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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 magneto-elastic coupling is strengthened at the pressures at which superconductivity appears; magneto-elastic 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 $T_C \approx 53$ K. Within the ferromagnetic state a crossover temperature $T_X \approx 30$ 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 γ 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

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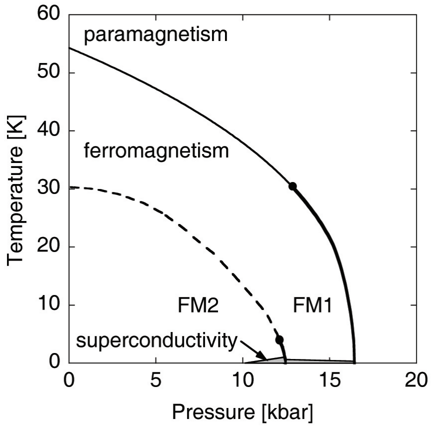

Figure 1. Schematic pressure-temperature phase diagram of UGe_2. Thin (thick) lines represent the second (first) order phase transitions. The dashed line marks the crossover regime and the dots mark the critical end points. The superconducting region is a dark area at the bottom.

the thermal expansion of two lattice constants under hydrostatic pressure conditions with the resolution of $\Delta d/d{\sim}1\times10^{-6}$[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 ≈1 degree (FWHM). Samples cut off the single crystals used in the neutron scattering measurements showed residual resistivity ratio, RRR≈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 ≈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 $\approx5\times10^{-5}$ 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 α were obtained by differentiating $\Delta d/d$ with respect to T (not shown). At the ferromagnetic transition α along *a* increases by $\approx1\times10^{-6}$ 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.

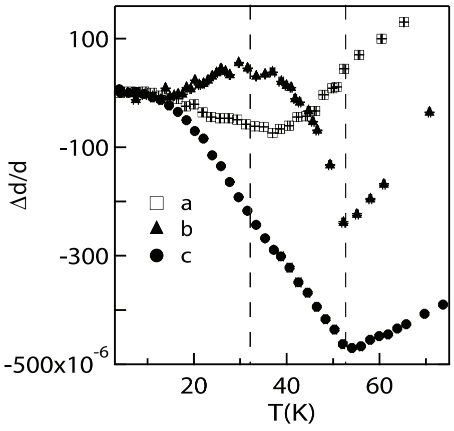

Figure 2. Temperature dependence of the linear lattice expansion along *a*, *b*, and *c* axes at ambient pressure. $\Delta d/d$ is relative to the lowest temperature measured (3 K). Error bars are the size of markers. Vertical dashed lines mark T_X and T_C.

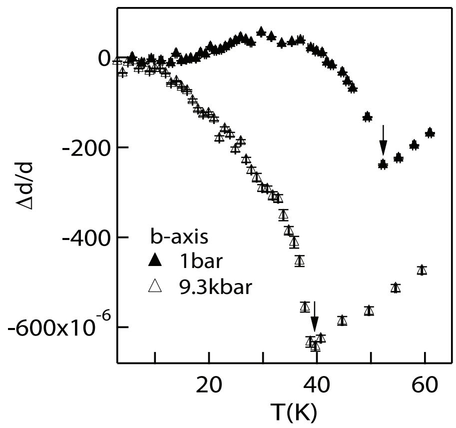

Figure 3. Temperature dependence of the linear lattice expansion along the *b* axis. $\Delta d/d$ is relative to the lowest temperature measured (3 K). Error bars are smaller than the size of the markers where not shown explicitly. Arrows mark T_C.

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 γ 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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