Microstructure and Crystal Perfection of Trigonal UPt_3

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Introduction

The structure of UPt_3 is key to the understanding of the normal and superconducting states in this important heavy fermion compound. Kycia et al. have shown that the residual resistivity ratio (RRR) of UPt_3, a measure of crystal perfection, decreases significantly with increased annealing temperature.[1] Since impurity concentrations are too low to explain this trend, some other source of defects must exist in the crystal lattice. Hong et al.[2] used TEM to study the annealing dependence of stacking faults and dislocations in UPt_3, but the overall density of such faults seems again too low to fully explain the annealing dependence of RRR.

Using high-energy x-ray diffraction, we recently discovered that the crystal structure of UPt_3 is not hexagonal but rather trigonal,[3] with space group $P\bar{3}m1$. Here we report further studies in which we attempt to connect the low-temperature properties to the microstructure of UPt_3.

Methods and Materials

Single crystals of UPt_3 were grown in a UHV zone-refining system. Most samples were subsequently annealed for six days and slowly cooled. X-ray diffraction measurements were performed in two ways: Experiments were performed using the rotating crystal method at beamline 5-BM-D of DND-CAT at the APS on a MAR CCD detector. The complete diffraction patterns of eleven samples, annealed at various temperatures, were collected to determine the Bragg-Williams long-range order parameter. Data analysis and fitting were performed in the same manner for all samples to reduce systematic errors. Lineshape measurements were performed on a four-circle diffractometer at beamline 1-ID-C of SRI-CAT, using the high-energy undulator and bent double-Laue monochromator. At both stations, 75-keV x rays were used; high-energy x rays were essential for minimizing absorption in this dense material.

Results and discussion

The Bragg-Williams long-range order parameter,[4] S_{BW}, is a measure of chemical order in an alloy; it equals the fraction of one type of atom on the correct site less the fraction of the same type of atom on the incorrect site. Figure 1 shows the very high values of SBW found for all samples, based on diffraction patterns collected at beamline 5-BM-D. The small deviation from unity could be attributed to systematic errors in fitting the data (e.g., values of anomalous scattering factors) or could be due to a small density of antiphase boundaries. The uniformity of S_{BW} with annealing temperature indicates the lack of an order-disorder transition in UPt_3 and shows that UPt_3 forms as an ordered intermetallic alloy directly from the melt. This is especially clear from the high S_{BW} value of the sample that underwent no additional annealing but, in essence, solidified directly from the melt. This measurement demonstrates the very high degree of chemical long-range order in UPt_3 but does not explain the annealing-temperature dependence of the RRR.

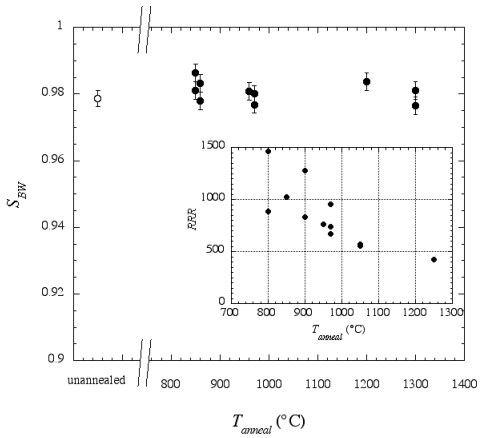

FIG. 1. Bragg-Williams order parameter of UPt3 vs. annealing temperature. Inset: Residual resistivity ratio vs. annealing temperature (from Ref. 1).

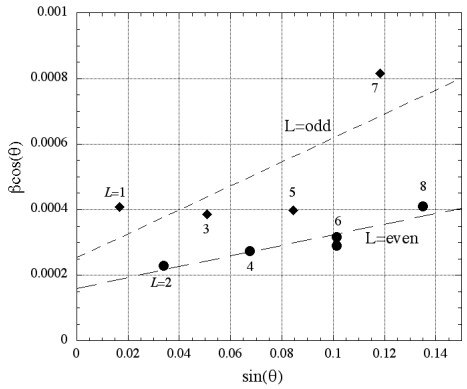

FIG. 2. Integral breadth b for the (00L) reflections of a UPt3 sample annealed at 1300° C, corrected for instrumental broadening. L is labeled for each reflection.

The crystal structure of UPt_3 is trigonal; a small relaxation along the c axis breaks the nearly hexagonal symmetry. Instead of the atoms being located at $z = \pm 1/4$, atoms are located at $z = \pm (1/4+\Delta z)$. Thus there is a slight variation in the spacing between planes along the c axis, alternating between $c(1+\Delta z)/2$ and $c(1-\Delta z)/2$. Spacing errors, which occur when the periodicity of the interlayer spacing is disrupted, will preferentially broaden reflections that are sensitive to the trigonal symmetry. Figure 2 shows integral widths of the (00L) peaks for a sample annealed at 1300°C and measured at beamline 1-ID-C. In this Williamson-Hall plot,[5] the intercept is inversely proportional to domain size

and the slope is proportional to strain. The odd L reflections, forbidden by hexagonal symmetry, are preferentially broadened, indicative of smaller effective domain sizes and greater strain. The positive slope indicates an excess of larger spacings $c(1+\Delta z)/2$. A quantitative model to explain details of the diffraction pattern broadening is being developed. Lineshape analysis is also being performed on samples annealed at other temperatures, to see if the density of spacing errors has any annealing-temperature dependence and therefore any connection to RRR and other low-temperature properties of UPt_3 samples.

Acknowledgments

Work at Northwestern University was supported by NSF grant No. DMR 9705473 and US DOE grant DE-F02-96ER45588. DND-CAT is supported by the E.I. DuPont de Nemours & Co., The Dow Chemical Company, the U.S. NSF through Grant DMR-9304725 and the State of Illinois through the Department of Commerce and the Board of Higher Education Grant IBHE HECA NWU 96. Use of the Advanced Photon Source was supported by the U.S. DOE, Basic Energy Sciences, Office of Energy Research under Contract No. W-31-102-Eng-38.

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