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Published in:
Journal of Applied Physics

DOI:
10.1063/1.361347

Citation for published version (APA):

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Magnetism in URhSi

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The magnetic susceptibility ($\chi$) of URhSi, measured as a function of temperature, shows two distinct regions. At low temperatures below 10 K, the susceptibility is dominated by the rare-earth contribution, while above 10 K, the magnetic anisotropy and low-temperature excitations of the cerium subsystem prevail. The onset of ferromagnetization in URhSi is accompanied by a significant increase in the magnetic moment per uranin atom, which is close to $\mu_B/2$. The specific heat $C_p$ shows a strong peak at low temperatures, indicating the presence of a magnetic phase transition. The specific heat data can be fitted to the theoretical expression for magnetic ordering, suggesting a magnetic ground state with a magnetic moment of approximately 0.11 $\mu_B$.

SQUID magnetometer measurements reveal an itinerant 5$f$ ferromagnet with very reduced $U$ magnetic moments. The magnetic susceptibility, specific heat, and electrical resistivity indicate that URhSi orders ferromagnetically at low temperatures with the $U$ magnetic moments aligned along the $c$ axis. Anomalies in the temperature dependence of the magnetic susceptibility, specific heat, and electrical resistivity are observed, indicating a magnetic phase transition. The high-field magnetization data obtained on oriented powder reveal a strong magnetocrystalline anisotropy. All the results obtained on polycrystalline samples classify URhSi as an itinerant 5$f$ ferromagnet with very reduced $U$ magnetic moments. © 1996 American Institute of Physics.
The strongly nonlinear part between 20 and 150 K can be roughly approximated by a modified CW law including a temperature independent term \( \chi_0 \)
\[
\chi = C/(T - \Theta_p) + \chi_0
\]
with significantly lower value of Curie constant, a positive paramagnetic Curie temperature of 9.7 K and \( \chi_0 = 10^{-8} \) m\(^3\)/mol. Although, the applicability of both approaches for polycrystalline data of an anisotropic material is rather questionable some tentative conclusions can be made. The value of the effective moment from the high temperature fit, which can be taken as the upper limit in this material is much lower compared to free ion \( U^{3+} \) or \( U^{4+} \) expectation values in localized systems. We can take it as a good evidence of delocalization of \( 5f \) moments in the present system. The drastically different \( \Theta_p \) values from the high- and low-temperature region point to very strong magnetocrystalline anisotropy in the paramagnetic range.

The magnetic ordering at \( T_C = 9.5 \) K is reflected by a relatively broad peak in the \( C_p/T \) vs \( T \) curve [Fig. 1(b)]. No other anomalies are seen between 1.6 and 250 K. The linear extrapolation of \( C_p/T \) vs \( T^2 \) from the region 1.6–8 K to \( T = 0 \) K gives a tentative \( \gamma \) value of 185.6±1.3 mJ/mol K\(^2\). A much better fit can, however, be obtained by the formula
\[
C/T = \gamma + \beta T^2 + \delta T^2 \ln(T)
\]
which involves the contribution of spin fluctuations through the additional logarithmic term. For the temperature region 1.6–9 K, the fitting parameters: \( \gamma = 180.9 \pm 0.6 \) mJ/mol K\(^2\), \( \beta = 2.4 \pm 0.1 \) mJ/mol K\(^2\) and \( \delta = -0.5 \pm 0.05 \) mJ/mol K\(^2\) were obtained. To estimate the magnetic entropy we tried to subtract the Debye function with a probable value of Debye temperature \( \Theta_D \). This task turns out to be very difficult. It seems impossible to assign one single value of \( \Theta_D \). The best agreement in the high-temperature part can be obtained for \( \Theta_D = 290 \) K together with the assumption of \( \gamma = 20 \) mJ/mol K\(^2\). In this way, we obtained by integrating \( C(T)/C_{Debye}(T) \) in the temperature region 0–100 K, a value of 9.1 J mol\(^{-1}\) K\(^{-1}\) for the magnetic entropy. This means a value of 1.58 \( R \ln 2 \) or, through the expression \( R \ln(2J + 1) \), a value for \( J \) of 1.0. In fact, this value represents the estimated upper limit. The magnetic entropy connected with the magnetic transition is only a very small fraction of this value, the rest originates probably from magnetic fluctuations.

The onset of magnetic ordering at \( T_C = 9.5 \) K is reflected also by the maximum in the temperature derivative of the electrical resistivity [Fig. 1(c)] near this temperature. The resistivity in the paramagnetic range is slightly decreasing with lowering temperature. It drops significantly below \( T_C \), where also a quadratic temperature dependence with normalized parameters \( \rho_0 = 0.4088\pm0.0004 \) and the quadratic coefficient \( A = 3.292\pm0.009\times10^{-3} \) K\(^{-2}\) is observed.

The high-field magnetization measured at 4.2 K up to 35 T on a field-aligned powder sample saturates slowly and at 35 T it reaches 0.67 \( \mu_B/\text{f.u.} \) (\( M_{\text{free}} \)). A value of 0.3 \( \mu_B/\text{f.u.} \) is obtained by extrapolation to zero magnetic field. For a powdered sample with randomly oriented grains, somewhat smaller values of 0.52 \( \mu_B/\text{f.u.} \) (\( M_{\text{fix}} \)) and 0.23 \( \mu_B/\text{f.u.} \), respectively, are recorded. Although the magnetization is by far not yet saturated in 35 T the ratio \( M_{\text{free}}/M_{\text{fix}} = 0.78 \) can be taken as a supporting indication for an easy-plane anisotropy.

The low-field magnetization measured at different temperatures on a fixed-powder sample are shown in Fig. 2, where also some points from the previous high-field magnetization measurements (big open points) are displayed. It is clear that both data sets compare well and that hysteresis effects set in at low temperatures, as can be seen from the inset in Fig. 2 yielding a value of the remanent magnetization of 0.10 \( \mu_B/\text{f.u.} \).
the size of magnetic moments 0.11

The magnetic structure was determined by fitting to the experimental data and the fit. The best fit. The full line at the bottom represents the difference between the experimental data and the fit.

IV. NEUTRON DIFFRACTION RESULTS
Two possible crystallographic structures were considered: the CeCu2 type of structure (space group Imma) and its ordered version TiNiSi (space group Pnma). The neutron-diffraction patterns obtained above the transition temperature, namely at 80 K (Fig. 3) and 24 K, can be indexed using the orthorhombic TiNiSi type of structure. Note that the lines indexed as 102, 111, 200, 112, 202, and 211 would not be present for the CeCu2 type. The structural parameters determined at 80 K are summarized in Table I. The absence of any unindexed reflection proofs the very low content of the URhSi secondary phase indicated by the electron microprobe analysis. A considerable improvement of fit is achieved by introducing a slightly higher fraction of U (0.9%) and a slightly lower atomic fraction of Rh (~0.6%). This is in a good agreement with the electron-microprobe analysis.

The spectrum recorded at 2.8 K (well below Tc inferred from bulk experiments) contains no additional reflections, but only additional magnetic contributions to the 101, 011, 200, 112, 202, and 211 nuclear Bragg reflections. This is consistent with identical magnetic and crystallographic unit cells. The magnetic structure was determined by fitting to the models possible within the experimental constrain. The best agreement was obtained for the model with all U moment parallel to the c axis, i.e., for the ferromagnetic model with the size of magnetic moments 0.11 ± 0.02 μB/U atom.

V. CONCLUSIONS
URhSi crystallizes in the orthorhombic TiNiSi-type of structure (space group Pnma) and orders ferromagnetically below 9.5 K with ordered U moments of about 0.11 μB. This rather small value in comparison with most UTX compounds together with magnetization, susceptibility, specific-heat, and resistivity behavior suggests that the magnetism in URhSi is governed by strongly delocalized uranium 5f electron states. The origin of this itinerant behavior can be found mainly in the strong 5f-ligand hybridization.

The ordered U moment determined by the neutron-diffraction is much smaller that the value obtained from the high-field magnetization experiments. This fact points to field-induced moments in the latter experiment. The presence of magnetic fluctuations which play an important role at zero field even well above the transition temperature can be inferred from the bulk measurements. Studies on single-crystalline URhSi are highly desirable to get more specific information on substantial issues of magnetism in URhSi as are the magnetocrystalline anisotropy, orbital and spin moments, etc.

ACKNOWLEDGMENTS
This work was sponsored by the “Stichting voor Fundamenteel Onderzoek der Materie” (FOM), and The Grant Agency of the Czech Republic (Project No. 202/93/0184). Support to K. P. in the framework of the E. C. funded training program HC & M is acknowledged. The work of V.S. and P.S. at HMI Berlin was supported by program PECO.