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Thermal expansion of the superconducting ferromagnet UCoGe

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We report measurements of the coefficient of linear thermal expansion, \(\alpha(T)\), of the superconducting ferromagnet UCoGe. The data taken on a single-crystalline sample along the orthorhombic crystal axes reveal a pronounced anisotropy with the largest length changes along the \(b\) axis. The large values of the step sizes \(\Delta\alpha\) at the magnetic and superconducting phase transitions provide solid evidence for bulk magnetism and superconductivity. Specific-heat measurements corroborate bulk superconductivity. Thermal-expansion measurements in magnetic fields \(B\parallel a, b\) show \(\Delta\alpha\) at \(T_C\) grows rapidly, which indicates the character of the ferromagnetic transition becomes first order like.

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The intermetallic compound UCoGe \((T_s=0.8\text{ K and }T_C=3\text{ K})\) belongs to the small group of superconducting ferromagnets.1 Superconducting ferromagnets (SCFMs) have the intriguing property that SC occurs in the FM phase, at a temperature \(T_s\) well below the Curie temperature \(T_C\), without expelling magnetic order.2 Until now, this peculiar ground state has been found in a few materials—all uranium intermetallics—only: in UGe2 state has been found in a few materials—all uranium compounds. Since the electronic and magnetic parameters of UTx compounds, with \(T\) a transition metal and \(X\) is Si or Ge, are, in general, strongly anisotropic,3 it is of uttermost importance to carry out further research on high-quality single-crystalline samples. Recently, Huy et al.10 reported the first magnetic and transport measurements on single crystals. Magnetization data revealed FM in UCoGe is uniaxial with \(m_0=0.07\mu_B\) pointing along the orthorhombic \(c\) axis. Resistance \(\rho(T)\) showed the upper critical field, \(B_c^\text{\(T\)}\), has an unusual large anisotropy with \(B_c^\text{\(T\)}\parallel a, b\) a factor \(\sim 10\) larger than for \(B_c^\text{\(T\)}\parallel c\).13

In this Brief Report we present measurements of the thermal properties of UCoGe single crystals. We find that the coefficients of linear thermal expansion measured along the crystal axes display a pronounced anisotropy with the largest length changes along the \(b\) axis. Large values of the step sizes \(\Delta\alpha\) at the FM and SC phase transitions provide evidence for bulk magnetism and SC. Specific-heat measurements support this conclusion. We use the Ehrenfest relation to analyze the uniaxial pressure dependencies of \(T_s\) and \(T_C\). Thermal-expansion measurements in applied magnetic fields indicate the nature of the FM phase transition changes to first order.

Single crystals of UCoGe were prepared by the Czochralski method as described in Refs. 14 and 15. The measurements were carried out on two samples, both shaped into a bar by means of spark erosion with typical dimensions of \(1 \times 1 \times 4\) mm\(^3\) and the long direction along the \(a\) sample \#1 and \(b\) axes (sample \#2). The samples were annealed15 and their good quality is attested by the high residual resistance ratio’s, \(\text{RRR=R(300 K)/R(1 K)}\), of \(\sim 30\) and \(\sim 40\) for sample \#1 and \#2, respectively. Sample \#1 was previously used to obtain the data in Refs. 14 and 16. It shows a large diamagnetic signal at the superconducting transition, \(T_s=0.5\text{ K},\) with a magnitude of 80% of the ideal screening value. The coefficient of linear thermal expansion, \(\alpha=L^{-1}(dL/dT)\), was measured using a three-terminal parallel-plate capacitance method using a sensitive dilatometer.17 Length changes along the \(a, b,\) and \(c\) crystal axes were measured along the short edges \((\sim 1\text{ mm})\) of the samples: \(\alpha_a\) and \(\alpha_b\) were measured on sample \#1 and \(\alpha_c\) on sample \#2. The data were taken in a \(^3\)He system in the \(T\) range 0.23–15 K.
and in a dilution refrigerator for $T = 0.05-1$ K. The specific
heat was measured on sample #2 (mass $\sim 0.1$ g) using a
semiadiabatic heat-pulse technique for $T = 0.15-1$ K.

In Fig. 1 we show $\alpha(T)$ for $T \leq 8$ K measured along the
main crystal axes. The data reveal a strong anisotropy. In the
paramagnetic phase, $\alpha_a$ and $\alpha_b$ are positive while $\alpha_c$ is
negative. The most pronounced variation is observed along the $b$
axis. For this direction, the transition to FM yields a negative
and to SC a positive contribution to $\alpha$. For the $a$ and $c$ axes
the contributions are smaller and the polarity is reversed. At
the FM and SC phase transitions large steplike changes,
the contributions are smaller and the polarity is reversed. At
bulk

The resulting

specific-heat data on single crystals of the heavy-fermion SC

polycrystal,1 where we assume $\beta = 3 \times \alpha$. The data show a

polycrystal,1 where we assume $\beta = 3 \times \alpha$. The data show a
nice overall agreement, but, obviously, the phase transitions
are much sharper for the single crystal.

Specific-heat, $c(T)$, data around the SC transition are re-
ported in Fig. 3(a). The phase transition for this crystal #2 is
broad with $\Delta T_s \sim 0.2$ K. An estimate for the step size
$\Delta c(T_s)$ can be deduced using an equal entropy method
[dashed line in Fig. 3(a)], which yields an idealized transition
at $T_s = 0.35$ K and $\Delta c(T_s)/\gamma_N \approx 0.7$, where $\gamma_N
= 0.062$ J/mol K$^2$ is the Sommerfeld coefficient. This value
is considerably smaller than the BCS value 1.43 for a con-
ventional SC. On the other hand, a smooth extrapolation of
$c(T)$ versus $T=0$ indicates the presence of a residual term
$\gamma_0 = 0.04$ J/mol K$^2$. Since orthorhombic SCFMs are, in
principle, two-band SCs (Ref. 21) with equal spin-pairing triplet
states $|\uparrow \downarrow \rangle$ and $|\downarrow \uparrow \rangle$ in the spin-up and spin-down bands,
respectively, a finite $\gamma_0$ value could be taken as evidence that
only one-band superconductors,22 in which case $\gamma_0 = \gamma_N/2$. However, in our case the broad transition and finite $\gamma_0$ value
strongly suggest sample quality is an issue. The low value
$\Delta c(T_s)/\gamma_N$ and finite $\gamma_0$ term remind one of the early
specific-heat data on single crystals of the heavy-fermion SC

UCoGe

and is reported in Fig. 2. Ideally, the

UCoGe

coefficients

UCoGe

of volumetric expansion is given by

$\beta = \sum_i \alpha_i$, where $i=a, b, c$ and is reported in Fig. 2. Ideally, the $\alpha_i(T)$ curves should be measured on one single sample. However, in our case we used two samples with slightly different RRR values. The resulting $\beta(T)$ data shows a large negative step at $T_C$ and a positive step at $T_s$. Since the phase transitions are relatively broad in temperature, we use an equal area construction30 to obtain idealized sharp transitions. In this way we extract $T_C^{bulk} = 2.6$ K and $T_s^{bulk} = 0.42$ K. In the inset of Fig. 2 we compare $\beta(T)$ of the single crystal with previous results on a

UCoGe

FIG. 1. (Color online) Coefficient of linear thermal expansion
versus temperature of UCoGe along the orthorhombic $a$, $b$, and $c$
axes as indicated. Arrows indicate the ferromagnetic (at $T_C$) and
superconducting (at $T_s$) transition temperatures.

UCoGe

FIG. 2. (Color online) Coefficient of volumetric thermal expan-
sion of single-crystalline UCoGe as a function of temperature. The
dashed lines represent idealized sharp FM and SC transitions at
$T_C = 2.6$ K and $T_s = 0.42$ K, respectively. The blue thicker arrow
locates the presence of an additional contribution in the FM state
(see text). Inset: comparison of $\beta(T)$ of single-crystalline (closed
circles) and polycrystalline (solid line) UCoGe. The dashed line
gives $\beta_{para}(T) = \alpha T$ (see text).

UCoGe

FIG. 3. (Color online) (a) Specific heat of UCoGe (single crystal
#2) in a plot of $c/T$ versus $T$. (b) $\alpha_i(T)$ of UCoGe (single crystal
#1). (c) Resistivity versus $T$ of UCoGe (single crystal #2). The
vertical dotted line indicates the approach to the zero-resistance
state coinciding with the onset temperature of bulk SC as seen in
$c(T)$ and $\alpha_i(T)$. The dashed lines in (a) and (b) represent idealized
sharp FM and SC transitions.
The relative volume change $\Delta V/V = [V(T) - V(0.05 \text{ K})]/V$ as a function of $T$ (solid blue line). The black dashed line gives $\Delta V/V$ in the absence of FM order. The red dotted line gives a smooth extrapolation of $\Delta V/V$ in the absence of SC. Inset: blow-up of the low-T part.

phase transition in $c/T$ [Fig. 3(a)] and $a_b$ [Fig. 3(b)] we obtain $\Delta k_{\text{bulk}}$ is 0.35 K and 0.42 K, for sample #2 and #1, respectively.

With the help of the Van der Waals relation for second-order phase transitions $dT_c/dp = V_m \Delta \alpha_b / \Delta (c/T_c)$ (where the molar volume $V_m = 3.13 \times 10^{-5} \text{ m}^3/\text{mol}$) one may extract the uniaxial pressure variation in $T_c$. Since not all steps $\Delta \alpha_b$ and $\Delta (c/T_c)$ have been measured on the same sample, we here restrict ourselves to a qualitative analysis. The largest effect is calculated for uniaxial pressure, $p_{ab}$, along the $b$ axis: $T_c$ increases and $T_c$ decreases. For $p_{ac}$ and $p_b$, the effect is smaller with reversed polarity. An estimate of the variation in $T_c$ as a function of hydrostatic pressure can be calculated using the relation: $dT_c/dp = V_m \Delta \alpha_b / \Delta (c/T_c)$. By combining the results obtained on the two crystals, using the values $\Delta \alpha_b = 1.19 \times 10^{-6} \text{ K}^{-1}$ (see Fig. 2) and $\Delta (c/T_c) = 0.038 \text{ J/mol K}^2$ (Fig. 3), we calculate $dT_c/dp = 0.098 \text{ K/kbar}$. This value is larger than the value 0.062 K/kbar deduced for a polycrystal. In the same way we calculate $dT_c/dp = -0.79 \text{ K/kbar}$, where we used $\Delta \alpha_b = -3.53 \times 10^{-6} \text{ K}^{-1}$ (Fig. 2) and the polycrystal value $\Delta (c/T_c) = 0.014 \text{ J/mol K}^2$ (Ref. 1). Notice, the pressure variations deduced from the Van der Waals relation are considerably larger than the experimental values $dT_c/dp = 0.03 \text{ K/kbar}$ and $dT_c/dp = -0.21 \text{ K/kbar}$, which tells us the quantitative analysis should be interpreted with care.

The relative volume changes due to FM order and SC are obtained by integrating $\beta(T)$ versus $T$. The result is shown in Fig. 4. The spontaneous magnetostriction is obtained by integrating $\beta_{\text{MAG}}(T)$, i.e., the difference between the measured $\beta(T)$ and the paramagnetic background term. The latter is approximated by a linear term $\beta_{\text{para}} = aT$ with $a = 4.4 \times 10^{-7} \text{ K}^{-2}$ (see inset Fig. 2). The relative volume change due to the spontaneous magnetostriction amounts to $\Delta V/V = 4.2 \times 10^{-6}$ for $T \rightarrow 0$ and is much larger (and has an opposite sign) than the estimated $\Delta V/V = -2.5 \times 10^{-7}$ due to SC (see inset Fig. 4). The latter value is due to the condensation energy of the SC state and agrees well with similar values obtained for heavy-fermion superconductors. Thus FM order is not expelled below $T_c$ and coexists with superconductivity.Muon spin rotation and relaxation experiments provide evidence for the coexistence of SC and FM on the microscopic scale.

A closer inspection of the volumetric thermal expansion in Fig. 2 reveals an additional contribution visible below $\sim 1.5 \text{ K}$ in the FM phase, just before SC sets in. This shoulder indicates the presence of a second energy scale, most likely related to low-energy spin fluctuations. It will be highly interesting to investigate whether these spin fluctuations provide the pairing interaction for SC. Notice, a second low-energy scale associated with spin fluctuations has also been identified in the thermal expansion and specific heat of URhGe and UGe$_2$.

Finally, we present measurements of $\alpha(T)$ around the Curie point in magnetic fields applied along the dilatation direction (see Fig. 5). Again we observe a large anisotropy. For $\alpha_c$ and $B||c||m_0$ the phase transition smears out rapidly in a field of $1 \text{ T}$, $\alpha_c(T)$ is virtually independent of temperature up to $10 \text{ K}$ and close to zero. For $B||a,b$ the magnetic contribution to $\alpha_c$ and $\alpha_b$ grows rapidly and attains the large values of $\sim 2 \times 10^{-5} \text{ K}^{-1}$ at $T_c$ in a field of $8 \text{ T}$. The large length changes show the nature of the FM transition becomes first orderlike in an applied magnetic field. This is in line with the phase diagram for an itinerant quantum FM when tuned to the critical point with the magnetic field playing the role of pressure. A recent analysis of the Landau free energy of FM UCoGe in a magnetic field predicts $T_c$ is reduced in a transverse field $B \perp m_0$. The variation in $T_c$ with magnetic field $B||a,b$ as determined from the thermal-expansion data in field is given in the insets of Figs. 5(a) and 5(b), respectively. $T_c$ shows a small increase in low magnetic fields, but then is rather insensitive for $B$ up to $8 \text{ T}$. Magnetotransport data...
reveal that for $B \parallel b$ the critical field at which $T_C \to 0$ is $\sim 15$ T.\textsuperscript{32}

In summary, we have investigated the thermal properties of the SCFM UCoGe. The use of single-crystalline samples enabled us to investigate the anisotropy in the coefficient of the linear thermal expansion. The largest length changes, $\Delta L/L$, are observed along the $b$ axis. Large phase-transition anomalies at $T_C$ and $T_p$ confirm bulk magnetism and bulk SC. By making use of Ehrenfest relations, the effect on $T_C$ and $T_p$ of uniaxial pressure was investigated. In the volumetric thermal expansion an additional contribution was observed which develops toward low $T$, just before SC sets in. Experiments on large, high-quality single crystals are required to further investigate this phenomenon as it may provide an important clue as regards low-energy spin fluctuations providing the glue for superconductivity.

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\textsuperscript{17}A. de Visser, Ph.D. thesis, University of Amsterdam, 1986.


\textsuperscript{20}An equal volume for the broadened and idealized contributions is imposed when integrating $\beta T$ with respect to the background signal.


\textsuperscript{25}The value of $dT_c/dp$ in Ref. 1 should read 0.02 K/kbar. Notice, this value and $dT_c/dp=-0.25$ K/kbar refer to uniaxial pressure dependencies, since they are evaluated using $\Delta \alpha$ rather than $\Delta \beta$.


