Magneto-acoustic study of single crystalline $UCu_{0.95}Ge$

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We present results of a magneto-acoustic study of $UCu_{0.95}Ge$. This compound exhibits antiferromagnetic ordering at 48 K and shows a metamagnetic phase transition at 38 T for the magnetic field applied along the c direction, seen as a jump in the magnetization. The sound velocity and sound attenuation demonstrate pronounced anomalies in the vicinity of both magnetic phase transitions proving the important role of magnetoelastic interactions in the physics of this actinide compound.

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I. INTRODUCTION

UCuGe belongs to a large group of equiatomic uranium-based intermetallic compounds (UTX) with a transition metal T (Cu, Ag, or Au) and a p metal X of the III–V groups in the periodic table. Magnetic and other electronic properties of UTX depend predominantly on the oxidation state of the uranium $5f$ electrons, which are in turn strongly influenced by their hybridization with the ligand electrons. In recent years, detailed investigations of the magnetic properties of UTX compounds, especially of isostructural series with different T and X elements, have been performed.[1,2] These studies have provided systematic information on the evolution of the character of the $5f$-electron states by varying the chemical composition in a controlled way and have enabled testing the theoretical models of the physics of actinide-based materials.[1]

The crystal structure of UCuGe is closely related to that of ferromagnetic UGa_2 (having the hexagonal structure of the AlB_2 type).[3,4] Besides their similar crystalline and electronic structure, UCuGe possesses very distinctive magnetic properties. UCuGe crystallizes in an ordered version of the AlB_2 structure, which is the BaLiSi type.[5] The ordering of Cu and Ge at the B sites in the AlB_2 structure reduces the symmetry from P6/mmm to $P\bar{6}m2$. In stoichiometric UCuGe, the atomic positions are U (000), Cu (1/3 2/3 1/2), and Ge (2/3 1/3 1/2). However, similar to some other representatives of the UTX group, UCuGe has a relatively wide homogeneity range. Study of the magnetic susceptibility of UCu_xGe ($0.85 \leqslant x \leqslant 0.95$) and $UCuGe_y$ ($0.800 \leqslant y \leqslant 0.997$) showed that the magnetic properties depended strongly on the composition.[6–8] The magnetic structure of the stoichiometric compound UCuGe was examined by neutron diffraction on polycrystalline samples.[9] According to this study, the magnetic unit cell contains four U ions, and the U magnetic moments lie in the basal ab plane. In the $z = 0$ layer, they form an angle of $\pi/3$ with the a axis. Within the layer, U ions form ferromagnetic chains along the a axis, which are coupled antiferromagnetically. In the neighboring layer at $z = 1/2$, the moments are turned by $\pi/2$ with respect to the layer with $z = 0$. The U magnetic moment μ_U is equal to $2.0\mu_B$.[9] This value might be overestimated, as suggested from a preliminary single-crystal neutron diffraction study that nevertheless confirmed the basal-plane arrangement of the magnetic moment.[10] In the off-stoichiometric samples, in addition to the antiferromagnetic (AF) low-temperature state, some peculiarities have been discussed in terms of spin-glass behavior, which has been suggested to occur in the temperature range up to 78 K.[6,7]

Anomalies in the temperature dependence of the magnetic susceptibility, electrical resistivity, and specific heat observed in the single crystal of $UCu_{0.95}Ge$ point to an AF ordering at the Néel temperature $T_N = 48$ K, without any noticeable peculiarities appearing above this temperature.[11] It has been found that the magnetic susceptibility in fields applied along the basal plane is somewhat higher than along the c axis. Within the basal plane, the largest susceptibility is observed for fields applied along the a axis for T below 25 K. This anisotropy vanishes above 25 K. High-field magnetization measurements for the hard c direction have revealed a field-induced transition at 38 T (at 4.2 K) to a state with $1.35\mu_B$ per formula unit (f.u.). No transition occurs for magnetic fields applied along either the a or b axis up to 42 T, where magnetization reaches $0.75\mu_B$/f.u.[11] The susceptibility along the b axis amounts to $\sim \cos(\pi/3)$ of the value along the a axis, which indicates a large magnetic anisotropy within the basal plane. Irreversible magnetic-history-dependent anomalies observed in fields applied along the basal plane (in the vicinity of 15 T at 4.2 K) are attributed to repopulation of antiferromagnetic domains.[11]

In order to understand the role of magnetoelastic interactions in the magnetic properties of $UCu_{0.95}Ge$, we have performed ultrasound investigations of this compound. One can expect that the spontaneous (magnetic ordering) and field-induced phase transitions observed in $UCu_{0.95}Ge$ should be accompanied by anomalies in the acoustic properties. Ultrasound investigations have been proven to be a powerful experimental tool for studying various phase transitions and critical phenomena.[12] In this paper, we present the results of a comprehensive ultrasonic study (temperature and field dependence of the sound velocity and sound attenuation) performed on a $UCu_{0.95}Ge$ single crystal for ultrasound waves propagating along the principal axes. Preliminary results obtained for one geometry (propagation along the c axis) have been reported in Ref. 13.

II. EXPERIMENTAL

The UCuGe single crystal was grown by use of the Czochralski method in a tri-arc furnace from stoichiometric melt. More details on the UCuGe crystal growth can be found in Ref. 11. The composition of the crystal determined by microprobe analysis shifted to the Cu-deficit side, namely $UCu_{0.95}Ge$. The lattice parameters have been found to be $a = 418.5$ pm and $c = 379.6$ pm, in agreement with literature data for this composition within the homogeneity range.[6] The magnetic properties, however, differ from those of polycrystals reported in Ref. 6. The sample was cut from the same ingot as was used for the magnetization study in Ref. 11. The back-scattered Laue patterns were used to orient the crystal. Two pairs of parallel facets perpendicular to the a and c axes with a distance between the facets of 1.91 mm in both cases, were polished for the ultrasound measurements. The measurements have been performed using a pulse-echo technique[14] in the frequency range from 40 to 85 MHz. A longitudinal acoustic wave with the wave vector **k** and polarization **u** was propagated along the a (acoustic c_{11} mode) and c axes (acoustic c_{33} mode). A pair of piezoelectric film transducers were glued to the surfaces in order to excite and detect the acoustic waves. The relative change of the sound velocity, $\Delta v/v$, and sound attenuation, $\Delta\alpha$, have been measured as a function of external parameters in these experiments. These as well as magnetization measurements have been performed in He-flow cryostats and in pulsed magnetic fields (up to 60 T) applied along the c axis. The magnetization signal was detected by integrating the voltage induced in a pick-up coil surrounding the sample. The absolute value of the magnetization was calibrated from steady-field measurements.

III. RESULTS

The renormalization of the phonon velocities and the sound attenuations in magnetic media are mostly the result of two causes.[12] First, sound waves change the ligand positions, and, therefore, the crystalline electric field of the ligands is affected. In turn, due to the strong spin-orbit interaction, it produces changes of the single-ion magnetic anisotropy of the U ions. It should change the effective g-factors of the U ions, and, thus, due to magnetoelastic interactions, the sound waves can change slightly the directions of the magnetic anisotropy, and vice versa, the magnetic anisotropy modifies the sound-wave parameters, such as the sound velocity and attenuation. This interaction between the sound waves and the magnetic subsystem exists at any temperatures lower than the characteristic energy of the single-ion magnetic anisotropy.

Second, sound waves change the magnetic-ion positions themselves and/or the positions of nonmagnetic ions, involved in the superexchange between magnetic ones. In that case, sound waves renormalize the effective exchange (and magnetic-dipole) interactions between magnetic ions. The manifestation of this effect is more pronounced as the influence of the sound on the single-ion magnetic properties, because the interionic magnetic and exchange interactions determine predominantly magnetic phase transitions.

Figure 1 shows the temperature dependence of the sound velocity, $\Delta v/v$, and the sound attenuation, $\Delta\alpha$, as measured

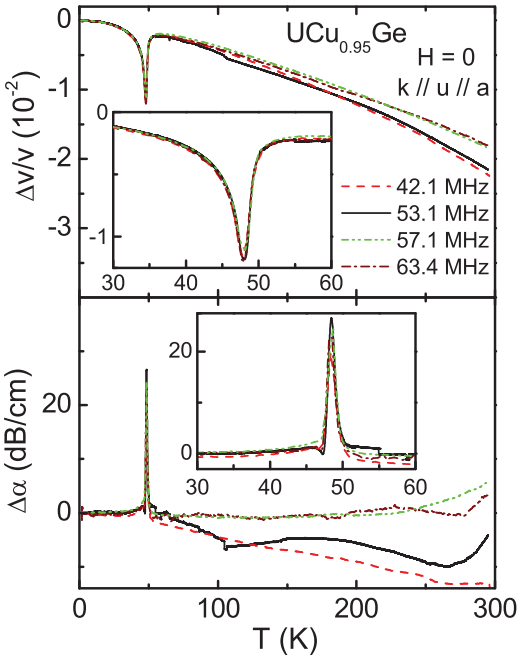

FIG. 1. (Color online) Temperature dependencies of the sound velocity, $\Delta v/v$, and the sound attenuation, $\Delta\alpha$, of the longitudinal ultrasonic waves propagating along the a axis (acoustic c_{11} mode) measured at various frequencies. Insets show enlargement of the data in the vicinity of T_N.

for several frequencies of the longitudinal acoustic wave propagating along the a axis at zero magnetic field. One can see well-pronounced anomalies in both quantities at the magnetic ordering, which are shown in more detail in insets of Fig. 1. The $\Delta v/v(T)$ curve exhibits a sharp softening of the acoustic mode by more than 1%, whereas $\Delta\alpha(T)$ displays a sharp maximum corresponding to a large energy dissipation at the transition. Both anomalies are frequency independent and occur at $T_N = 48$ K, exactly as determined from specific-heat measurements. Above T_N, a smooth change occurs due to the anharmonic contribution to the sound velocity (upper panel in Fig. 1).

A very different situation is observed when the longitudinal wave propagates along the c axis (Fig. 2). Similar to the wave propagation along the a axis, both acoustic quantities exhibit anomalies at T_N. But they are smaller, have a fine structure, and depend on frequency. At 42 and 51 MHz, the anomalies are qualitatively similar to those observed along the a axis, i.e., with a minimum in $\Delta v/v(T)$ and a maximum in $\Delta\alpha(T)$ at 47.3 K, both, however, smaller and broadened. With further increasing frequency, the $\Delta v/v$ anomaly splits into two minima, at 47.3 and 48 K. The second minimum becomes larger, being most pronounced at 65.7 MHz. Then the features become smaller again. The sound attenuation shows a small maximum at low frequencies, whereas for $f > 51$ MHz, the maximum at 47 K is followed by a minimum at 48 K. This is most pronounced at 61.4 MHz. At 65.7 MHz, an inversion of the curve occurs; there is first a minimum at 47 K and then a maximum at 48 K, with an additional smaller maximum in between. Finally, the single maximum restores at 85.6 MHz, shifted to 48 K compared to the low-frequency curve.

It is known that in many U compounds magnetic ordering from the paramagnetic to the ground state occurs in a

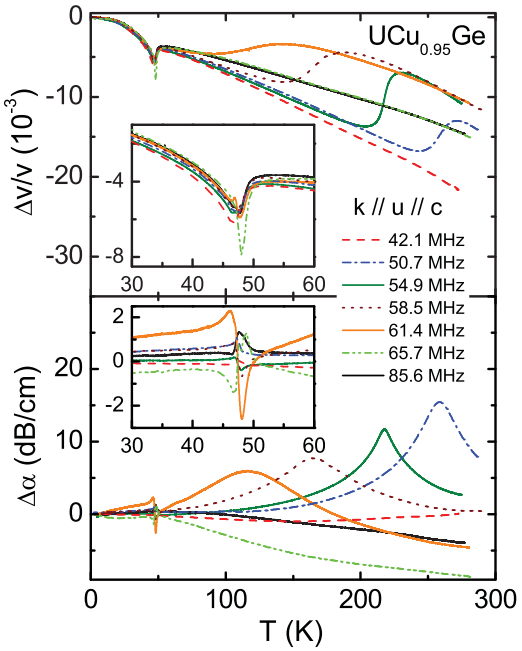

FIG. 2. (Color online) Temperature dependencies of $\Delta v/v$ and $\Delta\alpha$ of the longitudinal ultrasonic wave propagating along the c axis (acoustic c_{33} mode) measured at various frequencies. Insets show enlargement of the data in the vicinity of T_N.

narrow temperature range via several intermediate structures, incommensurate or with a long periodicity, such as in UNiGa[1] or UCu_2Si_2.[15] The fine structure of the acoustic anomalies at 47–48 K in our crystal may be related to similar transitions. Because of the off-stoichiometric composition with corresponding vacancies in the Cu sublattice, the local neighborhood of the U atoms varies to some extent over the crystal. A Mössbauer-effect study[8] revealed that in UCu_xGe vacancies in the Cu sublattice cause a strong perturbation of the magnetic structure due to a balance change of the competing exchange interactions. This is accompanied by the formation of the two new spin configurations. Different UCu_xGe compounds show antiferromagnetism with three typical coexisting U-moment configurations. One of them corresponds to the original magnetic structure of the stoichiometric UCuGe compound (with a hyperfine field $B_{hf} = 5.7$ T) and the two others ($B_{hf} = 6.5$ T and 9.7 T) appear due to vacancies in the Cu sublattice. The quantitative results of Ref. 8 should be taken with care because 0.5% Ge in the polycrystalline samples were substituted by Sn, enriched with the ^{119}Sn isotope in order to allow for the Mössbauer effect. This may as well influence the balance of the exchange interactions.

The magnetic structure of UCuGe, characterized by the AF arrangement of the magnetic moments in the basal plane, should be determined by competing exchange interactions and spin frustrations. Since the nearest atoms around U are Cu and Ge, it is quite probable that the competing interactions in this material originate in U-Cu-U and U-Ge-U indirect interactions mediated by the $5f$-ligand hybridization.[1] In this scenario, vacancies on the Cu sites in the nearest surrounding of the U atom change the balance of these exchange interactions. This results in changes of the local configurations of the U moments and, consequently, the effective exchange

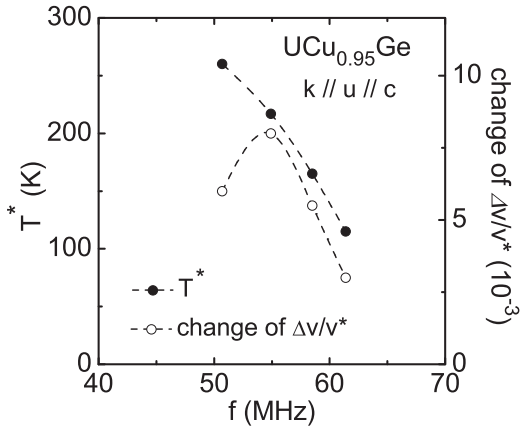

FIG. 3. Frequency dependence of the attenuation-maximum temperature (T^*) and of the height of the steplike change ($\Delta v/v^*$) in the sound velocity obtained from the data shown in Fig. 2.

fields are modified considerably. The effect should be rather gentle in our crystal because no anomalies pointing to this are observed in the susceptibility, electrical-resistivity, and specific heat measurements.[11] Indeed, all peculiarities that may be attributed to such inhomogeneities are observed in our ultrasound data only for one direction in a limited frequency range ($50 < f < 80$ MHz).

In addition to the anomalies accompanying the magnetic ordering, a broad and large step-like change of $\Delta v/v$ together with a large broad maximum of $\Delta\alpha$ are observed for the ultrasonic wave propagating along the c axis (Fig. 2). They occur at temperatures considerably above T_N and, thus, are not related to magnetism. For this measurement geometry, the anomalies are much larger than the ones at the magnetic ordering. Nevertheless, they do not exceed effects at T_N for the waves traveling along the a axis (1% and 28 dB/cm for sound velocity and attenuation, respectively) where no trace of anomalies above T_N are seen (Fig. 1). The observed "nonmagnetic"anomalies are strongly frequency dependent. The frequency dependencies of the attenuation-maximum temperature (T^*) and of the height of the steplike velocity change (v/v^*) are shown in Fig. 3. The inflection points of $\Delta v/v(T)$ coincide exactly with T^* found from the maximum in $\Delta\alpha(T)$. The anomalies disappear at 65.7 and 85.6 MHz. For $f = 42.1$ MHz, a maximum might occur above room temperature exceeding our measurement range. The observed anomalies might be caused by vacancy dynamics in the Cu sublattice. Obviously, frequency-dependent features detected at relatively high temperatures (Fig. 2) are not related to magnetic ordering and to magnetic degrees of freedom of the considered system; the characteristic frequencies of the magnetic movement are several orders of magnitude larger than the observed resonance frequencies. It is known that at a temperature T a crystal defect, which has a migration energy U_d, moves with the frequency $\nu_d = B\nu\exp(-U_d/k_BT)$, where ν is the frequency of the oscillating atoms and B is a constant of order unity.[16] Under the action of a small field F, such a defect moves with the velocity $v = DF/k_BT$, where $D = D_0\exp(-U_d/k_BT)$ is the diffusion coefficient. We can speculate that under the sound perturbation, defects (vacancies of Cu) present in $UCu_{0.95}Ge$, move along the c direction, and,

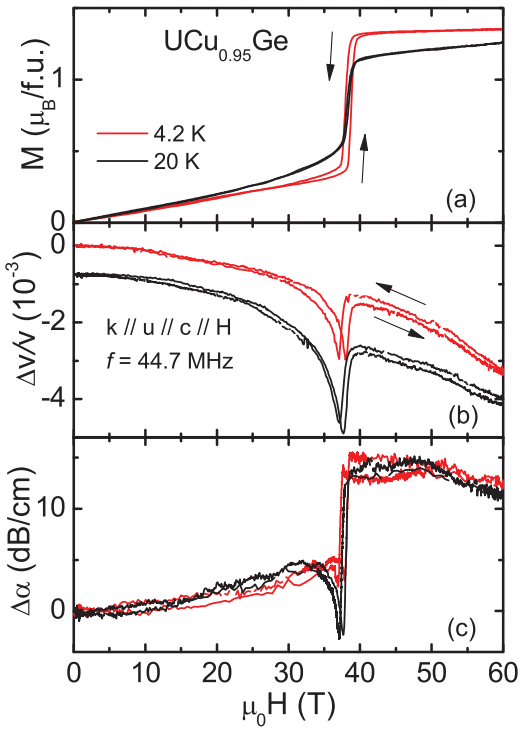

FIG. 4. (Color online) Field dependence of (a) the magnetization, (b) $\Delta v/v$, and (c) $\Delta\alpha$ measured in longitudinal geometry along the c axis in pulsed magnetic fields applied along the c axis, at 4.2 K and 20 K. Up and down field sweeps are shown.

hence, the characteristic frequency of their movement is equal to ν_d, which is a relatively small value. Such oscillations, according to the above analysis, can exist at temperatures higher than the temperatures of the magnetic ordering.

Figure 4 shows the field dependence of the magnetization as well as $\Delta v/v(T)$ and $\Delta\alpha$ measured for longitudinal ultrasound waves propagating along the c axis with magnetic fields applied along the same direction. The magnetization curve at 4.2 K increases linearly up to $\sim$ 36 T. Then an abrupt steep increase occurs between 38 T and 39 T with a hysteresis of 0.8 T. In Ref. 11, the transition was observed in a wider field interval, 38–44 T. A possible reason for that could have been an imperfect magnetic field alignment. In earlier measurements, the maximum field of 47 T did not allow determination of the saturation magnetization unambiguously.[11] Here we obtain the saturation value $1.35\mu_B$/f.u., having much higher fields at hand. The sharpness of the transition and its pronounced hysteresis point, in contrast to earlier assumptions,[11] to a first-order character of the transition. The saturation magnetization reached above $\sim$40 T suggests that parallel alignment of the U magnetic moments is achieved; consequently, the value $2\mu_B$ for μ_U reported in Ref. 9 seems to be overestimated.

The transition stays almost constant with increasing temperature up to 15 K but then becomes gradually smeared out and disappears at temperatures above T_N.[11] This can be seen as well in the magnetization curve measured at 20 K (Fig. 4), where the slope dM/dH, the magnetization gain over the transition, and the hysteresis become smaller compared to the 4.2 K data.

Both acoustic characteristics demonstrate well-pronounced anomalies at the transition. $\Delta v/v$ has a sharp minimum ($\sim$0.2%) with a 1 T hysteresis. Above the transition, $\Delta v/v$ recovers the value extrapolated from the fields below the transition. The attenuation exhibits an even sharper feature at the transition. After a sharp jump at 38 T, $\Delta\alpha$ remains at a strongly enhanced value. The observed hysteresis in the magnetization and in both acoustic characteristics is most probably an intrinsic property of the system, which is only slightly affected by the eddy-current heating during the pulse. The measurements performed in two pulsed magnets with various pulse durations (25 ms for the magnetization measurements and 150 ms for the ultrasound) provided very similar hysteresis widths in spite of a big difference in the time derivative of the magnetic field $\frac{\partial B}{\partial t}$. The anomalies in $\Delta v/v$ and $\Delta\alpha$ become even sharper at 20 K, despite the fact that the magnetization curve is smoother at this temperature. Thus, the ultrasound results confirm the first-order nature of the transition from the AF arrangement of the U magnetic moments to their parallel configuration along the c axis.

IV. CONCLUSIONS

We have performed magnetic and ultrasound investigations of $UCu_{0.95}Ge$ in a wide magnetic field and temperature range. Our pulsed magnetic-field data have revealed a first-order metamagnetic phase transition at 38 T to a spin-polarized state. This transition is accompanied by a jump in the magnetization and the sound attenuation as well as a softening of the acoustic c_{33} mode. Very pronounced acoustic anomalies, both at the metamagnetic transition and at the antiferromagnetic ordering at zero field, confirm the important role of magnetoelastic interactions in the magnetic properties of this actinide compound. Some unusual frequency-dependent features, observed in the sound velocity and attenuation above T_N, presumably can be related to the dynamics of Cu vacancies in $UCu_{0.95}Ge$.

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