$, free of Al phase. Using the density of states determined from specific-heat data, one obtains $l_e \sim 10$ Å. Thus *I* and FK phases of a given composition exhibit essentially similar transport properties. However, for melt-spun *I*-phase samples of composition $Al_{56.1}Cu_{10.2}Li_{33.7}$, ρ determined from critical-field measurements is a factor of 5–10 less than the value obtained by direct measurement. We attribute this observation to the presence of microcracks in the sample. It should be noted that large variations in ρ have also been reported for *I*-phase Al-Mn alloys in the literature. At the present, we cannot explain the apparent preferential presence of microcracks in certain melt-spun alloy systems. Noteworthy is the enhanced value of $l_e \sim 20$ Å in *I*-phase $Al_{56.1}Cu_{10.2}Li_{33.7}$, which greatly exceeds that observed in amorphous simple-metal alloys. The significant differences in transport and vibrational properties between melt-spun *I*-phase $Al_{56.1}Cu_{10.2}Li_{33.7}$ and other *I*-phase Al-Cu-Li phases of lower Al content are not understood.

SUMMARY

We have performed specific-heat and transport measurements on the *I* and FK phases of Al-Cu-Mg and Al-Cu-Li alloys in an attempt to observe the effects of crystalline and quasicrystalline order on the electronic, vibrational, and transport properties of these systems. In the Al-Cu-Mg alloys, the electronic density of states at the Fermi energy of the *I* and FK phases were similar and close to that predicted by the free-electron model. In the Al-Cu-Li alloys it was found that the *I* and FK phases obtained by the same method (conventional casting) and of similar composition had comparable density of states and Θ_D. The density of states for all Al-Cu-Li samples were substantially less (up to $\sim$75%) than the free-electron value, and were sensitive to compositional variations, suggesting possible effects of local atomic ordering.

Θ_D of I-phase Al-Cu-Li was seen to decrease with higher cooling rates, indicating decreasing order, but not necessarily characteristic of icosahedral symmetry since Θ_D of the as-cast I-phase crystals of $Al_{60}Cu_{10}Li_{30}$ (485 K) was found to be the same as the FK phase. The low density of states and high Θ_D of the as-cast I phase of the composition of approximately $Al_{55}Cu_{11}Li_{34}$ may be clues to its unusual stability. In the superconducting I-phase $Al_{56.1}Cu_{10.2}Li_{33.7}$, an electronic mean free path of ~ 20 Å was observed, which is substantially larger than that observed in other icosahedral alloys, and greatly exceeds that found in simple metallic glasses.

ACKNOWLEDGMENTS

The authors thank Y. Shen for useful discussions. This research was supported by National Science Foundation Grant No. DMR-85-12869.

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