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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 $B\|a,b$ show $\Delta\alpha$ at T_C grows rapidly, which indicates the character of the ferromagnetic transition becomes first orderlike.

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The intermetallic compound UCoGe (T_s=0.8 K and T_C =3 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 UGe_2 (Ref. 3) and UIr (Ref. 4) under pressure and in URhGe (Ref. 5) and UCoGe (Ref. 1) at ambient pressure. SCFMs attract much attention because their ground state does not obey the standard BCS scenario for phonon-mediated SC, because the FM exchange field impedes pairing-up of electrons in spin-singlet Cooper pairs.[6] Instead, the itinerant nature of the U 5f magnetic moments, together with the notion that these materials are close to a magnetic instability, has led to the proposal that SC is unconventional and promoted by a novel pairing mechanism:[7,8] critical FM spin fluctuations mediate pairing of electrons in spin-triplet states. In recent years ample evidence for such an unusual pairing mechanism in SCFMs has been put forward.[3,9–11] SCFMs are excellent laboratory tools to investigate the interplay of magnetism and SC, which is a key issue in unraveling the properties of a wide range of materials, such as heavy-fermion, high-T_s cuprate, and FeAs-based superconductors.

UCoGe crystallizes in the orthorhombic TiNiSi structure (space group *Pnma*).[12] The coexistence of SC and FM in UCoGe was first reported for polycrystalline samples.[1] Magnetization measurements show the emergence of a weak FM phase below T_C=3 K with a small ordered moment m_0 =0.03μ_B. The analysis of the magnetization data by means of Arrott plots corroborates itinerant FM. This is further substantiated by specific-heat data, which show that the entropy associated with the magnetic phase transition is small (0.3% of R ln 2). In the FM phase, SC is found with a transition temperature T_s=0.8 K, as determined by resistance measurements. The ac susceptibility shows large diamagnetic signals below T_s=0.6 K. Thermal-expansion and specific-heat measurements on polycrystalline samples[1] confirmed the bulk nature of the SC and FM phases with T_s^{bulk}=0.45 K and T_C^{bulk}=3 K, respectively.

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[13] it is of uttermost importance to carry out further research on high-quality single-crystalline samples. Recently, Huy *et al.*[14] reported the first magnetic and transport measurements on single crystals. Magnetization data revealed FM in UCoGe is uniaxial with m_0=0.07μ_B pointing along the orthorhombic c axis. Resistance measurements showed the upper critical field, $B_{c2}(T)$, has an unusual large anisotropy with $B_{c2}(T\to 0)$ for $B\|a,b$ a factor ~10 larger than for $B\|c$.

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 annealed[15] and their good quality is attested by the high residual resistance ratio's, $RRR=R(300\text{ K})/R(1\text{ K})$, of ~30 and ~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 K, with a magnitude of 80% of the ideal screening value. The coefficient of linear thermal expansion, α $=L^{-1}(dL/dT)$, 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 (~1 mm) of the samples: α_b and α_c were measured on sample #1 and α_a on sample #2. The data were taken in a ^{3}He system in the T range 0.23–15 K

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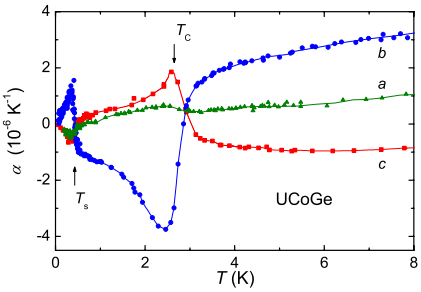

FIG. 1. (Color online) Coefficient of linear thermal expansion versus temperature of UCoGe along the orthorhombic a, b, and c axes as indicated. Arrows indicate the ferromagnetic (at T_C) and superconducting (at T_s) transition temperatures.

and in a dilution refrigerator for $T=0.05-1$ K. The specific heat was measured on sample #2 (mass ~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, α_a and α_b are positive while α_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 α. 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, $\Delta\alpha$, are found. The large values of $\Delta\alpha$ at T_C and T_s provide solid evidence that FM and SC are bulk properties. Notice, the step sizes at T_s are comparable to the ones obtained for the heavy-fermion SCs URu_2Si_2 (Ref. 18) and UPt_3.[19] The coefficient of volumetric expansion is given by $\beta=\Sigma_i\alpha_i$, 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 construction[20] 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

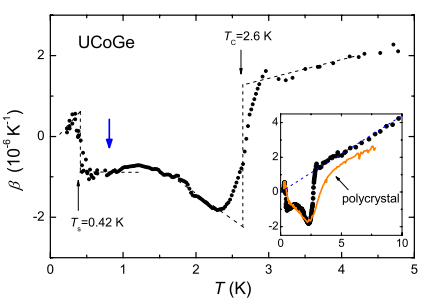

FIG. 2. (Color online) Coefficient of volumetric thermal expansion of single-crystalline UCoGe as a function of temperature. The dashed lines represent idealized sharp FM and SC transitions at $T_C=2.6$ K and $T_s=0.42$ K, respectively. The blue thicker arrow locates the presence of an additional contribution 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 $\beta_{para}(T)=aT$ (see text).

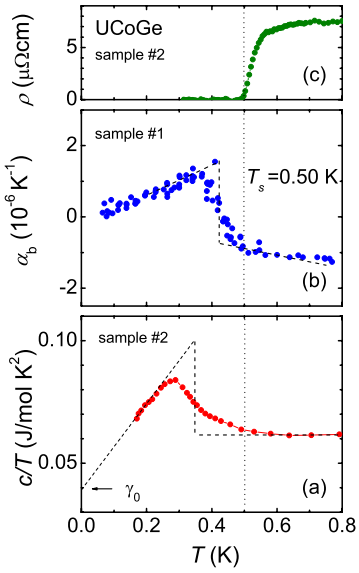

FIG. 3. (Color online) (a) Specific heat of UCoGe (single crystal #2) in a plot of c/T versus T. (b) $\alpha_b(T)$ of UCoGe (single crystal #1). (c) Resistivity versus T of UCoGe (single crystal #2). The vertical dotted line indicates the approach to the zero-resistance state coinciding with the onset temperature of bulk SC as seen in c/T and $\alpha_b(T)$. The dashed lines in (a) and (b) represent idealized sharp FM and SC transitions.

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 reported 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 conventional 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 $|\uparrow\uparrow\rangle$ and $|\downarrow\downarrow\rangle$ in the spin-up and spin-down bands, respectively, a finite γ_0 value could be taken as evidence that only one-band superconducts,[22] in which case $\gamma_0=\gamma_N/2$. However, in our case the broad transition and finite γ_0 value strongly suggest sample quality is an issue. The low value $\Delta(c/T_s)/\gamma_N$ and finite γ_0 term remind one of the early specific-heat data on single crystals of the heavy-fermion SC UPt_3.[23] Upon improving the sample quality the transition became more and more sharp, and eventually a split transition appeared as well as a much reduced γ_0 value.[24]

In Figs. 3(c) and 3(b) we compare $c(T)/T$ with resistivity, $\rho(T)$, data taken on the same sample, and $\alpha_b(T)$ measured on sample #1. The resistivity measurements were carried out with a four-point low-frequency ac method with a current of 100 μA along the b axis. The zero-resistance state is reached at 0.5 K, which corresponds to the onset temperature T_s^{onset} for bulk SC. Using idealized constructions for the SC

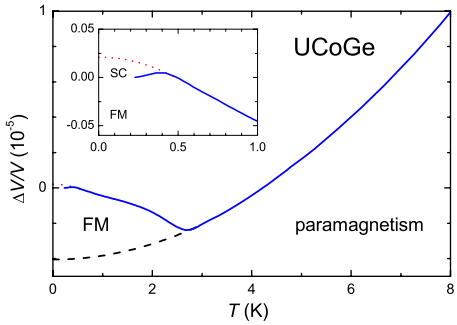

FIG. 4. (Color online) The relative volume change $\Delta V/V = [V(T) - V(0.05\ \mathrm{K})]/V$ as a function of T (solid blue line). The black dashed line gives $\Delta V/V$ in the absence of FM order. The red dotted line gives a smooth extrapolation of $\Delta V/V$ in the absence of SC. Inset: blow-up of the low-T part.

phase transition in c/T [Fig. 3(a)] and α_b [Fig. 3(b)] we obtain T_s^{bulk} is 0.35 K and 0.42 K, for sample #2 and #1, respectively.

With the help of the Ehrenfest relation for second-order phase transitions $dT_{s,C}/dp_i = V_m \Delta\alpha_i / \Delta(c/T_{s,C})$ (where the molar volume $V_m = 3.13 \times 10^{-5}\ \mathrm{m^3/mol}$) one may extract the uniaxial pressure variation in T_s and T_C. Since not all steps $\Delta\alpha_i$ and $\Delta(c/T_{s,C})$ have been measured on the same sample, we here restrict ourselves to a qualitative analysis. The largest effect is calculated for uniaxial pressure, p_b, along the b axis: T_s increases and T_C decreases. For p_a and p_c the effect is smaller with reversed polarity. An estimate of the variation in T_s as a function of hydrostatic pressure can be calculated using the relation: $dT_s/dp = V_m \Delta\beta / \Delta(c/T_s)$. By combining the results obtained on the two crystals, using the values $\Delta\beta = 1.19 \times 10^{-6}\ \mathrm{K^{-1}}$ (see Fig. 2) and $\Delta c/T_s = 0.038\ \mathrm{J/mol\,K^2}$ (Fig. 3), we calculate $dT_s/dp = 0.098\ \mathrm{K/kbar}$. This value is larger than the value 0.062 K/kbar deduced for a polycrystal.[1,25] In the same way we calculate $dT_C/dp = -0.79\ \mathrm{K/kbar}$, where we used $\Delta\beta = -3.53 \times 10^{-6}\ \mathrm{K^{-1}}$ (Fig. 2) and the polycrystal value $\Delta c/T_C = 0.014\ \mathrm{J/mol\,K^2}$ (Ref. 1). Notice, the pressure variations deduced from the Ehrenfest relation are considerably larger than the experimental values $dT_s/dp = 0.03\ \mathrm{K/kbar}$ and $dT_C/dp \sim -0.21\ \mathrm{K/kbar}$,[16,26] 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 $\beta_{\mathrm{FM}}(T)$, i.e., the difference between the measured $\beta(T)$ and the paramagnetic background term. The latter is approximated by a linear term $\beta_{para} = aT$ with $a = 4.4 \times 10^{-7}\ \mathrm{K^{-2}}$ (see inset Fig. 2). The relative volume change due to the spontaneous magnetostriction amounts to $\Delta V/V = 4.2 \times 10^{-6}$ for $T \to 0$ 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.[18,19] Thus FM order is not expelled below T_s and coexists with superconductivity. Muon spin rotation and relaxation experiments[27] provide evidence for the coexistence of SC and FM on the microscopic scale.

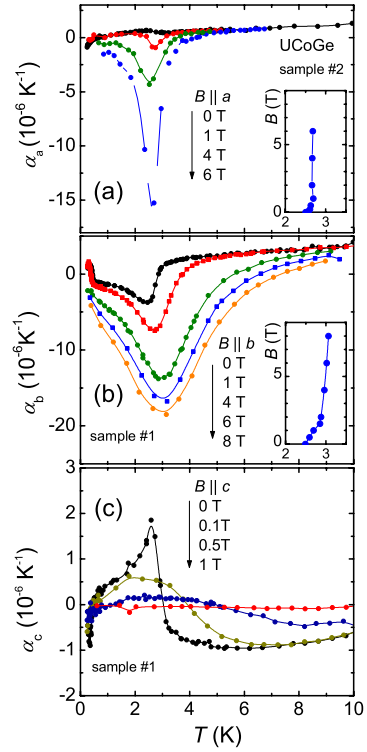

FIG. 5. (Color online) Coefficient of thermal expansion of UCoGe in applied magnetic fields along the dilatation direction as indicated. (a) α_a for $B \| a$; (b) α_b for $B \| b$; and (c) α_c for $B \| c$. Insets: T_C as a function of $B \| a, b$.

A closer inspection of the volumetric thermal expansion in Fig. 2 reveals an additional contribution visible below ~ 1.5 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.[28,29]

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 α_c and $B \| c \| m_0$ the phase transition smears out rapidly in a field of 1 T, $\alpha_c(T)$ is virtually independent of temperature up to 10 K and close to zero. For $B \| a, b$ the magnetic contribution to α_a and α_b grows rapidly and attains the large values of $\sim -2 \times 10^{-5}\ \mathrm{K^{-1}}$ at T_C in a field of 8 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[30] 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 $B \perp m_0$.[31] 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 T. Magnetotransport data

reveal that for $B \| b$ the critical field at which $T_C \to 0$ is ~ 15 T.[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_s confirm bulk magnetism and bulk SC. By making use of Ehrenfest relations, the effect on T_C and T_s 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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