PHYSICAL PROPERTIES OF Al-Cu-Fe-Mn ICOSAHEDRAL ALLOYS

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ABSTRACT

The results of Mössbauer spectroscopy, electrical conductivity, and synchrotron-radiation-based resonant photoemission spectroscopy studies of the icosahedral alloys $Al_{62}Cu_{25.5}Fe_{12.5-x}Mn_x$ (x=2.5, 5, and 7.5) are presented. The distribution of the electric quadrupole splittings indicates the presence of the topological/chemical disorder, which is also observed in the temperature dependence of the electrical conductivity. The main features of the valence band are identified with the Fe(Mn) 3d and Cu 3d states. The presence of the theoretically predicted minimum of the density of states around the Fermi level and of their spikiness is not observed within the resolution of the photoemission experiment.

1. Introduction

The icosahedral (i) alloys of high structural quality exhibit some unusual electronic properties.[1] Their most striking features are the low electrical conductivity (σ) that decreases with improved structural quality of the sample, and its generally positive temperature co-efficient. Additionally, σ is extremely sensitive to sample composition. These and other anomalies have been interpreted almost exclusively in terms of a Hume-Rothery mechanism, which implies the existence of a pseudogap in the electronic density of states (DOS) at the Fermi level E_F.

Transition-metal constituents of i-alloys seem to play an important role in determining their electronic properties.[2] In this paper we report the first investigation of the electronic properties of the i-alloy $Al_{62}Cu_{25.5}Fe_{12.5}$ in which the Fe atoms are partially replaced by the Mn atoms.

2. Experimental

Ingots of nominal composition $Al_{62}Cu_{25.5}Fe_{12.5-x}Mn_x$ (x=2.5, 5, and 7.5) were produced by melting high-purity source elements in an argon atmosphere. They were used to produce ribbons by melt spinning. The x-ray diffraction studies showed[3] that the x=2.5 and 5 samples contained a few % of the second phase, whereas the x=7.5 sample was a mixture of the second phase and i-phase in approximately equal proportions.

Mössbauer measurements were performed at room temperature using a standard spectrometer. Thin Mössbauer absorbers were used.[3] Electrical conductivity measurements were conducted with a standard dc four-probe method between 10 K and room temperature. Photoemission spectra were collected at room temperature on beam line U14A at the National Synchrotron Light Source at the Brookhaven National Laboratory. They were corrected for various instrumental factors, as described elsewhere.[1,3]

3. Results and Discussion

A typical Mössbauer spectrum (Fig. 1) consists of two broad and structureless lines and is very similar to the spectra of non-magnetic metallic glasses. Its broad lines are caused by the presence of the distribution of the electric quadrupole splittings $P(\Delta)$.[4] The spectrum in Fig. 1 could be fitted successfully with two components: a doublet due to the presence of $P(\Delta)$, which is shown in Fig. 2, and a doublet due to the presence of a small amount of impurity.
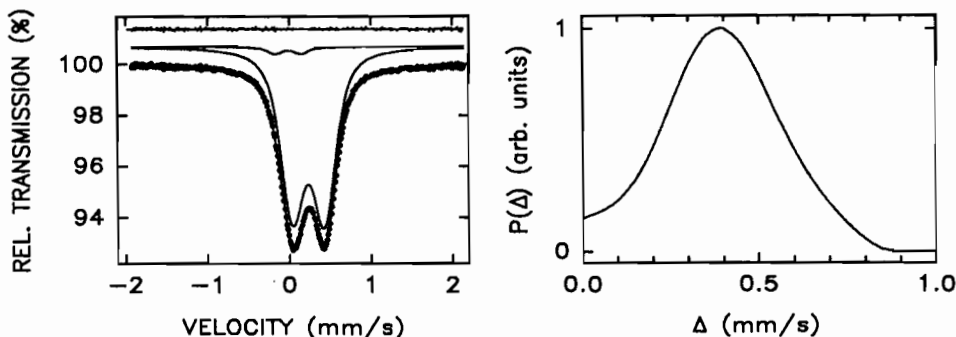

Fig. 1 (left): ^{57}Fe Mössbauer spectrum of i-Al$_{62}$Cu$_{25.5}$Fe$_{10}$Mn$_{2.5}$ fitted with a distribution of the electric quadrupole splittings (doublet) and an impurity contribution (doublet). The difference between the spectrum and the fit is also shown.

Fig. 2 (right): Distribution of the electric quadrupole splittings corresponding to the main doublet component in Fig. 1.

The distribution $P(\Delta)$ observed here (Fig. 2) and in other i-alloys of high structural quality, which exhibit only resolution-limited Bragg-peak widths,[3,4] proves that there is a chemical and/or topological disorder even in the "perfect" quasicrystals. Such a disorder can be expected to influence other physical properties.

The temperature dependence $\sigma(T)$ for the studied alloys (Fig. 3) is similar to that observed for the i-Al-Cu-Fe alloys.[5] A significant amount of a second phase can explain the larger $\sigma(T)$ values for the x=7.5 sample as compared to the values for the x=2.5 and 5 samples. As is similar to the situation in the i-Al-Cu-Fe alloys,[5] the $\sigma(T)$ curves for the x=2.5 and 5 samples are parallel.

Below about 200 K, the temperature dependence $\sigma(T)$ can be well fitted to the formula based on quantum interference effects.[3] These effects result in changing the slope of the curve in Fig. 4 from about 0.5 (electron-electron interaction effect) to about 1.0 (weak localization effect). The applicability of these effects to the studied alloys is consistent with the presence of some disorder inferred from the Mössbauer data.

Typical valence bands consist of two broad features (Fig. 5) located at the binding energy (BE) of about -0.6 and -3.8 eV. Resonant photoemission spectra (Fig. 5) indicate that, as the photon energy $h\nu$ increases, the relative intensity of the peak at BE=-0.6 eV with respect to the peak at BE=-3.8 eV decreases first, reaches its minimum at $h\nu$=53 eV, and then starts to increase for higher values of $h\nu$. The suppression of the BE=-0.6 eV feature at $h\nu$=53 eV indicates that it is mainly of the Fe $3d$ character (the Mn $3d$-like states also contribute to this feature). This is confirmed by the resonance photoemission spectra measured in the so-called constant-initial-state[1] (CIS) mode: the largest resonance occurs

(curve A in Fig. 6) for the initial energy corresponding to the position A in Fig. 5. The feature located at BE=−3.8 eV is mainly of the Cu 3d character.
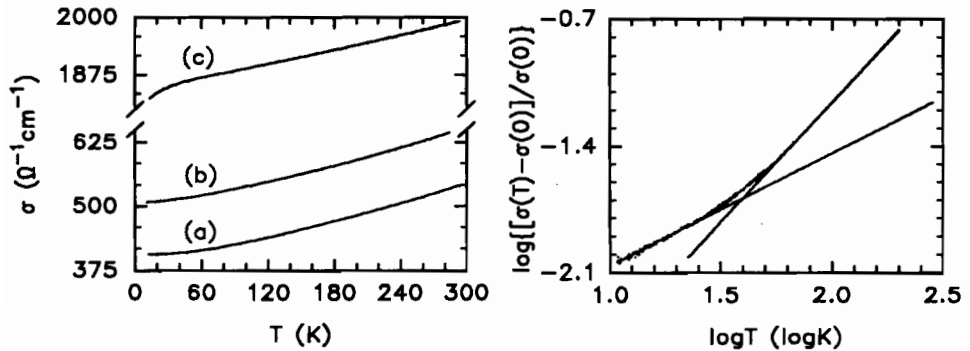
Fig. 3 (left): Temperature dependence of the electrical conductivity of i-Al$_{62}$Cu$_{25.5}$Fe$_{12.5-x}$Mn$_x$ with (a) x=5, (b) x=2.5, and (c) x=7.5.
Fig. 4 (right): Temperature dependence of the normalized conductivity relative to that at 0 K on a log-log scale of the x=2.5 sample. The solid lines of different slope are the fits to the T^n dependence.

Since there are no theoretical DOS calculations for the approximant of the i-Al-Cu-Fe-Mn alloys, we compare the valence band of the x=2.5 sample with the lowest Mn concentration with the theoretical valence band (Fig. 7) which is based on the theoretical DOS calculated for an approximant of the i-Al-Cu-Fe alloy.[2] The calculations predict (Fig. 7) the presence
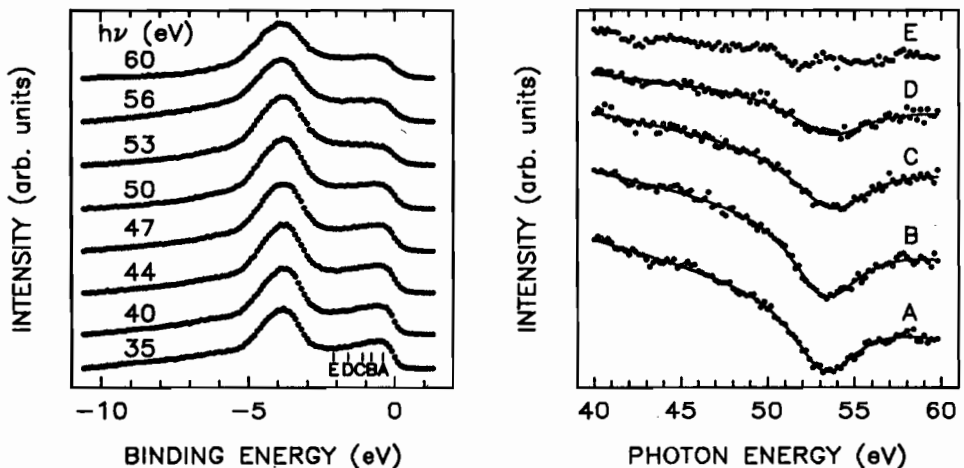
Fig. 5 (left): Valence bands of i-Al$_{62}$Cu$_{25.5}$Fe$_{10}$Mn$_{2.5}$ measured for different photon energies around the Fe 3p →3d transition. A, B, C, D, and E identify positions for which CIS spectra were measured (Fig. 6).
Fig. 6 (right): CIS spectra measured for the valence band positions A, B, C, D, and E in Fig. 5, which are identified here by the same latters. The solid line is a fit to a Fano profile and a linear background.

of a pseudogap with a width of about 0.5 eV whose center is located at about 0.3 eV *above* E_F and of many fine spiked peaks. In order to make a meaningful comparison between an experimental and a theoretical valence band, the latter has to be appropriately broadened to account for the lifetime broadening effects inherent to the photoemission spectroscopy technique and for the finite resolution (0.4 eV) of a photoemission experiment.[1] Such a comparison (Fig. 8) shows that the positions of the two main features due to the Fe and Cu 3d states are reproduced relatively well by theory. No pseudogap at E_F is observed in the valence band (Fig. 8). This may be due either to the insufficiently high energy resolution of the experiment[1] or to the possibility that such a pseudogap does not exist. It should be stressed that the low σ values do not necessarily imply the minimum of DOS(E_F). The theoretically predicted spikiness in the DOS is not observed. If it exists, it can be detected only on the vicinity of E_F and would require a photoemission experiment of the highest possible energy resolution.[1]
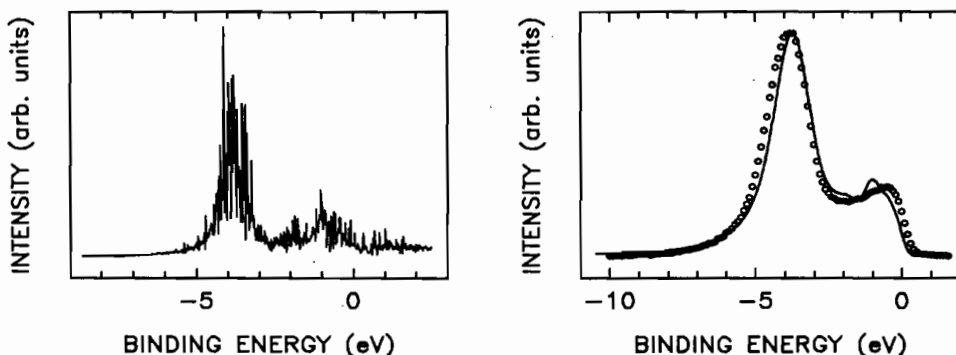

Fig. 7 (left): Theoretical $h\nu$=100 eV PES valence band of an *i*-Al-Cu-Fe alloy obtained by summing the theoretical partial Al, Cu, and Fe DOS associated with different angular momenta from Ref. 2 weighted by the corresponding photoionization cross sections and by the composition and the number of electrons, as described in Refs. 1 and 3.
Fig. 8 (right): Comparison between the valence band of *i*-Al$_{62}$Cu$_{25.5}$Fe$_{10}$Mn$_{2.5}$ measured at $h\nu$=100 eV (o) and the broadened theoretical valence band (solid line) from Fig. 7, as descibed in the text.

4. Acknowledgements

5. References

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