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LETTER TO THE EDITOR

The structural transition and magnetism of the $Al_{65}Cu_{20}Fe_{15}$ quasicrystal

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Abstract. $Al_{65}Cu_{20}Fe_{15}$ is a thermodynamically stable quasicrystal. There are two kinds of quasicrystal present in such a material: 3D quasicrystal—icosahedral (I phase) and 2D quasicrystal—decagonal (D phase). The I phase is dominant in rapidly quenched samples and exhibits Pauli paramagnetism. For annealed samples, however, the D phase becomes dominant and exhibits diamagnetism. Macrostructural transitions from the I phase to the D phase during annealing were observed by SEM. The results of UPS experiments show a gradual increase in Fermi energy, implying that the unfilled energy level of Fe is being filled by conduction electrons during such a structural transition. This causes the magnetism to change from Pauli paramagnetism to diamagnetism.

Since the quasicrystal Al_6Mn was first identified in 1984 by Shechtman *et al* [1], several kinds of quasicrystalline structure have been discovered, for example icosahedral, decagonal, dodecagonal and octagonal structures. Some physical properties, in particular magnetic properties, have since been studied in many quasicrystals [2].

The stable $Al_{65}Cu_{20}Fe_{15}$ quasicrystal was first reported by Tsai and co-workers [3] in 1987, and was the I phase in rapidly quenched $Al_{65}Cu_{20}Fe_{15}$. However, He and co-workers [4] indicated that a D phase also existed in this sample. Some studies [5] of its magnetic properties showed it to be paramagnetic, while others [6] found it to be diamagnetic. There should be a close relation between the structure and magnetic properties of the quasicrystal. This work attempts to ascertain whether this 'stable' quasicrystal undergoes any relevant structural changes during heat treatment and the implications for its magnetic properties.

The $Al_{65}Cu_{20}Fe_{15}$ ingot was prepared by arc melting in an Ar atmosphere of high-purity Al, Cu and Fe. Rapidly quenched specimens were obtained by the melt spinning technique. Both ingot and quenched specimens were annealed at 1098 K for 2, 3, 4, 5, 6, 7, 8, and 10 days in an evacuated quartz tube. The structural transitions of these samples were monitored by x-ray diffraction and differential thermal analysis (DTA). Transmission electron microscopy (TEM) was carried out on a Phillips EM400 electron microscope for the as-quenched sample and the sample annealed for 8 days.

Scanning electron microscopy (SEM) was carried out using a TEOL JSM-35CF electron microscope. The morphology patterns of the as-quenched and annealed samples were observed. The magnetic properties were analysed by an extracting sample magnetometer. The ultraviolet photoemission spectra (UPS) were also obtained.

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There were no indications of phase transitions occurring from the results of the DTA and x-ray diffraction, but this does not necessarily mean that no structural change has taken place—some structural changes can involve a long relaxation process. The results of the electron diffraction show that there are two quasicrystalline phases in both the as-quenched and annealed $Al_{65}Cu_{20}Fe_{15}$ samples: one is the I phase with five-, three- and twofold axes, which is present principally in the as-quenched sample; the other is the D phase with a tenfold axis and periodic structure in the perpendicular direction, which is prevalent in the annealed samples.

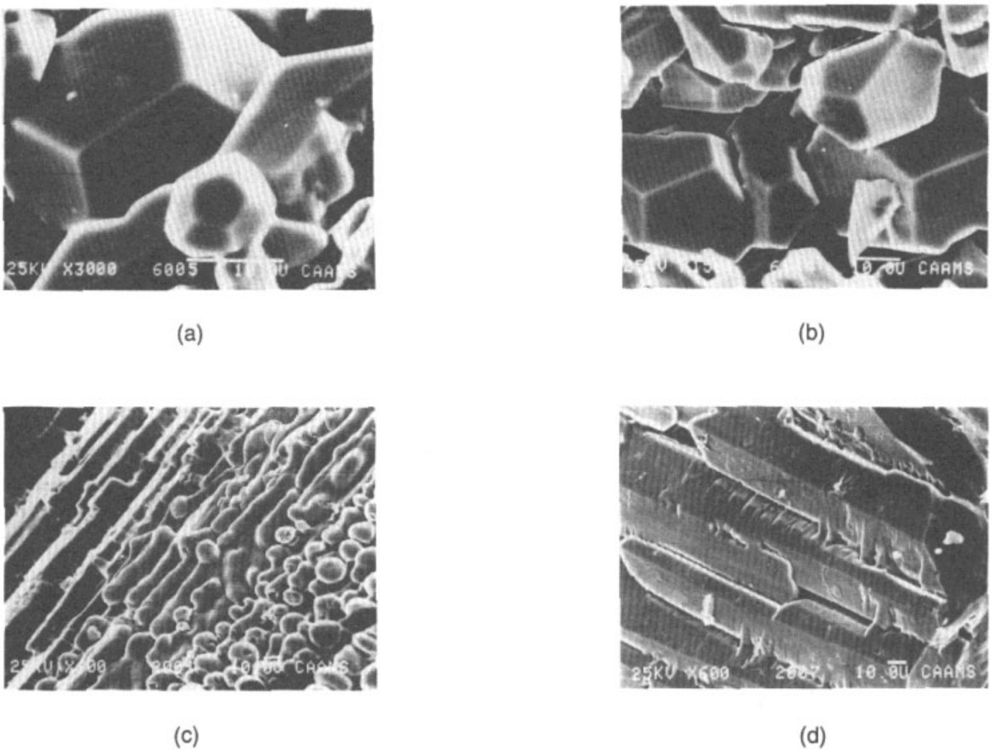

Figure 1. SEM of $Al_{65}Cu_{20}Fe_{15}$ quasicrystals from as-quenched to annealed samples.

We investigated the transformation of the I phase to the D phase carefully by SEM. Some of the results are shown in figure 1. It can be seen that the pentagonal dodecahedral morphology of the I phase is the dominant structure in the as-quenched sample and that the degree of decagonal columnar morphology gradually increases in annealed samples. There are obvious differences in macromorphology even though they are all quasicrystals. It is very hard to obtain completely single-phase structure in samples because this transformation is a slow process. The phase proportions are different for different annealing times, and the physical properties should also be different.

Figure 2 shows the variation in the magnetic properties of the $Al_{65}Cu_{20}Fe_{15}$ quasicrystal: the as-quenched sample in curve a is Pauli paramagnetic; the three annealed samples b, c and d also appear to be Pauli paramagnetic, with the paramagnetic value decreasing gradually. In curves e and f, the other two annealed samples are shown to be diamagnetic. This illustrates

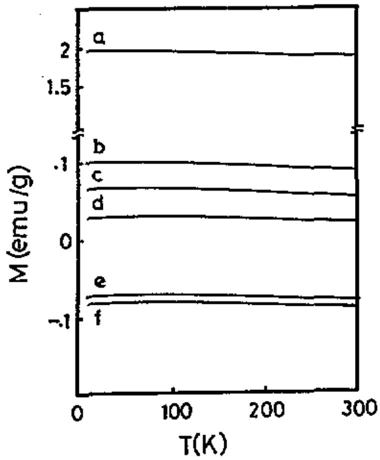

Figure 2. Magnetization versus temperature obtained in an applied field of 30 kOe for $Al_{65}Cu_{20}Fe_{15}$ specimens: a—as-quenched; b, c, d, e, f—annealed for 2, 4, 6, 8 and 10 days, respectively.

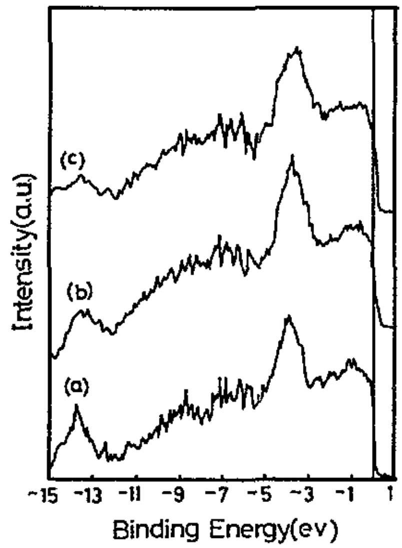

Figure 3. UPS results of three $Al_{65}Cu_{20}Fe_{15}$ samples: a—as-quenched, b— and c—annealed for 4 and 10 days, respectively.

that the $Al_{65}Cu_{20}Fe_{15}$ I phase is Pauli paramagnetic and transforms to a diamagnetic state gradually with increasing degree of D phase, in agreement with the result reported by Stadnik and co-workers [6]. This implies that the vacant part of the 3d band in Fe is gradually filled up by conducting electrons during the transformation from the I phase to the D phase, i.e. the Fermi level rises progressively. This is supported by UPS data, indicated in figure 3 for three samples: as-quenched, annealed for 4 days and annealed for 10 days. We utilize the ratio I_{E_F}/I_{Cu} to express the rising tendency of the Fermi level E_F with the increase of annealing time. I_{Cu} is the spectral intensity of Cu in the UPS curve. The binding energy of Cu is −4.1 eV (its relative intensity should be kept constant for different samples in our case) and I_{E_F} is the UPS intensity at the Fermi level. This rising tendency can be observed by comparing the I_{E_F}/I_{Cu} values. The Fermi level of the annealed sample is higher than that of the as-quenched sample, and the Fermi level increases with increasing annealing time. It is because the Fermi level rises during the heating process that the $Al_{65}Cu_{20}Fe_{15}$ quasicrystal can transform from paramagnetic to diamagnetic.

Finally, we conclude that the I phase transforms to the D phase in $Al_{65}Cu_{20}Fe_{15}$ alloys progressively during the heating process. The quasicrystal structure in the I phase is 3D, while in the D phase, the crystalline structure is along the axis direction. Therefore in $Al_{65}Cu_{20}Fe_{15}$ alloys, the D-phase quasicrystal structure is thermodynamically more stable than that of the I-phase quasicrystal. This is why the as-quenched sample will transform to the D-phase structure from the I-phase structure with higher energy, and hence the magnetism will transform from paramagnetism to diamagnetism.

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