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High-field magnetism and magnetoacoustics in uranium intermetallic antiferromagnets

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ABSTRACT

We report on studies on the magnetic and magnetoacoustic properties of uranium intermetallic antiferromagnets UCo_2Si_2, $UCu_{0.95}Ge$, UIrGe and U_2Ni_2Sn in pulsed magnetic fields up to 60 T, where they undergo metamagnetic transitions. The measurements were performed in the temperature range from 1.4 to 100 K. The magnetic ordering and the metamagnetic transitions are accompanied by pronounced anomalies in the ultrasound velocity and the ultrasound attenuation. All studies were performed on single crystals grown by the Czochralski method in a tri-arc furnace.

1. Introduction

Uranium intermetallic compounds are subjects of intensive fundamental research because of the special character of the 5*f* electron states. New interesting phenomena (e.g., heavy fermions, non-conventional superconductivity, non-Fermi-liquid scaling) have been discovered in the course of this research. Magnetic, magnetoelastic and other electronic properties of uranium intermetallics are connected with the degree of itinerancy of the 5*f* electrons and the inter-uranium exchange interactions. The hybridization of the uranium 5*f* electron states with the valence electron states of ligands plays a major role in the formation of 5*f* magnetic moments and in the magnetic ordering [1]. The magnetic properties of the uranium intermetallics vary starting from temperature-independent paramagnetism over different stages characterized by high-temperature Curie–Weiss paramagnetism, which may condense either to a non-magnetic ground state with spin fluctuations or into ferromagnetic or antiferromagnetic (AF) ordering at low temperatures. The AF compounds are of special interest because of possible field-induced transitions. Parameters of such transitions (e.g., the critical field value) give important information about the strength of exchange and anisotropic interactions in the compound.

Magnetically ordered uranium compounds typically have a huge magnetic anisotropy. Therefore, their study requires high-quality single crystals (their growth and characterization have been performed at the Joint Laboratory for Magnetic Studies, Charles University and Institute of Physics of Academy of Sciences, Prague) and the application of very high magnetic fields (available at the Dresden High Magnetic Field Laboratory, Helmholtz-Zentrum Dresden-Rossendorf). In the present work, we report on recent collaborative studies of the magnetism and magnetoacoustics of several less-studied representatives of uranium intermetallic antiferromagnets with different crystal structures in pulsed magnetic fields up to 60 T, with special attention to UCo_2Si_2.

2. Experimental

All single crystals were grown by the Czochralski method from the 8-gram stoichiometric mixtures of the pure elements (99.9% U and better for other components) using a tri-arc furnace with a copper water-cooled crucible under argon protective atmosphere. A tungsten rod was used as a seed and the pulling speed varied from 6 to 15 mm/h. The phase purity and composition of the samples were checked by standard x-ray powder diffractometry and x-ray microanalysis. The back-scattering x-ray Laue patterns showed the good quality of the crystals.

For the magnetization measurements, the samples were cut into parallelepipeds of 1.5 mm size with edges parallel to the principal axes. The magnetization curves were measured at 1.4–80 K in pulsed fields up to 60 T along the *c*-axis using a non-destructive pulsed-field magnet with pulse duration of 25 ms. The magnetization signal was detected by an induction method with a standard pick-up coil system. A detailed description of the setup is given elsewhere [2]. The absolute values of the magnetization were calibrated by steady-field measurements up to 14 T in a PPMS-14 magnetometer (Quantum Design).

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For the ultrasound measurements, two piezoelectric film transducers were glued onto parallel polished facets of the samples (typical diameters of the facets were 2–3 mm with thicknesses of the samples between 1.5 and 3 mm) and the measurements were performed using a pulse-echo technique [3] in a He^4-flow cryostat in pulsed magnetic fields up to 60 T above 1.5 K.

3. Results and discussion

3.1. UCo_2Si_2

UCo_2Si_2 belongs to the wide group of uranium ternary intermetallics UT_2X_2 where T is a late 3*d*, 4*d* or 5*d* metal and X is a *p*-element (Si and Ge) crystallizing into two related structural types. These compounds exhibit a large variety of magnetic states starting from AF (UCr_2Si_2), complex ferrimagnetic (UNi_2Si_2) and ferromagnetic structures (UCu_2Si_2) through Pauli paramagnets (UFe_2Si_2) to compounds which have a superconducting together with an AF state (URu_2Si_2) [1]. UCo_2Si_2 crystallizes in the tetragonal $ThCr_2Si_2$ crystal structure (space group *I*4/*mmm*). The structure is built up of U, Co and Si basal-plane atomic layers extended along the *c*-axis. UCo_2Si_2 orders antiferromagnetically below the Néel temperature T_N=83–85 K. Its magnetic structure obtained from powder neutron diffraction consists of ferromagnetic basal-plane layers of U magnetic moments of 1.42 μ_B (at 4.2 K) oriented parallel to the *c*-axis which are coupled in a simple sequence $+-+-$ (AF type-I structure) in the same direction [4]. The magnetic moment is localized only on the U atoms.

Results of previous studies of the magnetization, electrical resistivity and specific heat performed on a UCo_2Si_2 single crystal [5] point to T_N=83 K. The strong anisotropy is preserved in the paramagnetic range as well. The magnetic susceptibility along the moment direction (the *c*-axis in UT_2X_2) is much larger than along the basal-plane and obeys the Curie–Weiss law with an effective moment μ_{eff}=2.55 μ_B and paramagnetic Curie temperature $\Theta_p=-29$ K. The $\chi_{(T)}$ curve along the *a* axis is smooth and does not obey the Curie–Weiss law.

Because of the plain magnetic phase diagram with only one AF phase of simplest structure below T_N, UCo_2Si_2 did not attract as much attention as its isostructural analogs. UNi_2Si_2 has in its phase diagram in addition to the AF type-I structure ferrimagnetic and incommensurate AF phases. In the ground state, it shows a $++-$ sequence with longitudinally modulated magnitude of the magnetic moment resulting in the spontaneous moment 0.53 μ_B, which is 1/3 of the uranium moment μ_U [6]. Substitution of 10–15% Ni by Pd leads to the stabilization of the AF type-I structure, the ferrimagnetic $++-$ arrangement is restored by a metamagnetic transition (MT) at about 5 T and this state remains stable after the external field is removed [7]. The $++-$ phase is also observed in high magnetic fields in UPd_2Si_2 (with AF type-I structure in zero field). The transition occurs at the critical field μ_0H_{cr}=15 T and is accompanied by huge hysteresis and relaxation effects [8,9]. It is interesting to check whether UCo_2Si_2, where the 5f electrons should be more itinerant than in UNi_2Si_2 and UPd_2Si_2 due to a stronger 5f–d electron hybridization, exhibits this phase as well in a large magnetic field. In this work we used the same UCo_2Si_2 crystals as in Ref. [5] (the lattice parameters are *a*=392.1 pm and *c*=963.9 pm).

We have indeed observed such a transition. Fig. 1 shows magnetization curves measured along the principal axes at 1.4 K. In a field applied along the *c*-axis, the MT is observed at μ_0H_{cr}=45 T. The transition is extremely sharp; it is really “a jump” as it should be in the case of an ideal first-order transition. On the other hand, it has a very small hysteresis ($\mu_0\Delta H_{cr}$=0.16 T), especially in comparison with the huge hysteresis (> 15 T) found for an analogous transition in UPd_2Si_2 [8,9]. The observed transition is characterized by a magnetization jump ΔM=0.52 μ_B. The ΔM value corresponds roughly to 1/3 of the U magnetic moment of 1.42 μ_B determined by neutron powder diffraction [4]. Therefore, we can suppose that the state above the transition is ferrimagnetic with a $++-$ arrangement of ferromagnetic basal-plane layers (data for UNi_2Si_2 where such a state is spontaneous are shown in Fig. 1 for comparison). This means that a further transition from the $++-$ sequence to the parallel orientation of the magnetic moments can be expected in much higher fields, as observed for UNi_2Si_2 [9].

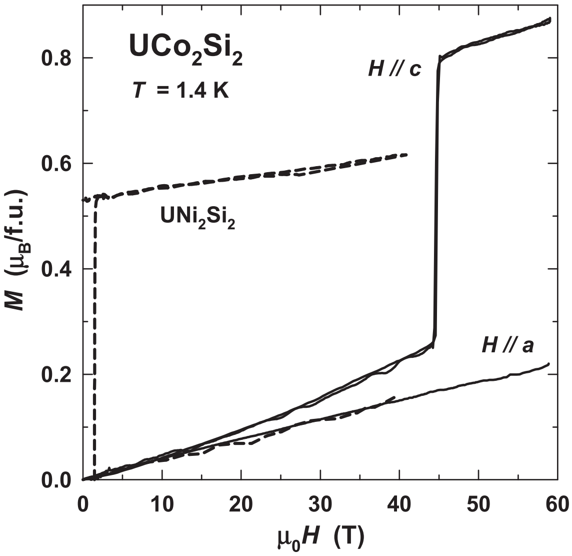

Fig. 1. Magnetization curves measured along the principal axes of a UCo_2Si_2 crystal at 1.4 K (solid lines). Analogous curves for UNi_2Si_2 [10] are shown by dashed lines.

The magnetization measured along the *a* axis shows no transition and is linear up to the highest fields applied in the experiment. The initial “isotropy” of the magnetic susceptibility for fields applied along the *a* and *c* axes is broken at about 15 T where the *c*-axis isotherm starts to exhibit a positive curvature. Just below the transition, the magnetization along the *c*-axis exceeds that along the *a*-axis by a factor of 1.5.

The *a*-axis susceptibility of 3.7×10^{-3} μ_B/T per U atom, the same as in UNi_2Si_2, is a typical value for the hard-axis magnetization slopes of U intermetallics independent of the crystal structure and type of the magnetic ground state and reflects mostly the Pauli paramagnetism of conduction electrons [1]. UCo_2Si_2, like many other U compounds, behaves as a paramagnet in fields applied perpendicular to the moment direction.

The transition is still very sharp at 20 K (Fig. 2). At higher temperatures, it becomes drastically wider and cannot be considered anymore to be of first order. The transition field, μ_0H_{cr}, determined as the field of the maximum *dM*/*dH* derivative, decreases with increasing temperature. Whereas the maximum *dM*/*dH* values are almost the same at 1.4 and 20 K (1.8 and 1.5 μ_B/T, respectively), it is only 0.14 μ_B/T at 40 K. A very small *dM*/*dH* maximum indicating the transition is still observed at 80 K.

The acoustic properties exhibit drastic anomalies both at the magnetic-ordering temperature in 0 T (data not shown) and at the MT. The changes in the ultrasound velocity $\Delta v/v$ and the peak in the attenuation $\Delta\alpha$ at the field-induced transition are more than an order larger than at the spontaneous transition. Whereas $\Delta\alpha$ exhibits only a very sharp peak at the MT (Fig. 3), $\Delta v/v$ has a more complex behavior, much “richer” than the corresponding *M*(*H*) curves (Fig. 4). It has a non-monotonous temperature evolution