PHYSICAL REVIEW B, VOLUME 65, 144429

Canted ferromagnetic structure of UNiGe in high magnetic fields

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(Received 19 May 2001; revised manuscript received 20 December 2001; published 1 April 2002)

UNiGe adopts the orthorhombic TiNiSi-type of structure and exhibits in zero field two different antiferromagnetic structures. Both are noncollinear with significant *a* axis component. Magnetic measurements indicate two metamagnetic-like transitions with the field applied along the *b* or the *c* axis. Neutron diffraction studies in magnetic fields applied along the *c* axis reveal above the second metamagnetic-like transition a canted ferromagnetic structure. U magnetic moments of $1.16\pm0.04\mu_B$ exhibit a ferromagnetic *c* axis component of $1.04\pm0.04\mu_B$ and an antiferromagnetic *a* axis component of $0.52\pm0.05\mu_B$. Observation of such a canted ferromagnetic structure in UNiGe provides strong evidence for presence of anisotropic exchange interactions. The fact that the *a* axis component cannot be well aligned above the second metamagneticlike transition indicates that the antiferromagnetic interaction between the *a* axis components is much stronger than between the others.

DOI: 10.1103/PhysRevB.65.144429 PACS number(s): 75.25.+z, 75.30.−m, 61.12.−q, 75.50.Cc

I. INTRODUCTION

Systems with *f* electrons display a rich variety of cooperative phenomena as different types of magnetic ordering, superconductivity, or heavy-fermion state, which are mostly very sensitive to variations of external variables such as magnetic field.[1] Although related to one-electron background, these phenomena are characterized by strong electron-electron correlations. Recently, progress has been made in description of magnetic properties of these compounds using a semiphenomenological approach based on hybridization of *f* states with other electron states. The hybridization is strongly anisotropic and affects both the delocalization of $5f$ states and intersite magnetic coupling effects.[2] To draw conclusions regarding general relations between type of coupling, directions of magnetic moments, and type of crystal structures, single crystals of good quality have to be studied. In this contribution we address these issues in view of a newly determined field-induced magnetic structure of single-crystalline UNiGe studied by means of neutron diffraction in fields up to 14.5 applied along the *c* axis.

II. BULK PROPERTIES

UNiGe is one of the most extensively studied equiatomic uranium ternary compounds that adopt the orthorhombic TiNiSi-type of structure[3–6] in which U atoms form zig-zag chains running along the *a* axis (with an amplitude of 0.1 c). The shortest U-U distance of 3.547 Å is found within the chains. The separation of chains of 3.653 Å gives the next-nearest U-U distance. Magnetic measurements revealed that UNiGe orders magnetically below $T_N=50$ K with an additional magnetic phase transition at 42 K (Refs. 1,4,7–9) although at first, the transition at 50 K has been overlooked.[10,11] The low-temperature magnetic structure, determined both by non-polarized and polarized neutron-diffraction experiments,[6,12] is commensurate with a propagation vector $\boldsymbol{q}=(0,1/2,1/2)$ (see Figs. 1 and 2 in Ref. 12). The magnetic structure between 42 K and T_N is reported to be incommensurate. Both structures are noncollinear with significant *a* axis components ($\mu_x=0.35\mu_B/U$ at 20 K).

Both magnetic structures can be modified by application of a magnetic field along the *b* or along the *c* axis. At 4.2 K, there are two metamagnetic-like transitions, at 2.5 and 9.5 T, in UNiGe if the field is applied along the *c* axis and two (at 17 and 25 T) with the field applied along the *b* axis.[1,4,7] While the magnetic structure above the highest critical field is forced ferromagnetic for both field orientations, in the intermediate region uncompensated antiferromagnetic structures exist.[1,8,9,12] For the *a* axis orientation no metamagnetic-like transition is found up to 38 T.[7] Strong magnetic anisotropy persists also in the paramagnetic state.

III. EXPERIMENTAL METHODS

A single crystal of UNiGe used for present experiment originates from the same batch as that used previously for magnetic measurements[4,7] and neutron-scattering experiments.[6,8,9,12] The crystal has been grown from a stoichiometric melt by a modified tri-arc Czochralski technique in continuously gettered Ar atmosphere at FOM-ALMOS center at the University of Amsterdam. X-ray Laue diffraction and electron microprobe analysis revealed an excellent quality of the crystal. A small sphere with a diameter of 3 mm was spark-eroded from this crystal and used in the neutron-diffraction experiments.

The integrated intensities were measured on the E4 normal beam diffractometer at the BER-II reactor at Hahn-Meitner-Institute, Berlin. The crystal was mounted with its c axis parallel to the rotational axis of the diffractometer, which was also the direction of the applied field. Magnetic field up to 14.5 T was produced using a superconducting split-pair cryomagnet manufactured by Oxford Instrument, which offers currently the world's highest static magnetic field in connection with neutron research. The fact that the split has only 20 mm and additional $\pm 2°$ opening restricted us to a $(hk0)$ diffraction plane. The crystal was wrapped in an aluminum foil and measured with an incident-neutron wavelength of 2.44 Å. The E4 diffractometer is equipped with a single detector and $\lambda/2$ contamination filter leaving residual contamination on the level of less than 1×10^{-3}.

The single crystal was oriented using several sufficiently strong and well centered nuclear reflections. The individual ω scan profiles were analyzed by the Lehman-Larson algorithm[13] and by fitting to a Gaussian profile.

The crystallographic and magnetic structures were determined by fitting procedures using the program FULLPROF.[14] The function minimized during least-squares refinement was $\Sigma w|F_{\text{obs}}-F_{\text{calc}}|^2$, $w=1/\sigma^2$. The scattering lengths were taken from Ref. 15 and the U^{3+} $(\langle j_0\rangle + c_2\langle j_2\rangle)$ magnetic form factor from Ref. 16. During the experiment we have collected three identical sets of reflections (so-called rocking curves) at 60 K and zero field, at 2 K and zero field and at 2 K and at 12 T, a field which is well above the upper metamagnetic-like transition. Moreover, we have followed intensity of few reflections as a function of magnetic field up to 14.5 T.

IV. RESULTS

Using 40 (13 inequivalent) reflections observed at 60 K, i.e., in the paramagnetic state, we confirmed that UNiGe forms in the structure that conforms with the orthorhombic TiNiSi-type structure (space group *Pnma*). Among the observed reflections were also (110) and (310) peaks, which are forbidden for space group *Pnma*. However, by measuring (110) type reflections with $\lambda/2$ wavelength (graphite filter removed) we were able to prove that these reflections originate from a multiple scattering, proving thus a very good quality of our crystal. This conclusion was possible to make thanks the fact that the scattering angle does not depend linearly on the wavelength and that no other reflection is allowed at the scattering angle of the (110) type of reflections. As a consequence, some of the low-indexed reflections are measured with a certain uncertainty leading to an error in the structural determination and the overall scaling factor. Affected reflections appear to be smaller. The intensity due to magnetic order is affected in the very same manner leading to incorrect moment magnitudes and/or to direction of moments. However, the appearance of intensity on certain nuclear Bragg reflections (as discussed below) suggest that main conclusions of this contribution are independent on this problem.

During the fit of integrated intensities onto the structural model we had to fix the z-position numbers for all three atoms because of limited set of reflections with $l=0$. Results of the fit, which included absorption, scale factors, x-position numbers and isotropic temperature factors of the three atoms as free parameters compare well with previous publications. Similar results were obtained from the data set collected in zero field at 2 K. Note that in zero field below T_N we were unable to observe any magnetic reflections because propagation vectors for both zero-field magnetic structures have both the k and l components. We were confined during our experiment to the $l=0$ plane.

In Fig. 1 we show field dependence of the integrated intensity of three representative reflections measured up to 14.5 T. For comparison, magnetization curve measured at 4.2 K with field applied along the c axis is shown at the bottom marking two metamagnetic-like transitions at 2.5 and 9.5 T. As can be seen, both critical fields are clearly visible on (200) and (210) reflections while on (120) reflection only the latter transition can be discerned. The increase in intensity at 2.5 T is much smaller than at 9.5 T for (200) and (210) reflections. This is in agreement with proposed uncompensated antiferromagnetic structure with $\boldsymbol{q}=(0,1/3,1/3)$ which exhibits a ferromagnetic component between the two critical fields (see bottom of Fig. 1). The ferromagnetic component amounts to one third of the magnetization attained in fields above 9.5 T applied along the c axis. Note that between the two metamagnetic-like transitions we were, in analogy to the zero-field situation, unable to observe any magnetic reflections.

Up to now, a simple collinear field-forced ferromagnetic structure was supposed to be formed in UNiGe above the second critical field (for the c axis direction above 9.5 T) although we have speculated previously about the presence of the antiferromagnetic a axis component at high fields as well.[17] However, due to necessity of rather high magnetic fields a clear experimental proof was up to now missing.

On the basis of the bulk magnetic studies, a ferromagnetic (canted or collinear) or a ferrimagnetic structure are possible in UNiGe at 2 K in high fields. Reciprocal scans at high fields suggest that propagation vector $\boldsymbol{q}=(0,0,0)$ is established under these conditions. By subtracting the integrated intensity measured in zero field at 2 K from the integrated intensity obtained at 12 T, additional magnetic intensity has been unambiguously found on top of 26 reflections belonging to eight inequivalent groups (020), (120), (200), (210), (220), (320), (400), and (410).

In order to fit our data to all possible magnetic structures that are compatible with paramagnetic *Pnma* space group and with the magnetic-structure propagation vector $\boldsymbol{q}=(0,0,0)$, we have used irreducible-representation (IR) theory.[18] Group analysis for the Wyckoff position 4(c), which is occupied by U atoms that are the only magnetic species in

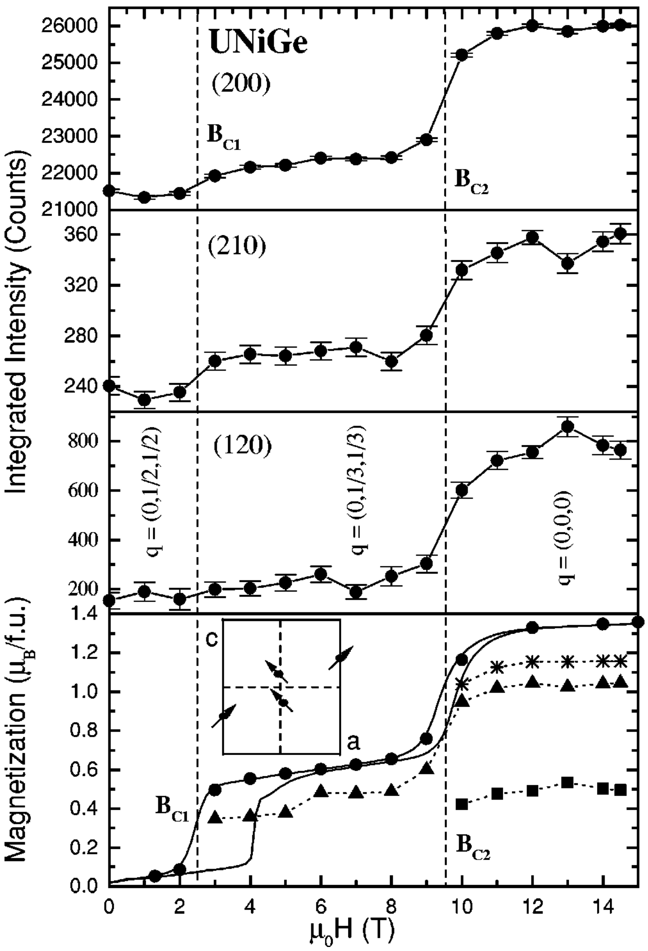

FIG. 1. Field dependence of integrated intensity of the (200), (210), and (120) reflection. For comparison, field dependence of the magnetization measured along the *c* axis (d) during continuous field sweeps (line) and quasistatic fields (●) is also shown. In the bottom panel, ferromagnetic *c* axis component (▲), the antiferromagnetic *a* axis component (■), and the total U magnetic moment (*) obtained from our neutron diffraction experiment are depicted. The model of the refined magnetic structure is shown as well. For details see the main text.

UNiGe leads to eight one-dimensional IR's. The basis vectors (possible magnetic structures) associated with various IR's can be divided in two subgroups. In four models only U moments along the *b* axis are allowed. In the remaining models are U moments confined to the *a*-*c* planes, i.e., perpendicular to the *b* axis. Among all the models, there is only one magnetic structure that allows for ferromagnetic coupling of U moments along the *c* axis. This magnetic structure allows both the *a* and *c* axis components. The question now is if they both are realized. Let us note that we suppose equal U magnetic moments in all magnetic structure models (discarding thus ferrimagnetic structures) what is fairly reasonable assumption because all U atoms occupy the same crystallographic site and there is no sign of frustration in the system, i.e., no competing interactions exist in UNiGe along the same crystallographic directions.

It can be shown that only the magnetic structure with both the *a* and *c* axis components nonzero is compatible with observed magnetic reflections. Moreover, the intensity of the (120) and (320) magnetic reflections is directly proportional only to the *a* axis component of U magnetic moments and not to the *c* axis component. The fact that we do observe magnetic intensity on top of these reflections suggests that the forced-ferromagnetic structure of UNiGe in high magnetic fields above 9.5 T applied along the *c* axis is noncollinear and has a significant antiferromagnetic *a* axis component. Refinement leads to the antiferromagnetic *a* axis component $\mu_{\mathrm{AF}}=0.51\pm0.05\mu_B$ and to the ferromagnetic *c* axis component $\mu_F=1.04\pm0.04\mu_B$. The total U magnetic moment is $\mu=1.16\pm0.04\mu_B$. The projection onto the *b*-*c* plane of the resulting magnetic structure is depicted in Fig. 1.

Thanks to the fact that intensity of the (200) and (210) reflections contain information concerning the ferromagnetic *c* axis component and the (120) about the antiferromagnetic *a* axis component we were able to deduce these components at each magnetic field. Resulting field dependencies are depicted at the bottom of Fig. 1. Of course, between 2.5 and 9.5 T only the *c* axis component can be refined. From the μ_{AF} and μ_F values above 9.5 T we were able to calculate the tilting angle which make U moments with respect to the *c* axis. This angle amounts to 25±1 degrees and compares well with the out-off *b*-*c* plane tilting of 20° found for the ground-state antiferromagnetic structure in UNiGe (Ref. 6) and with 17 degrees found in the incommensurate phase of UNiGe.[12]

V. DISCUSSION AND CONCLUSIONS

We have investigated the field-forced ferromagnetic structure in UNiGe by means of a microscopic method, namely, using neutron diffraction in fields up to 14.5 T, to be able to compare these results with previous bulk magnetization measurements. Clearly, the size of U magnetic moments determined from our diffraction experiment is 15% smaller with respect to the magnetization data. There are several possible scenarios to explain the disagreement. The first deals with the fact that during the course of refinement of the integrated intensities we had to suppose a certain magnetic form factor. We have chosen for calculation the U^{3+} magnetic form factor calculated in the dipole approximation. Clearly, due to lack of decisive information on the U valence state in UNiGe, other magnetic form factors are possible. However, it should be noted that magnetic form factors for various uranium valence states are quite similar and therefore, the influence on the refined size of moments is small, of the order of 5%. The other possibility is that during the magnetization measurement one records the magnetic response of all three species, i.e., including Ni and Ge. While Ge does not carry any magnetic moment, small induced magnetic moment can be expected on Ni atoms. Similarly induced moment of the order of $0.1\mu_{\mathrm{B}}$/transition metal were detected using a polarized-neutron scattering experiments in URhAl,[19] UNiAl,[20] and UNiGa.[20] *Ab initio* electron-structure calculations using a fully relativistic optimized linear combination of atomic orbitals method[1,21] supports the idea that a significant hybridization between U $5f$ states and Ni $3d$ states leads to almost complete filling of the $3d$ band on Ni and to small induced moments on these atoms. Yet another possibility to explain the 15% smaller U magnetic moments is change of extinction or multiple scattering across the metamagnetic-like phase transition. Indeed, the magnetic state of UNiGe at high magnetic field should be monodomain with higher perfectness of the lattice leading to stronger extinction. Magnetostriction-driven change of lattice constants opens a possibility for modified multiple scattering. Both latter processes are, however, hard to evaluate.

Another consequence of the hybridization together with a significant orbital part and a strong spin-orbit coupling existing for $5f$ states is that these materials (containing $5f$ electrons and d electron states) exhibit an enormous anisotropy. Hybridization enhanced anisotropy of the type depending on the geometrical surrounding of the $5f$-electron atom is present in a vast majority of uranium-based compounds and its strength is larger than in $4f$ compounds.[22] Hybridization generally leads to delocalization of electronic states responsible for magnetic properties and in the strong limit to loss of magnetic ordering. However, in the weak-hybridization limit it promotes the magnetic ordering because it acts as an effective interaction mechanism between $5f$ electron states residing predominantly at actinide ions. Especially, $5f$-d hybridization acts in this way.

At this point it would be interesting to compare the field-induced magnetic structure of UNige with the ground-state magnetic structures in URhSi (Refs. 23,24) and URhGe (Refs. 23,25) that have the same crystal structure and are ferromagnets. In the case of URhSi the magnetic structure has been identified as collinear with strongly reduced magnetic moments parallel to the c axis. On the contrary for URhGe Tran *et al.*[23] published a non-collinear magnetic structure with reduced moments confined to the a-c plane that is of the same type as the here presented field-induced magnetic structure of UNiGe. This suggests that both structures might be intimately connected due to a microscopic coupling mechanism, for instance, due to hybridization between $5f$ and d states. On the other hand, Aoki *et al.*[25] reported for URhGe a collinear ferromagnetic structure that contradicts Tran's work.

The most striking feature of our observation is the robust nature of μ_x, the antiferromagnetic x component of the magnetic moments of uranium atoms. The presence of this component in the antiferromagnetic phase was identified[17] as direct evidence for anisotropic interactions. The observation that the phenomenon prevails in the forced ferromagnetic phase suggests that the anisotropic interaction is at least of the same order of magnitude as the isotropic one.

The argument in favor of an anisotropic antiferromagnetic exchange interaction goes as follows. The low value of χ_{xx}, the a-axis susceptibility, and the absence of any sign of a metamagnetic transition for $B_x \leqslant 38$ T show that crystal fields suppress magnetization in the x direction with an anisotropy field $B_x^{(a)} > 40$ T. Evidently, the antiferromagnetic interaction overcomes this anisotropy field. However, an isotropic interaction of the form $J(\hat{\mathbf{s}}_1 \cdot \hat{\mathbf{s}}_2)$, $J>0$, would generate ordered moments only in the b-c plane, so as to avoid the positive anisotropy energy associated with μ_x.

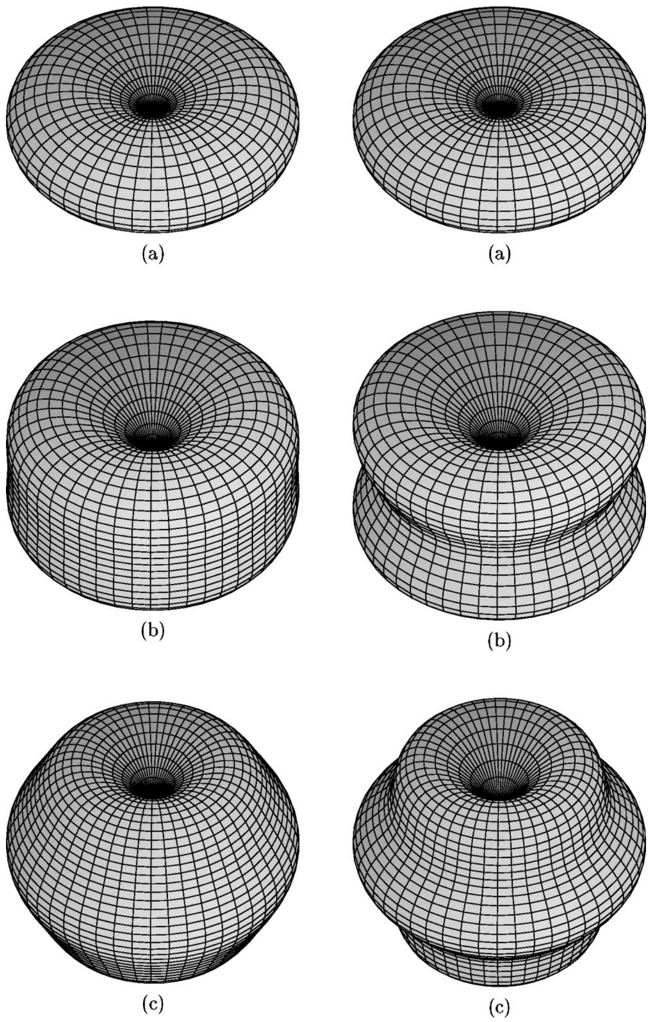

FIG. 2. Charge densities (normalized to one electron; left panel) and spin densities (angular distribution of the magnitude of the spin vector; right panel) for the Hund's-rule ground state of the $f^1(a)$, $f^2(b)$, and f^3 (c) configurations.

Given the large spin-orbit coupling and the possibility of a large orbital angular momentum of uranium, one can speculate about numerous mechanisms of anisotropic exchange. As the uranium atoms are embedded in the lattice of a ternary compound, an indirect mechanism is likely, either through ligand states (superexchange)[26] or through Bloch states [Ruderman-Kittel-Kasuya-Yosida (RKKY)].[27,28] When considering such indirect interactions between two atoms, it is not the medium that introduces the anisotropy. The anisotropy of the interaction is due to the nonspherical charge distribution of the f electrons and due to the spin-orbit coupling, which introduces a nonspherical and noncollinear spin distribution. In the theory of the anisotropic RKKY interaction, these anisotropies are mediated by a fully isotropic jellium, since the f states couple to spherical waves of the same angular pattern.

Figure 2 shows such patterns for the Hund's-rule ground states of the f^1, f^2, and f^3 configurations. For a single f electron [Fig. 2(a)] we recognize the familiar torus-shaped probability density, in accordance with the classical picture

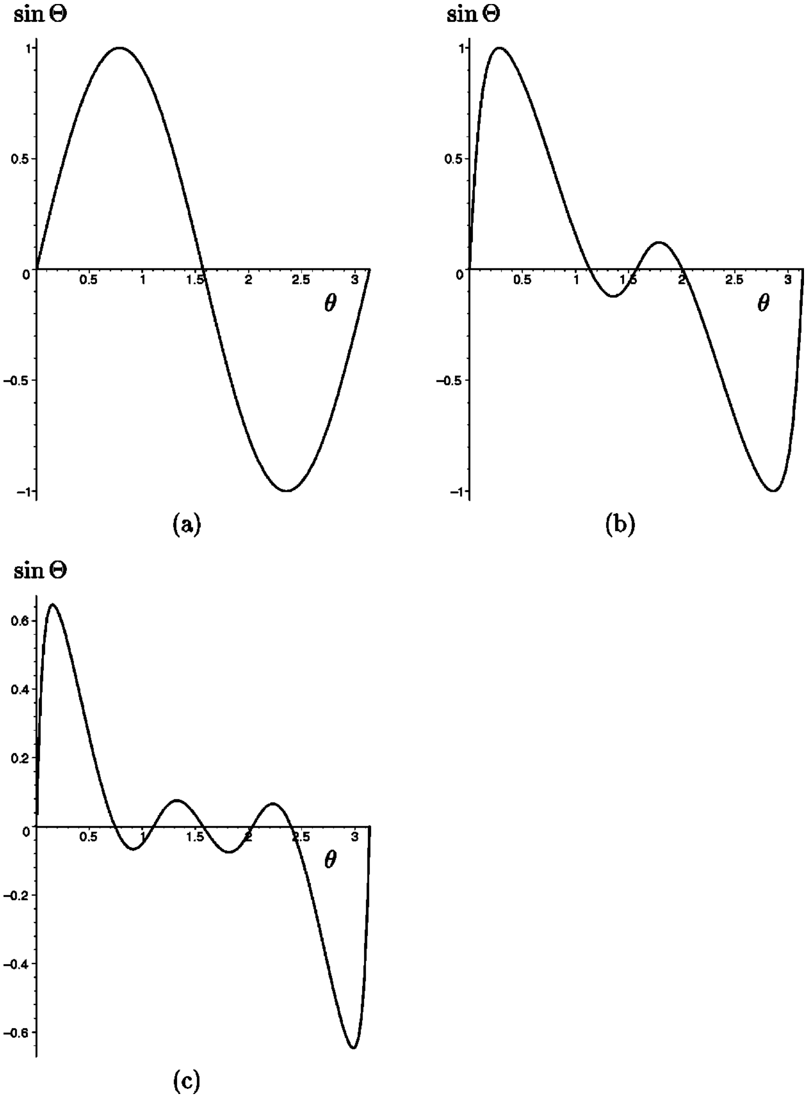

FIG. 3. Noncollinearity of the spin distribution in the f shell for the Hund's-rule ground state of the $f^1(a)$, $f^2(b)$, and $f^3(c)$ configurations represented as the perpendicular component of the spin density, in units of its magnitude, as a function of the colatidudal angle θ, $\sin\Theta = S_x(\theta)/|\mathbf{S}(\theta)|$.

of an electron orbiting the nucleus. The spin density is seen to have the same shape, as it should, since for a single electron ($s=1/2$), $|\langle \mathrm{s}\rangle|=1/2$, constant. On the other hand, Fig. 3(a) shows that the spin density is strongly noncollinear, though it points downwards, as expected for $J=L-S$, in the region where the probability density is substantial. The f^1-f^2-f^3 sequence shows a trend towards a spherically symmetric and collinear spin distribution with increasing numbers of f electrons. (Ultimately, at f^7 this high symmetry is reached with the fully spin-polarized orbital s states of gadolinium.) This tendency suggests that Russell-Saunders coupling will reduce anisotropic interactions. In the case of UNiGe, the high-temperature susceptibility and high-field magnetization show signs of the breakdown of this coupling. In particular, the effective moment is about $3\mu_B$ (slightly anisotropic), as opposed to $3.6\mu_B$, which is the value expected for the f^2 and f^3 configurations, and in the forced ferromagnetic state the magnetization remains below half of its expected saturation value appropriate to the Hund's rule ground state.

Staunton *et al.*[27] have derived the orientation dependence of the interaction energy mediated by relativistic scattering between two magnetic "impurities." They found that the isotropic RKKY interaction has just one of a large number of possible orientation dependencies. In general, the interaction energy contains terms of the form

$$J^{(i,j)}(\hat{\mathbf{s}}_1\cdot\hat{\mathbf{s}}_2)[(\mathbf{R}_{12}\cdot\hat{\mathbf{s}}_1)(\mathbf{R}_{21}\cdot\hat{\mathbf{s}}_2)]^i[\mathbf{R}_{12}\cdot(\hat{\mathbf{s}}_1\times\hat{\mathbf{s}}_2)]^{2j}, \qquad (1)$$

where $\hat{\mathbf{s}}_1$ and $\hat{\mathbf{s}}_2$ are unit vectors pointing in the direction of an effective local magnetic field at sites 1 and 2, respectively, and $\mathbf{R}_{21}=\mathbf{R}_2-\mathbf{R}_1$ is the vector connecting the two sites. The exponents i and j run from 0 to $2l_{\max}$, where $l_{\max}$ is the largest angular momentum quantum number contributing to the scattering amplitude; for lanthanides and actinides $l_{\max}=3$. The dependence of the interaction energy on the distance R_{12} is of the usual $\cos(2k_FR_{12})/R_{12}^3$ form so that for $i=j=0$ the isotropic RKKY interaction emerges. The higher-order terms contain pseudodipolar contributions (when $i\neq 0$) and even powers of the Dzialoshinsky-Moriya interaction[26,29] ($j\neq 0$). Odd powers can only occur if inversion symmetry with respect to the midpoint between spins 1 and 2 is broken, which is obviously not the case in this model.

In UNiGe, there is no inversion symmetry with respect to points half way between two uranium atoms in the buckled chain along the *a* axis. However, the Dzialoshinsky-Moriya interaction favors a perpendicular arrangement of $\hat{\mathbf{s}}_1$ and $\hat{\mathbf{s}}_2$ in the plane perpendicular to the vector $\mathbf{R}_{21}$. Confinement to this plane is dictated by the dot product in $\mathbf{R}_{21}\cdot(\hat{\mathbf{s}}_1\times\hat{\mathbf{s}}_2)$, to which the x component of $\hat{\mathbf{s}}_1$ or $\hat{\mathbf{s}}_2$ hardly contributes, if one considers interaction between uranium atoms in the buckled *a*-axis chain. Therefore, the Dzialoshinsky-Moriya interaction can not account for the spontaneous generation of a μ_x component. The U-U separation being shortest along this chain, it is reasonable to speculate about interactions of another symmetry between these atoms, rather than about interactions between more distant neighbors. Indeed, a combination of the isotropic and the second-order pseudodipolar forms $[i=2$ and $j=0$ in Eq. $(1)]$ in the in-chain interaction will insist on a component of $\hat{\mathbf{s}}_{1,2}$ along $\mathbf{R}_{21}$, favoring antiferromagnetically oriented μ_x components, if $J^{(2,0)}>0$. In fact, higher order terms cannot be excluded and they will dictate the same preferred orientations if i is even and $J^{(i,0)}>0$. The odd-power pseudodipolar interaction is indifferent to the relative orientation of $\hat{\mathbf{s}}_1$ and $\hat{\mathbf{s}}_2$, since, if i is odd, both $(\hat{\mathbf{s}}_1\cdot\hat{\mathbf{s}}_2)$ and $[(\mathbf{R}_{12}\cdot\hat{s}_1)(\mathbf{R}_{21}\cdot\hat{\mathbf{s}}_2)]^i$ change sign when the $\hat{\mathbf{s}}$ vectors are flipped from parallel to anti-parallel orientation. However, combined with a strong isotropic $(i=0)$ exchange interaction, for $J^{(i,0)}>0$ such interaction can also overcome the anisotropy field and generate μ_x components.

ACKNOWLEDGMENTS

K.A. gratefully acknowledges the support of Kordofan University, the University of Amsterdam and the ‘‘Steufonds Soedanese Studenten,’’ Leiden, The Netherlands, which enabled him to participate in this work. H.N. acknowledges support through the NSF (Grant No. DMR-0094241).

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[1] V. Sechovský and L. Havela, in *Handbook of Magnetic Materials*, edited by K. H. J. Buschow (North-Holland, Amsterdam, 1998), Vol. 11, p. 93.

[2] T. Gasche, M. S. S. Brooks, and B. Johansson, J. Phys.: Condens. Matter **7**, 9499 (1995); T. Gasche, M. S. S. Brooks, and B. Johansson, *ibid.* **7**, 9511 (1995).

[3] F. Canepa, P. Manfrinetti, M. Pani, and P. Palenzona, J. Alloys Compd. **234**, 225 (1996).

[4] K. Prokeš, H. Nakotte, E. Brück, F. R. de Boer, L. Havela, V. Sechovský, P. Svoboda, and H. Maletta, IEEE Trans. Magn. **30**, 1214 (1994).

[5] S. Kawamata, K. Ishimoto, Y. Yamaguchi, and T. Komatsubara, J. Magn. Magn. Mater. **104**, 51 (1992).

[6] A. Purwanto, V. Sechovský, L. Havela, R. A. Robinson, H. Nakotte, A. C. Larson, K. Prokeš, E. Brück, and F. R. de Boer, Phys. Rev. B **53**, 758 (1996).

[7] L. Havela, V. Sechovský, F. R. de Boer, E. Brück, and H. Nakotte, Physica B **177**, 159 (1992).

[8] V. Sechovský, L. Havela, A. Purwanto, A. C. Larson, R. A. Robinson, K. Prokeš, H. Nakotte, E. Brück, F. R. de Boer, P. Svoboda, H. Maletta, and M. Winkelmann, J. Alloys Compd. **293-294**, 536 (1994).

[9] V. Sechovský, L. Havela, P. Svoboda, A. Purwanto, A. C. Larson, R. A. Robinson, K. Prokeš, H. Nakotte, F. R. de Boer, and H. Maletta, J. Appl. Phys. **76**, 6217 (1994).

[10] R. Troć and V. H. Tran, J. Magn. Magn. Mater. **73**, 389 (1988).

[11] A. Murasik, P. Fischer, R. Troć, and V. H. Tran, J. Phys.: Condens. Matter **3**, 1841 (1991).

[12] H. Nakotte, A. Purwanto, R. A. Robinson, Z. Tun, K. Prokeš, A. C. Larson, L. Havela, V. Sechovský, H. Maletta, E. Brück, and F. R. de Boer, Phys. Rev. B **54**, 7201 (1996).

[13] M. S. Lehman and F. K. Larsen, Acta Crystallogr., Sect. A: Cryst. Phys., Diffr., Theor. Gen. Crystallogr. **31**, 245 (1974).

[14] J. Rodrigues-Carvajal, FULLPROF version 3.2 JAN97 (unpublished).

[15] V. F. Sears, Neutron News **3**, 26 (1992).

[16] A. J. Freeman, J. P. Desclaux, G. H. Lander, and J. Faber, Phys. Rev. B **13**, 1168 (1976).

[17] P. F. de Châtel, K. Prokeš, H. Nakotte, A. Purwanto, V. Sechovský, L. Havela, E. Brück, R. A. Robinson, and F. R. de Boer, J. Magn. Magn. Mater. **177-181**, 785 (1998).

[18] E. F. Bertaut, Acta Crystallogr., Sect. A: Cryst. Phys., Diffr., Theor. Gen. Crystallogr. **24**, 217 (1968).

[19] J. A. Paixão, G. H. Lander, P. J. Brown, H. Nakotte, F. R. de Boer, and E. Brück, J. Phys.: Condens. Matter **4**, 829 (1992).

[20] M. Olšovec, J. Schweitzer, L. Paolasini, V. Sechovský, and K. Prokeš, Physica B **241-243**, 678 (1998).

[21] M. Diviš, M. Richter, and M. Olšovec (unpublished).

[22] G. H. Lander *et al.*, J. Appl. Phys. **69**, 4803 (1991).

[23] V. H. Tran, R. Troć, and G. André, J. Magn. Magn. Mater. **186**, 81 (1998).

[24] K. Prokeš, M. Diviš, V. Sechovský, P. Svoboda, M. Richter, K. H. J. Buschow, and F. R. de Boer, Solid State Commun. **104**, 597 (1997).

[25] D. Aoki *et al.*, Nature (London) **413**, 613 (2001).

[26] T. Moriya, Phys. Rev. Lett. **4**, 228 (1960).

[27] J. B. Staunton, B. L. Györffy, J. Poultner, and P. Strange, J. Phys. C **21**, 1595 (1988).

[28] H. Shiba, O. Sakai, and R. Shiina, J. Phys. Soc. Jpn. **68**, 1988 (1999).

[29] I. Dzialoshinsky, J. Phys. Chem. Solids **4**, 241 (1958).