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Magnetization densities in URhSi studied by polarized neutron diffraction

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Polarized neutron experiment has been performed on a URhSi single crystal at 2 K with field applied along the *c* axis that is the easy magnetization direction. Maximum entropy approach has revealed that the magnetization distribution is significantly prolongated along the easy magnetization direction. Refinement of the magnetic structure factors yields the spin ($0.33\mu_B$) and orbital components ($0.99\mu_B$) of the uranium magnetic moments that are oriented antiparallel to each other and strongly reduced with respect to the free U^{3+} or U^{4+} ion. A small induced moment on the Rh atoms of about $0.05\mu_B$ has been found. The difference with respect to total bulk magnetization is ascribed to the polarization of the conduction electrons. The deduced ratio between the orbital and spin part suggests that $5f$ electron moments are delocalized and close to the U^{4+} ionic state. The agreement with theoretical calculations is only limited.

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I. INTRODUCTION

The magnetic and electronic properties in general of uranium intermetallic compounds are dominated by the $5f$ electron states. They are not well screened by outer electrons as the $4f$ electron states in the rare-earth compounds. As a result, $5f$ states are always to a certain extent delocalized. There are two main delocalizing mechanisms: (1) the direct overlap between $5f$-electron wave functions of neighboring uranium atoms and (2) the hybridization between the uranium $5f$ states and the electron states of neighboring atoms (ligands). Both delocalization mechanisms depend strongly on the crystallographic details in the uranium atom surrounding (the coordination number, symmetry, nature of ligands, and their distances). As the number of various crystal structure types is more than a handful, it is not at all surprising that materials containing $5f$ electrons show a large variety of intriguing electronic properties.[1,2] Depending on the constituent *X* and *T* components, the magnetic properties of these compounds span from the Pauli paramagnetism via spin-fluctuation effects and metamagnetism to a long-range magnetic ordering with stable uranium magnetic moments. There are even two ferromagnetic superconductors [URhGe (Ref. 3) und UCoGe (Ref. 4)] known to date among the orthorhombic U*TX* compounds. The development in the magnetic properties is clearly observable when moving from the left to the right within the Periodic Table for a given *d* series as a result of reduced $5f$-d hybridization due to gradual filling of the *d* band. Similar tendency can be found also for movement from the top to the bottom within a column or for *X* constituents.

In order to reduce the number of "free parameters," systematic studies of large groups of isostructural compounds having the same geometrical surrounding of $5f$ ions can play an essential role in deeper understanding of the physics of $5f$ intermetallics. Indeed, the systematic investigation of isostructural groups of U*TX* compounds (*T*: a transition metal from the end of a *d*-electron series and *X*: a *p*-electron element) revealed clear tendencies in the type of magnetic ordering, direction of magnetic moments, and thermal and electronic properties with respect to *T* and/or *X* species.[1,2] There are several crystal structure types within the U*TX* intermetallics. Besides the most populated group of compounds possessing the hexagonal ZrNiAl type of structure, the second largest group of U*TX* intermetallics exhibits the orthorhombic TiNiSi or a closely related orthorhombic $CeCu_2$ type of structure.

As a result of a large spatial extent of $5f$ orbitals, most of the $5f$ compounds exhibit strong magnetocrystalline anisotropy, the type which depends on the crystal structure. For hexagonal structures uniaxial anisotropy is found; for the orthorhombic one an easy-plane magnetocrystalline anisotropy is realized. Early polarized neutron-diffraction experiments suggested that the strong magnetocrystalline anisotropy in the case of the hexagonal compounds URuAl (Ref. 5) and URhAl (Ref. 6) originates from a strongly anisotropic hybridization between the uranium $5f$ states and the *d* states of the transition metals. Relatively large magnetic moment induced by the $5f$-d hybridization has been observed on transition-metal site, which lies in the basal plane together with the U atoms. The other transition-metal site building *T*-*X* plane, which is approximately at the same distance from uranium, was found not to carry sizable magnetic moment. On the contrary, later but similar polarized neutron diffraction on UNiAl and UNiGa (Ref. 7) showed that for these two compounds a larger magnetic moment is found on the transition-metal site within the basal plane containing no U atoms. However, a nonzero magnetization density has been found in a region that does not correspond to any occupied crystallographical position, and it was speculated that it is caused by a small fraction of interstitially positioned Ni atoms.[7,8] In the case of UCoAl (Refs. 9 and 10) comparable moments on both cobalt sites have been reported, and for UPtAl (Ref. 11) the size of induced moments was dependent on the method used. To our knowledge, no magnetization density studies have been published forU*TX* compounds crystallizing in the orthorhombic structure. Similar study, however, exists for ferromagnetic superconductor UGe_2.[12] This fact has motivated us to undertake such a study for ferromagnetic URhSi.

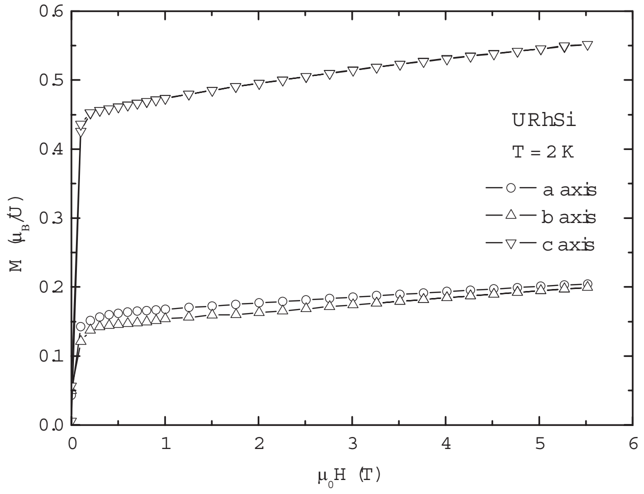

FIG. 1. The field dependence of the magnetization of URhSi in fields up to 5.5 T applied along the principal axes at the temperature of 2 K (after Ref. 21).

URhSi may be considered as a material, which is situated in the crossover region between magnetically ordered and paramagnetic materials. It has been studied experimentally by several groups in a polycrystalline form[13–18] or using single crystals.[19–21] It crystallizes in the TiNiSi type of structure (space group *Pnma*) in which U atoms form zigzag chains along the *a* axis.

Bulk properties point to a ferromagnetic (*F*) order in URhSi with Curie temperature of 10.2 K for a single crystal.[19–21] A spontaneous moment of about $0.3\mu_B$/f.u. has been derived from the early free-powder magnetization experiments performed at 4.2 K.[16] The neutron powder-diffraction data obtained at low temperatures suggested an existence of ferromagnetically ordered U moments oriented along the *c* axis. Their magnitudes, however, ranging from 0.11 (Ref. 17) to $0.5\mu_B$ (Ref. 18) were a matter of dispute for some time. Single crystal neutron data[21] finally resolved some of the controversial observations in the literature. Collinear U moments of $0.58\mu_B$ oriented along the *c* axis have been found. However, magnetization measurements[19–21] contradict neutron-diffraction data. Namely, significant *a* and *b* axis components (Fig. 1), both of the order of $(0.11-0.15)\mu_B$/U, have been observed. This would suggest that U moments are in URhSi tilted out from the *c* axis. Our attempts to observe in unpolarized neutron experiments such a tilt, however, failed.[21] Such an unsatisfactory situation only adds a reason to the motivation mentioned above to perform a polarized neutron-diffraction study on URhSi.

II. EXPERIMENTAL

It is well known that in order to get accurate results from the polarized neutron experiment one has to know rather well the crystal structure which is usually in the course of the refinement kept fixed. Therefore, two types of neutron-diffraction experiments have been performed. To refine the atomic positions and other nuclear structure parameters including the extinction parameters of our crystal, integrated intensities of nuclear reflections have been collected on the multicounter diffractometer E2 installed at the Berliner Neutron Scattering Centre. The incident wavelength used in the experiment was 1.21 Å. The crystal was mounted in an orange cryostat in several orientations in order to be able to collect as many as possible nuclear Bragg reflections. In total, we have measured at 20 K (i.e., above the magnetic phase transition) 64 unique reflections. In order to refine the structure parameters of URhSi the SORGAM and POLSQ programs of the Cambridge Crystallography Subroutine Library[22] were used for the refinement. The Becker-Coppens secondary-type extinction correction[23] that describes the decrease in reflection intensities due to an angular distribution of large mosaic blocks have been applied. The effect of the extinction turned out to be rather weak, only about 5% of the intensity for the three most intense reflections, and led to a crystal mosaicity of 6 arc min. The plot of observed intensities versus calculated ones after the use of extinction and correction for Lorentz geometrical factor is shown in Fig. 2. The tabulated values of the scattering length of atoms present in the sample were used.[24] Because no sizable effect of anisotropic factors on the refined parameters or the fit quality has been found we have restricted ourselves to the use of isotropic temperature factors. The quality of the fit could be improved slightly by lowering the occupancy at the Si site by about 4 at. % in agreement with previous results.[21] The final results of our crystal structure refinement are summarized in Table I.

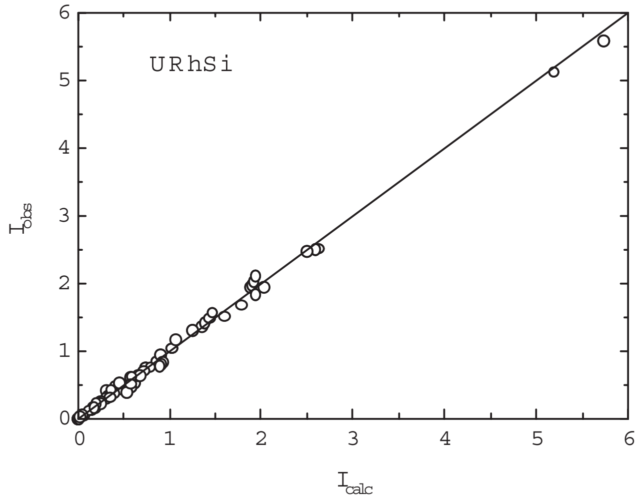

FIG. 2. Plot of the observed versus calculated intensities collected on URhSi single crystal after correction for the extinction.

The polarized neutron experiment has been carried out on the very same single crystal used in previous magnetic bulk measurements[19,20] and previous unpolarized neutron experiment.[21] The details regarding the sample preparation and characterization can be found in Ref. 21. It should be noted that a slight deficiency of Si has been found previously in the unpolarized neutron experiment and using the electron probe analysis. The crystal had a shape of a cube $2\times2\times2$ mm^3 with principal axes perpendicular to the cube faces. The flipping ratios were measured on the lifting-counter diffractometer 5C1 using neutrons with $\lambda=0.845$ Å obtained with a Heusler alloy monochromator in-

TABLE I. Structural parameters of URhSi determined at 20 K.

URhSi	Site	Positions	B_{iso} (Å)	Occupancy
U	4*c*	$(x_{\text{U}}, 1/4, z_{\text{U}})$ $x_{\text{U}}=0.0015(14)$ $z_{\text{U}}=0.1941(11)$	0.07(2)	1.00 (fixed)
Rh	4*c*	$(x_{\text{Rh}}, 1/4, z_{\text{Rh}})$ $x_{\text{Rh}}=0.1425(3)$ $z_{\text{Rh}}=0.5793(1)$	0.26(3)	1.00 (fixed)
Si	4*c*	$(x_{\text{Si}}, 1/4, z_{\text{Si}})$ $x_{\text{Si}}=0.7807(8)$ $z_{\text{Si}}=0.6015(2)$	0.41(8)	0.96(1)
R factors:		$\chi^2=2.44$		$R=5.6\%$

stalled at the ORPHÉE 14 MW reactor of the Léon Brillouin Laboratory, CEA/CNRS Saclay. The polarization of the incident neutron beam was 91%. A magnetic field of 6 T has been applied along the *c* axis (with precision of about 1°) which has been found previously to be an easy magnetization direction. In total, we have collected at 2 K and field of 6 T 544 reflections (132 unique ones) of the $(h\ k\ 0)$, $(h\ k\ 1)$, and $(h\ k\ 2)$ type within the $0.098 \le \sin\theta/\lambda \le 0.956$ range.

III. RESULTS OF THE POLARIZED NEUTRON EXPERIMENT

A. Direct refinement of the flipping ratios

One way to treat the experimental data is the direct refinement of the measured flipping ratios. We assume all the magnetic moments to be centered on the given atomic sites. Various atoms are characterized by appropriate magnetic form factors $f(\mathbf{Q})$ that have in general orbital (μ_L) and spin (μ_S) parts. The magnetic amplitude of elastic neutron scattering at the scattering vector $\mathbf{Q}$ from a magnetic ion with the moment μ is then proportional to $(\mu_L+\mu_S)f(\mathbf{Q})=\mu f(\mathbf{Q})$. While for the Rh atom we considered the magnetic form factor having only spin part, on the U site, we took into account both contributions. The uranium magnetic form factor is usually expressed by formula $f(\mathbf{Q})=\langle j_0(\mathbf{Q})\rangle + C_2\langle j_2(\mathbf{Q})\rangle$ valid in the dipole approximation. Here $C_2=\mu_L^{\text{U}}/(\mu_S^{\text{U}}+\mu_L^{\text{U}})=\mu_L^{\text{U}}/\mu^{\text{U}}$ depends on the hybridization degree of the 5*f* electrons and j_i are the radial integrals. Unfortunately, since these integrals for the magnetic form factors of the U^{3+}, U^{4+}, and U^{5+} ions are very close the data are not sensitive to the uranium valence at all.[25] On the contrary, the $\mu_L^{\text{U}}/\mu_S^{\text{U}}$ ratio depends very strongly on the degree of hybridization.[26] The stronger hybridization (strong coupling) causes the $\mu_L^{\text{U}}/\mu_S^{\text{U}}$ ratio to be smaller. A fit using magnetic form factors of the U^{3+} without magnetic moment on the Rh site gives an agreement factor of $\chi^2=9.8$. Including the Rh moment in the fit decreases the χ^2 value by a factor of 2. Finally relaxing of constraints between the spin and orbital parts of the U moment improves the fit by an additional factor of 2 and gives $\chi^2=2.44$. The results of such a type of fit are summarized in Table II. From these results one can calculate the coefficients C_2 and $R_L=C_2/(C_2-1)=-\mu_L^{\text{U}}/\mu_S^{\text{U}}$, which are tabulated as well. The experimental values $\mu^{\text{U}}=0.66(2)\mu_B$ and $\mu_L^{\text{U}}=0.99(4)\mu_B$ are to be compared with the theoretical ones calculated within the generally accepted intermediate coupling scheme.[27] These values are given for the U^{3+} and U^{4+} free ions at the bottom of Table II. In Fig. 3 the observed values and the calculated uranium form factor (multiplied by the total U magnetic moment and assuming a U^{3+} valence, respectively) are shown. Clearly, a small amount of reflections do not obey the smooth expected curve. One possible reason of that might be a nonzero polarization of the interstitial regions.

TABLE II. Magnetic moments and related parameters of URhSi determined at 2 K with field of 6 T applied along the *c* axis resulting from fitting to the direct refinement of the flipping ratios. The value μ^{cond} ascribed to the polarization of the conduction electrons has been obtained by comparing the sum of the moments resulting from the polarized experiment and the bulk magnetization. The theoretical values for the C_2 and R_L calculated within the intermediate coupling scheme are given at the bottom of the table. The theoretical values according to Refs. 14 and 31 resulting from fully relativistic spin-polarized calculations including an orbital polarization term and the empirically estimated value of the Racah parameter $E'=2.6$ mRy, the value $E''=4.3$ mRy calculated *ab initio* from Slater-type integrals, and spin-polarized relativistic linearized-augmented plane-wave methods are listed as well.

	URhSi		
	This work	Theor[a]	Theor[b]
μ^{bulk} (μ_B)	0.58(1)		
μ^{U} (μ_B)	0.66(2)	0.09,[c] 0.28[d]	0.3[e]
$-\mu_S^{\text{U}}$ (μ_B)	0.33(6)	0.15,[c] 0.20[d]	1.27[e]
μ_L^{U} (μ_B)	0.99(4)	0.24,[c] 0.48[d]	1.57[e]
μ_{Rh} (μ_B)	0.05(2)		
Deduced values			
$C_2=\mu_L^{\text{U}}/\mu^{\text{U}}$	1.50(10)	2.66,[c] 1.71[d]	5.23[e]
$R_L=-\mu_L^{\text{U}}/\mu_S^{\text{U}}$	3.00(32)	1.60,[c] 2.40[d]	1.23[e]
$\mu^{\text{cond}}=\mu^{\text{bulk}}-\mu^{\text{U}}$ (μ_B)	−0.12(3)		
Free ion values	U^{4+} $(5f^2)$	U^{3+} $(5f^3)$	
$C_2=\mu_L^{\text{U}}/\mu^{\text{U}}$	1.42	1.63	
$R_L=-\mu_L^{\text{U}}/\mu_S^{\text{U}}$			

[a] Reference 14.
[b] Reference 31.
[c] Value of the Racah parameter $E'=2.6$ mRy.
[d] Value $E''=4.3$ mRy calculated *ab initio* from Slater-type integrals.
[e] Spin-polarized relativistic linearized-augmented plane-wave methods.

The obtained C_2 parameter is close to the theoretical value for the U^{3+} free ion. On the Rh atoms a moment of $\mu^{\text{Rh}}=0.05(2)\mu_B$ has been refined. However, when comparing the total magnetic moment determined from the neutron $[\mu=\mu^{\text{U}}+\mu^{\text{Rh}}=0.66(2)+0.05(2)=0.71(4)\mu_B$, all of them along the *c* axis] and bulk measurements $[0.58(1)\mu_B]$,[21] a discrepancy of $0.12(5)\mu_B$ is found. There are several possible sce-

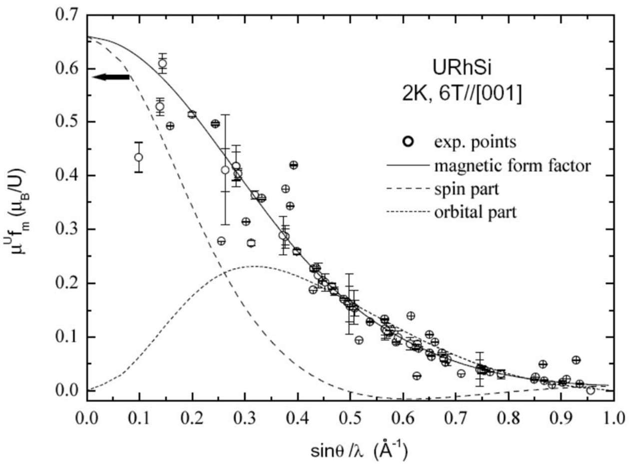

FIG. 3. Uranium magnetic form factor in URhSi multiplied by μ^U, as a function of sin θ/λ. The open points represent the measured experimental data. The full, dashed, and dotted lines represent the best fit supposing the U^{3+} configuration, spin, and orbital parts only, respectively. The bulk magnetic moment deduced from the magnetization measurement is denoted by the arrow.

narios to explain the discrepancy. Among the most probable ones is a possibility that some interstitial position is polarized and/or conduction electrons significantly contribute to the magnetization. That is why we have used a maximum entropy method to identify the origin of the discrepancy.

B. Maximum entropy method: Magnetization density maps

The maximum entropy gives the most probable magnetization distribution map compatible with the measured structure factors and their experimental uncertainties.[28] It is more powerful than the usual Fourier synthesis since it does not make any *a priori* assumption concerning the unmeasured Fourier components. As a result, it reduces both the noise and truncation effects. At the same time, no detailed atomic model is needed for the refinement. All the required information is the space group of the material, lattice constants, experimental flipping ratios together with corresponding measured or calculated nuclear structure factors, and other applicable correction parameters. For the refinement, we have used the program MEND.[29] The unit cell of URhSi was divided into $82\times82\times82=551\,368$ cells, in which the magnetization is assumed to be constant. The reconstruction was started from a flat magnetization distribution with a total moment in the unit cell equal to the bulk magnetization measured experimentally at 2 K with field of 6 T applied along the *c* axis (according to Ref. 21 the unit-cell magnetization in URhSi is $4\times0.58\mu_B$). As the local magnetization density can also be negative, one has a certain degree of freedom to select the maximal and minimal values with a difference that gives the bulk magnetization. In the course of the refinement we have tried several pairs of values. All the resulting density maps were quite similar.

As the final result, we have obtained the most probable reconstructed three-dimensional density of magnetic moment, i.e., the map which fits the data and for which the entropy is maximum. A common way to represent such a density is a projection of several density layers onto a certain crystallographic plane.

In Fig. 4(a) we show the projection of half of the crystallographic unit cell $[y=(0,0.5)]$, a region where all three different atoms are situated on the *a*-*c* plane. In Figs. 4(b) and 4(c) we show similar projections on the *b*-*c* plane $[x=(0,0.5)]$ and onto the *a*-*b* plane $[z=(0,0.5)]$, respectively. Relevant atomic positions are shown in Figs. 4(d)–4(f), respectively. As can be easily seen, the major part of the positive density is clearly centered at uranium positions that are shown by blue-filled circles. However, smaller but significant clouds are seen at positions that do not correspond to Rh (red-filled circles) nor Si (green-filled circles) atoms. This is at best seen in Figs. 4(a) and 4(d). Both Rh and Si atoms reside rather at the border between the positive and negative polarized regions (the zero-polarized boundary is shown by thick solid line and negative is shown by dashed lines) but they are clearly connected with U atoms by bridges of positive polarization that do not correspond to neither atomic position.

The density distribution is very anisotropic. Comparison of Figs. 4(a)–4(c) shows clearly that the magnetization density has shape of a "cigar" oriented along the *c* axis. This elongation parallel to the *c* axis is probably due to a low vertical resolution typical for a lifting-counter scattering geometry, but it corresponds at the same time well with the easy magnetization direction in URhSi.

IV. DISCUSSION

The obtained results show quite clearly that the magnetism in URhSi is caused mainly by uranium moments. The orbital and spin parts have been found to be oriented antiparallel to each other. The induced magnetic moment on the Rh sites deduced from the direct refinement of the flipping ratios is about $0.05\mu_B$. Similarly, a magnetic moment has been found on transition metal in various uranium equiatomic compounds.[5–8,10,11] However, one cannot compare these values directly as the present compound possesses another crystal structure. No magnetic moment has been found in our system at the position of silicon atoms. It is interesting to note that the magnetization density map suggests that the cloud of positive magnetization is not exactly situated on the rhodium atoms but in a region that connects rhodium and silicon with uranium atoms. As has been discussed, e.g., in Refs. 7 and 10, such a nonzero magnetization distribution could be caused by crystallographic disorder that would mix transition-metal atoms and the *p*-element atoms and/or by a certain distribution of transition-metal atoms in the interstitial regions. As we can practically reject mixing or interstitial uranium atoms, the disorder in the transition-metal or *p*-element sublattice would need to be rather significant. In our case it is not that easy to identify such a disorder as the scattering lengths of Rh (5.88 fm) and Si (4.15 fm) are not that different (the scattering length of uranium is 8.42 fm). It seems that the only remaining explanation is the polarization of the conduction electrons along the transition-

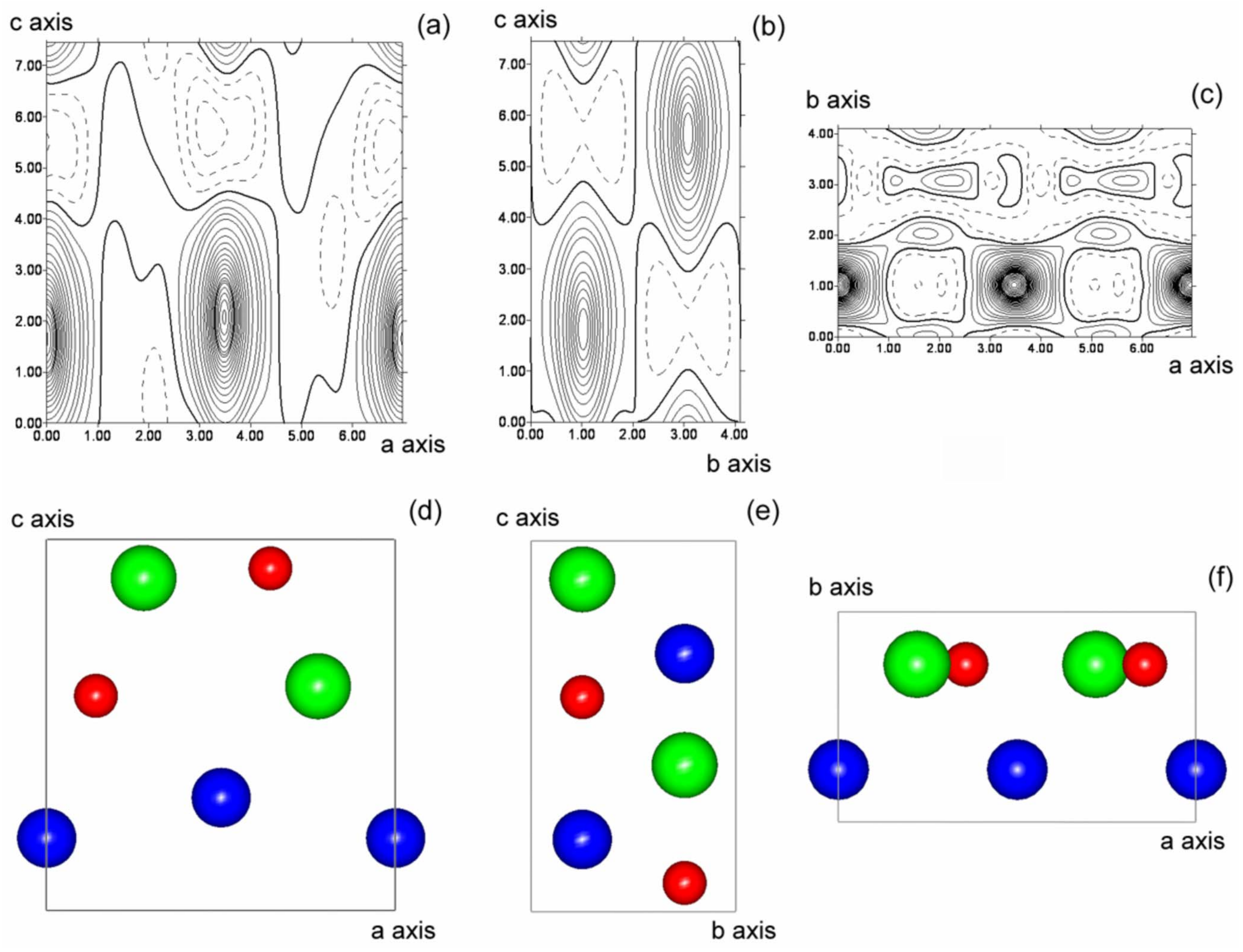

FIG. 4. (Color online) Projections of the magnetization distribution onto (a) the *a*-*c*, (b) *b*-*c*, and (c) *a*-*b* planes. For comparison, crystallographic unit cell is projected onto the relevant plane in panels (d), (e), and (f). In all cases only half of the unit cell is projected. The U atoms are shown in blue (middle-size spheres), Rh in red (smallest spheres), and Si in green (largest spheres).

metal–*p*-element–uranium atoms bonds. Comparing the total sum of all the moments with the bulk magnetization value, one can deduce the total conduction-electron polarization of $-0.12(3)\mu_B$. This "moment" is oriented parallel to the uranium spin moment, i.e., antiparallel to the total magnetic moment residing on uranium atoms. Negatively polarized conduction electrons were found in many uranium compounds. Among them, also in UGe$_2$, in which some of the Ge atoms (those lying in the same plane as uranium atoms) were found to carry non-negligible magnetic moment.[12] In our study, however, we have found negligible polarization of Si atoms.

Unfortunately, the magnetic form factors of U^{3+} and U^{4+} are nearly identical. That is why the refinement using just the radial functions j_i cannot reveal the valency of uranium atoms that is connected with the hybridization of the 5*f* states. However, the ratio $R_L=-\mu_L^U/\mu_S^U$ depends strongly on the degree of hybridization.[26] Our measurements providing both spin and orbital values with quite high precision give $R_L=3.00(32)$. This value lies between the R_L values expected for the U^{4+} and U^{3+} configurations (both calculated within the intermediate coupling scheme). The associated parameter $C_2=1.50(10)$ is slightly higher than that expected for the U^{4+} but smaller than one for the U^{3+} configuration. As has been shown in previous experiments a delocalization of 5*f* states causes a decrease in the value of the orbital part of the magnetic moment leading thus to reduction in the R_L value.[30] The experimental values thus suggest that the magnetism in URhSi is caused by itinerant 5*f* electrons that are closer to the U^{4+} configuration. This finding is in agreement with the theoretical calculations[14,31] that suggest that the magnetism in URhSi is of an itinerant nature due to hybridization of U 5*f* states with Rh 4*d* states. However, the agreement with theory is far from perfect. Striking is the fact that the observed R_L value is higher than the one found in other typically itinerant uranium intermetallics. For instance, in UGa$_3$, UNiAl, UNiGa$_5$, and UPtGa$_5$ ranges R_L value between 1.66 for the former and 2.10 for the latter system.[7,32,33] This suggests that 5*f* electronic states are in URhSi more localized than in these systems. On the other hand, R_L value in URhSi is clearly smaller than the value found for UAsSe ($R_L=3.2$).[34]

Moreover, the experimentally determined values of moments are much smaller than those expected for a free U^{3+}

($\mu_L^{3+}=5.585\mu_B$, $\mu_S^{3+}=-2.169\mu_B$) or U^{4+} ions ($\mu_L^{3+}=4.716\mu_B$, $\mu_S^{3+}=-1.432\mu_B$), respectively. Comparison with various theoretical calculations does not provide a good agreement. The calculation based on fully relativistic spin-polarized calculations including an orbital polarization term and the Racah parameter $E''=4.3$ mRy calculated *ab initio* from Slater-type integrals leads to values of the orbital and spin parts of the uranium moments that are roughly half of those found experimentally (see Table II, Ref. 14, superscript d). Other calculations (see Table II) yield either much smaller or much larger values except for the fact that all of them suggest a significant cancellation of the orbital and spin parts, in agreement with the experimental finding.

V. CONCLUSIONS

We have presented results of polarized neutron-scattering measurements performed on a single crystal of URhSi in its ferromagnetic state. The polarized neutron-scattering measurements allowed after correction for extinction obtained from a separate unpolarized neutron experiment the determination of the $5f$ electron orbital and spin contributions in field of 6 T applied at 2 K along the c axis. The experimentally determined ratio R_L between orbital and spin moment values that serves as a measure of the hybridization strength between uranium $5f$ and the surrounding electronic states is higher than the free-ion U^{3+} value, however, smaller than that for the U^{4+} configuration. This feature is the signature of rather moderate hybridization of $5f$ electrons compared to other itinerant uranium-based intermetallics. We point out that small and positively induced magnetization clouds are detected at positions that do not correspond to Rh nor Si atoms. The conduction electrons are found to be polarized in the same direction as uranium spins.

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