Low-Temperature Specific Heat of Icosahedral and Amorphous Pd-U-Si Alloys

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Received August 10, 1987

Icosahedral (I) Pd$_{0.588}$U$_{0.206}$Si$_{0.206}$ can be obtained from melt-spun amorphous (A) ribbons by annealing. The specific heat $C$ (measured between $T=0.1$ K and 20 K) shows very similar behavior for both phases. The main features of $C$ are as follows. (i) The vibrational heat capacity $C_{ph}$ dominates $C$ at high $T$. $C_{ph}$ is almost identical in both phases, in agreement with recent inelastic neutron scattering data. (ii) Shallow maxima in $(C-C_{ph})/T$ vs. $T$ are found at 5.4 and 4.3 K for I and A phases, respectively, associated with magnetic order. These maxima are suppressed by $\approx 20\%$ in an applied magnetic field of 6 T. (iii) A large quasi-linear contribution is observed with a low-$T$ coefficient $\gamma = 165$ mJ/mole U K$^2$ for the I phase and $\gamma = 120$ mJ/mole U K$^2$ for the A phase. In the low-$T$ region, $C$ is hardly affected by a field of 6 T. This hints at the formation of a narrow $5f$ band with a comparable density of states for both I and A phases.

I. Introduction

The discovery of icosahedral (I) point symmetry in certain Al-Mn alloys [1] has generated considerable interest in these "quasi-crystalline" materials. Since information on the atomic positions is still lacking, it is of considerable interest to compare structurally sensitive physical properties of quasicrystals with those of other phases. In this respect, Pd-U-Si quasicrystals are very suitable since they can be prepared by a proper heat treatment of initially amorphous (A) samples [2], thus readily lending themselves to a comparison of these two metastable structures.

Previous studies have shown that magnetic ordering—presumably some type of complex antiferromagnetism—occurs around 5 K in both I- and A-Pd-U-Si [3]. On the other hand, it had been inferred from the temperature-independent susceptibility that the electronic properties of both phases might be vastly different [2]. It thus appeared highly desirable to determine the electronic density of states directly from heat-capacity measurements, in particular at very low temperatures, in order to facilitate a separation of electronic and magnetic contributions. As will be shown below, the magnetic-field dependence of the specific heat is capable of distinguishing between those two contributions. In addition, measurements to higher temperatures facilitate a comparison of the vibrational density of states of the two phases [4].

The present work also offers the opportunity to investigate the properties associated with the possible formation of a narrow $5f$ band or with a Kondo resonance in quasicrystalline and amorphous U compounds. In particular, it is worthwhile to check if coherence effects encountered in crystalline dense Kondo systems (Kondo lattices) might conceivably also occur in quasicrystalline or even amorphous metals.
II. Experimental

Ingots of composition Pd$_{0.588}$U$_{0.206}$Si$_{0.206}$ (Pd-U-Si for brevity) were prepared in an argon-arc furnace from standard materials with U$^{235}$ depleted to 0.2% in order to avoid radioactive self-heating of the samples at low temperature. Melt-spun amorphous (A) ribbons obtained from these alloys were heat treated at 495°C for 100 min in sealed evacuated pyrex tubes also containing Zr chips as oxygen getters. The particular composition and heat treatment chosen have been proven to yield the largest fraction of icosahedral (I) phase without crystallizing into the equilibrium UPd$_3$ phase. The I samples still contained ~20% of A phase as determined from X-ray and TEM analysis [5].

The specific heat of a piece of ribbon (~9 mg) was measured with the heat-pulse technique (see [6] for details) in a dilution refrigerator for 0.08 K < T < 2.5 K and a He$^4$ cryostat for 1.5 K < T < 25 K, each equipped with a superconducting magnet providing fields up to 6 T. The thermometers (Matsushita and Allen-Bradley carbon resistors) were calibrated against Ge (T < 2.5 K) and carbon-glass resistors (T > 1.5 K) located in the compensated field region of the magnets. For temperatures below 1 K, the calibration of the Ge resistor was checked with a CMN thermometer and also with a superconducting fixed point device [7]. Two sets of A- and I-Pd-U-Si samples were investigated. The results were the same within the errors of the measurements and the uncertainty of the amount of the A phase content within the I phase.

III. Results

Figure 1 shows the specific heat C vs. T on a log-log plot over the whole temperature range investigated. The main point of this figure is to emphasize the overall similarity of C in both phases. At low T, C varies roughly linearly with T. In fact, the data can be represented quite well as C ~ T$^{-1.1}$ over a considerable T range. A shallow structure is visible in C around 5 K which is more clearly seen in Figs. 2 and 3 (see below) and which coincides with the magnetic ordering temperatures of 5.4 and 4.3 K for the I and A phase as determined by the maxima in the magnetic susceptibility [3]. The somewhat steeper slope of C at higher T is of course due to the vibrational contribution C$_{ph}$.

These data can be more closely analyzed in plots of C/T vs. T$^2$ shown in Fig. 2a and b. In these plots, the magnetic-field data taken in 6 T are included. Note that the magnetic-field effect on C is strongest in the vicinity of the ordering temperature. A depression of C by about 20% is observed in 6 T, again very similar for I and A phase. The magnetic-field effect is very small (<5%) both at high and low temperatures, except at very low T where a contribution due to the Zeeman splitting of Pd$^{105}$ nuclei can be observed (data not shown).

Although a linear C/T vs. T$^2$ dependence typical of nonmagnetic metals is not observed in either phase because of contributions from the U moments, the rise of C/T towards high T can be unambiguously attributed to phonons [8]. From their very recent inelastic neutron scattering study, Suck et al. [8] calculated the vibrational specific heat. These data are reproduced in Fig. 2a and b as solid lines. Again, the I data very closely resemble the A data. The good overall agreement of the calculated C$_{ph}$ with the slopes of the measured specific heat lends strong support to the above assignment.

In the remainder of this section, we comment briefly on C$_{ph}$, before turning to the more interesting low-temperature behavior of C. A smooth extrapolation to T~0 yields a Debye temperature Θ$_D$ = 180 K for A and 190 K for I. As structural relaxation of amorphous