Neutron scattering studies of the lattice expansion in a ferromagnetic superconductor UGe$_2$ under pressure.

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Abstract. We report high-resolution measurements of the lattice constants of UGe$_2$ under pressure probed by a novel technique which utilises Larmor precession of polarised neutrons and surpasses the resolution of conventional scattering methods by two orders of magnitude. We confirm the presence of sharp anomalies in the lattice parameters at both the Curie and crossover temperatures at ambient pressure. We find that for pressure of 9.3 kbar the anomaly at the Curie temperature shifts to lower temperature in agreement with the known phase diagram. At 9.3 kbar, the pressure corresponding to an onset of superconductivity, the lattice expansion at the ferromagnetic transition is much stronger than at ambient pressure. The results indicate a complex evolution of the electronic structure of UGe$_2$ with pressure and suggest that magnetoelastic coupling is strengthened at the pressures at which superconductivity appears; magnetoelastic coupling therefore may play an important role in stabilising superconductivity.

1. Introduction

UGe$_2$ is the first material in which bulk superconductivity co-exists rather than competes with ferromagnetism[1, 2]. The schematic pressure-temperature phase diagram of UGe$_2$ is shown in Fig. 1. At ambient pressure UGe$_2$ orders at $T_C$≈53 K. Within the ferromagnetic state a crossover temperature $T_X$≈30 K separates two ferromagnetic states, FM1 (high pressure) and FM2 (low pressure) characterised by different values of the ordered moment. At low pressures the line of $T_C$ is of second order, whereas $T_X$ is a crossover. At pressure higher than ≈11.5 kbar the $T_X$ line is a first order transition with a critical end point for this transition line at ≈11.5 kbar[2]. The electronic specific heat coefficient $\gamma$ increases from ≈50 mJ/mole-K$^2$ to ≈100 mJ/mole-K$^2$ when approaching the FM2 to FM1 transition at zero temperature[3] suggesting that the unconventional superconductivity in UGe$_2$ arises from the strong spin fluctuations associated with the critical end point of $T_X$.

Since the ferromagnetism in UGe$_2$ is suppressed with pressure we expect a strong anomaly in the thermal expansion at $T_C$ and also near the pressure driven transition between FM1 and FM2 states. Use of a novel neutron scattering method, Larmor diffraction allows us to measure...
the thermal expansion of two lattice constants under hydrostatic pressure conditions with the resolution of $\Delta d/d \sim 1 \times 10^{-6}$[4, 5]. Recently the potential of Larmor diffraction to study the thermal expansion and the distribution of the lattice constants of non-ferromagnetic systems has been demonstrated[6, 7] but our study is the first study of a ferromagnetic state.

2. Experiment

Single crystals of UGe$_2$ were grown using the Czochralski technique under a protective atmosphere of purified Ar to suppress the vapour pressure of Ge. Single crystals were oriented using white beam X-ray back scattering "Laue method" and sparkcut to fit into a pressure cell. The mosaic of the samples used was $\approx 1$ degree (FWHM). Samples cut off the single crystals used in the neutron scattering measurements showed residual resistivity ratio, RRR$\approx$70-120 and $T_C=52.6$ K indicating the good quality of the crystals. Pressure of up to 12 kbar was applied using Cu:Be piston-cylinder pressure cells with Fluorinert as the pressure transmitting medium. The pressure cells were the same used as used in previous studies[6, 7]. Neutron Larmor diffraction measurements were performed with the TRISP spectrometer at FRMII(Germany); details of the method are given in Refs.[6, 7]. The temperature and pressure dependence of all three lattice constants of the orthorhombic structure of UGe$_2$ were measured at (200), (060) and (002) positions in reciprocal space. The lattice expansion was measured on heating between 3 K and 75 K. The pressure was estimated at low temperatures from the measured temperature of the paramagnetic to ferromagnetic transition determined from the feature in the intensity of the (040) peak and known phase diagram of UGe$_2$[2, 8]. A demagnetisation procedure was applied to ensure equal populations of oppositely polarised ferromagnetic domains and to prevent depolarisation of the diffracted neutrons by the ordered moment of the sample.

3. Results and Discussion

At ambient pressure the lattice expansion in UGe$_2$ is quite anisotropic, Fig. 2. The lattice contracts on cooling as evidenced by the positive slope of $\Delta d/d$ at $T>T_C$. The temperature dependence of $\Delta d/d$ shows a sharp anomaly at $T_C$ for $b$ and $c$ axes but merely a change of slope for the $a$ axis, whereas the crossover regime in the ferromagnetic state at $\approx 30$ K is marked by broad anomalies along $a$ and $b$ axes but not along $c$ axis in a good quantitative agreement with results of strain gauge measurements[9]. The relative length along the $b$ axis increases by $\approx 5 \times 10^{-5}$ on cooling through $T_X$, while the change along $a$ axis is of the opposite sign and of roughly the same magnitude. The linear thermal expansion coefficients $\alpha$ were obtained by differentiating $\Delta d/d$ with respect to $T$ (not shown). At the ferromagnetic transition $\alpha$ along $a$ increases by $\approx 1 \times 10^{-6}$ K$^{-1}$ in agreement with strain gauge and dilatometry measurements[9, 10].
The crossover region $T_X$ manifests itself as a broad feature along $b$ axis near 30 K. Observation of rather large ($\Delta d/d \approx 5 \times 10^{-5}$) feature at $T_X$ indicates that the electronic structure of UGe$_2$ changes strongly when crossing between FM1 and FM2.

![Figure 2. Temperature dependence of the linear lattice expansion along $a$, $b$, and $c$ axes at ambient pressure. $\Delta d/d$ is relative to the lowest temperature measured (3 K). Error bars are the size of markers. Vertical dashed lines mark $T_X$ and $T_C$.](image)

![Figure 3. Temperature dependence of the linear lattice expansion along the $b$ axis. $\Delta d/d$ is relative to the lowest temperature measured (3 K). Error bars are smaller than the size of the markers where not shown explicitly. Arrows mark $T_C$.](image)

The results of the high pressure neutron Larmor diffraction measurements are shown in Fig. 3 for the $b$ axis. As the pressure increases the anomaly at $T_C$ moves to lower temperatures in agreement with the pressure-temperature phase diagram. Unexpectedly we find that the change in value of $\Delta d/d$ between 3 K and the transition temperature increases under pressure by a factor of $\sim 3$, indicating strong magneto-elastic coupling in UGe$_2$ near the pressure at which superconductivity is induced. A broad maximum corresponding to $T_X$ is no longer visible in the data at 9.3 kbar. This increase is bigger than that of $\gamma$ and is consistent with an increase in the Grüneisen parameter as predicted approaching a quantum critical point[11]. Based on ambient pressure results we expected to see a stronger signature of $T_X$ at higher pressure, however it appears that the transition at $T_C$ dominates the lattice expansion.

4. Conclusions

We have measured the lattice expansion of UGe$_2$ under pressure and shown that the lattice expansion increases near the pressure at which the superconductivity is induced. Our results indicate that the magnetoelastic coupling in UGe$_2$ is strong and its electronic structure is quite sensitive to the variation of the lattice parameters. These are to our knowledge the first measurements that show Larmor diffraction can be applied in the ferromagnetic state.

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