

Stacking faults in UPt_3

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Atomic resolution transmission electron microscopy and X-ray diffraction measurements have been combined to investigate the microstructure of superconducting UPt_3 . Regions of a second double hexagonal phase with a typical dimension of 25–30 Å are found to occupy approximately 3% of the total sample volume.

1. Introduction

The heavy-fermion superconductor UPt_3 has attracted much experimental and theoretical interest, due to the multiple superconducting phases observed as functions of magnetic field and temperature [1]. The details of the superconducting phase diagram are sample-dependent [2], and are strongly affected by compositional modifications [3,4], departures from stoichiometry [5], and by the application of both uniaxial [6] and hydrostatic pressures [7]. In addition, high-temperature annealing is required to obtain the sharpest superconducting transitions and highest transition temperatures. Taken together, these observations suggest that structural defects may play an important role in the unusual superconducting properties of UPt_3 .

2. Experimental details

We report here results on two separate polycrystalline specimens of UPt_3 , prepared by arc-melting the constituent materials in an argon atmosphere. The first sample, prepared at Los Alamos National Laboratory, was allowed to cool slowly, and had a superconducting transition temperature of 0.48 K, as determined from AC susceptibility measurements. A thin section for transmission electron microscopy was spark cut from the arc-melted button, mechanically thinned, and finally ion-milled to electron transparency. The specimen was examined at room temperature in a JEOL

4000EX atomic resolution transmission microscope operating at 400 kV.

The second specimen, prepared in Amsterdam, was annealed at 950°C for seven days, and was the subject of previously reported electrical resistivity and specific heat measurements [4]. The superconducting transition temperature of this sample was determined by electrical resistivity to be 0.56 K, substantially higher than that of the first sample. What is more, we observed two well defined peaks separated by ~60 mK in the low-temperature specific heat of this sample. X-ray scattering experiments on this sample were carried out at room temperature using a rotating anode system operating at a wavelength of 0.71 Å.

3. Experimental results

The most direct evidence for the nature and distribution of stacking faults in UPt_3 comes from atomic resolution transmission electron microscopy. Figure 1(a) shows a representative region of the sample looking down the $[2\bar{1}\bar{1}0]$ zone axis, i.e. perpendicular to the c -axis of UPt_3 . The (0002) and (02 $\bar{2}$ 0) planes are clearly resolved, with lattice spacings of 2.45 and 2.49 Å, respectively, in good agreement with published values [8]. As indicated by the arrows, there are several regions ~20–30 Å in extent in which there appear to be extra planes of atoms. Figure 1(b) is an enlargement of one such ‘defected’ region. The extra atomic planes appear midway between the (0002) planes, and approximately one-third of the distance between the (02 $\bar{2}$ 0) planes of the host UPt_3 hexagonal

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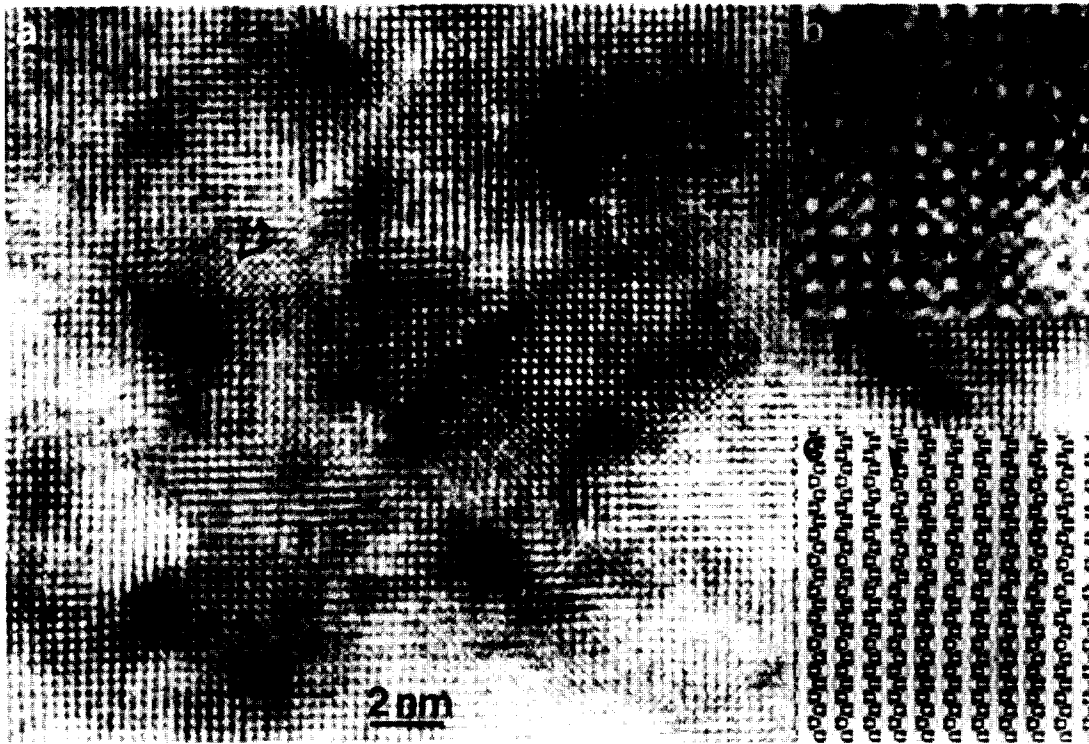


Fig. 1. (a) Image of (0002) and (0220) planes obtained from transmission electron microscopy. The c -axis is in the vertical direction. Arrows indicate regions with additional planes of atoms. (b) Enlargement of the 'defected' region. (c) Numerical simulation of the 'defected' region, assuming Ni_3Ti structure.

structure. The computed diffraction pattern for the Ni_3Ti structure (fig. 2b) and the diffraction pattern obtained from a fast Fourier transform of the minority phase region (fig. 2d) are compared in fig. 2, and show excellent agreement. We interpret the additional rows of atoms appearing in fig. 1(b) as projections from adjacent planes, which in the Ni_3Ti structure lie one-eighth of a unit cell dimension above and below the $[2\bar{1}10]$ plane. We cannot definitively rule out the presence of a more complicated and longer period defect layering structure, such as those known to exist in $U(Pd, Pt)_3$ [9], as well as in a number of rare earth [10] and transition metal [11] intermetallics. However, if present, such a structure must have predominantly double hexagonal character on short length scales.

X-ray diffraction is a powerful adjunct to our electron microscopy studies, providing statistical information about the density and structure of stacking faults. We note that previous neutron scattering studies [12] of single crystals of UPt_3 reported forbidden (0001) reflections and streaking parallel to the c^* -axis, observations which were taken as indications of stacking defects. The existence of large, oriented single-crystal grains in our second polycrystalline sample of UPt_3

permitted us to perform scans in selected directions of the crystal reciprocal space.

A $\theta-2\theta$ scan through the $(10\bar{1}2)$ peak is presented in fig. 3(a). As the instrumental resolution of this scan is $\sim 0.2^\circ$, no additional broadening is apparent. Figure 3(b) shows a rocking curve through the $(10\bar{1}2)$ peak. As indicated in the inset of fig. 3(b), this scan is approximately parallel to the c^* -axis. By comparing the background levels for figs. 3(a) and (b), we see that for the c^* -scan, the $(10\bar{1}2)$ peak lies on top of a ridge of diffuse scattering, which is missing for scans in the transverse direction. What is more, the instrumental broadening for the rocking curve (fig. 3b) only accounts for about one-third of the observed width of the $(10\bar{1}2)$ peak. Since there is no evidence for appreciable disorder or particle size broadening in the $\theta-2\theta$ scan, both the diffuse scattering and the excess width of the $(10\bar{1}2)$ peak in the rocking curve indicate the presence of substantial stacking disorder.

Assuming that the stacking faults have the double hexagonal structure imaged in the electron microscopy experiment, we used the simple analysis described by Warren [13] to determine their density. We find from the full width at half maximum of the $(10\bar{1}2)$ peak that

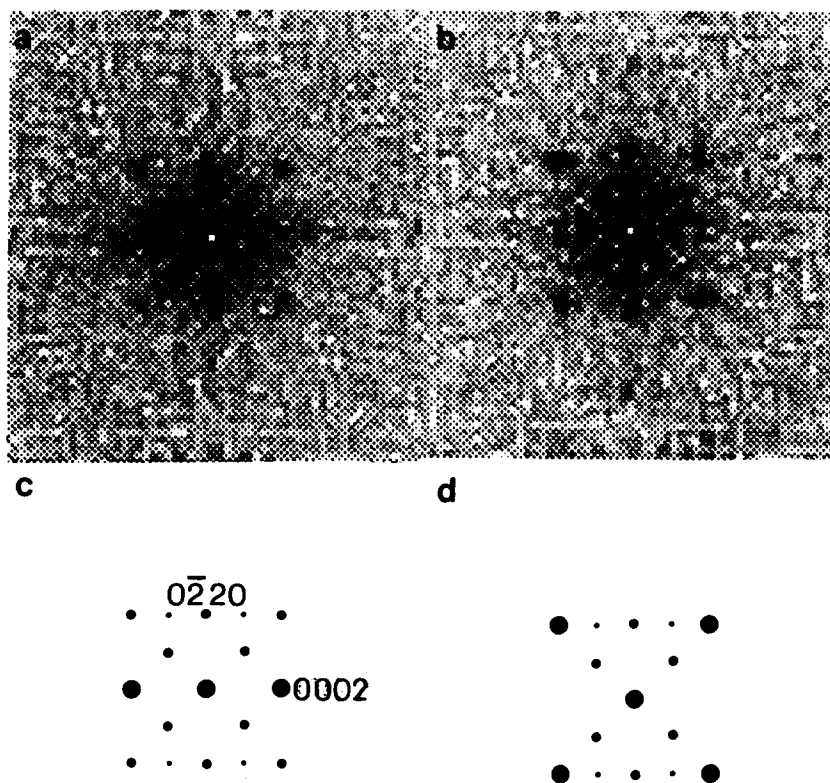


Fig. 2. Observed electron diffraction patterns for hexagonal host (a) and double hexagonal minority phase (b), compared with the computed diffraction patterns for hexagonal Ni_3Sn (c) and double hexagonal Ni_3Ti (d) structures.

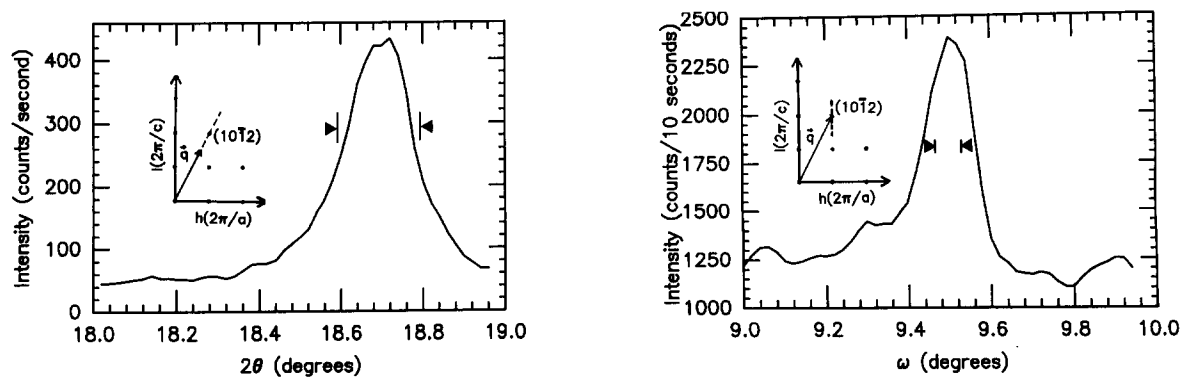


Fig. 3. Left: $\theta-2\theta$ X-ray scan through the $(10\bar{1}2)$ peak. Right: rocking curve through the $(10\bar{1}2)$ peak in the c^* -direction. Note the large diffuse scattering background. Arrows indicate the instrumental resolution in each scan. The respective directions in reciprocal space for each scan are indicated in the insets.

the probability that any (0002) plane is at the site of a stacking fault is approximately 3%. As the model predicts, there is no detectable shift of the $(10\bar{1}2)$ peak from the position indicated by the reported lattice parameters [8].

4. Discussion

Taken together, the electron microscopy and X-ray diffraction results indicate that our specimens of UPt_3 are best regarded as predominantly simple hexagonal

embedded with small double hexagonal regions with a typical dimension of 25–30 Å. The second phase occupies approximately 3% of the overall sample volume. We can immediately rule out the possibility that the secondary phase is simply contamination by one of the other known U–Pt compounds (UPt, UPt₂ or UPt₃) by noting that none of these three compounds is hexagonal or double hexagonal. In addition, since evidence for stacking defects is found in both the electron microscopy and X-ray measurements, we can discount the possibility that the stacking faults were introduced in the preparation of the microscopy specimen.

Perhaps the most striking finding of our study is that stacking defects are present not only in an unannealed sample with a rather low superconducting transition temperature (0.48 K), but also in a fully annealed sample with an exceptionally high superconducting transition temperature (0.56 K). Although it is intriguing to note that the size of the double hexagonal regions is comparable with estimates for the zero-temperature superconducting correlation length [14], the detailed relationship between the presence of this minority phase and the superconducting and magnetic properties of UPt₃ is at present unknown. Annealing and impurity studies to address this point are under way.

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