Rapid Communications

Microscopic nature of drastic influence of hydrogen on the magnetic anisotropy of 5*f*-electron systems: The case of U_2Ni_2Sn

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Recent experiments showed that hydrogenation of U_2Ni_2Sn leads to a dramatic change of the magnetic anisotropy (MA) from strongly uniaxial type to easy-plane type with easy axis and easy plane orthogonal to each other. We applied first-principles calculations aiming to understand the microscopic origin of the drastic MA change and distinguish between discontinuous and continuous scenarios of the transformation. The calculations combined with symmetry analysis revealed that the hydrogenation leads to the instability of both uniaxial and easy-plane states caused by the reduced symmetry of the atomic lattice. The obtained noncollinear noncoplanar magnetic states have the features of both apparently competing magnetic structures, which indicates the validity of the continuous scenario of the transformation. An insight into the active interatomic interactions shows that Dzyaloshinskii-Moriya interaction contributes to magnetic transformations and must be taken into account on the same footing as MA.

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Because of the chemical properties and small size of the H atoms they can easily penetrate into the bulk of materials, occupying interstitial positions, which leads to modification of properties. The character of the modification varies widely for different systems and has been intensively studied (see, e.g., Refs. [1–21]). In particular, the influence of the hydrogenation on magnetic properties has been the topic of many publications [1,4,6,7,10–12,17,19,20]. The potential to modify the properties of the absorbing systems provides an avenue for design of materials with new functionalities. Closely related to this applied aspect, the modification of the properties of the materials raises new fundamental questions about the physics of the underlying processes. In 5*f* materials, H provides an interesting tool for strengthening of magnetism, related to a large extent to a volume expansion increasing interactinide spacing. This explains, e.g., the ferromagnetism of U hydrides and increase of ordering temperature in U ternaries [22,23].

In this Rapid Communication we focus on the U_2Ni_2Sn (UNS) compound where recent experiments revealed a dramatic change of magnetic anisotropy (MA) under the influence of hydrogen. In the H-free form, UNS possesses very strong uniaxial magnetic anisotropy with easy axis parallel to the *z* axis (Fig. 1). The experiment on a single crystal of UNS gave an estimation of the MA amounting to 1.07 mRy per U atom [25]. In sharp contrast, the hydrogenated system was reported to show the *xy* plane as the easy plane [26]. This striking change of the MA raises an important question about the physical mechanism, by which the presence of hydrogen changes so strongly the magnetic properties of the system.

At least two different scenarios are possible. The first possibility is a discontinuous transformation. In this case both easy-axis and easy-plane states correspond to local energy minima. If for H-free UNS the easy-axis minimum is deeper and the hydrogenation makes the easy-plane minimum deeper, a discontinuous transformation from one state to the other takes place. In the second possible scenario, intermediate states of the system are available to allow a continuous transformation.

Since the experiment could be performed either for the H-free system [25] or the system with high hydrogen concentration of about one H atom per one U atom [26], it is difficult to reveal experimentally how the process of the transformation from one physical state to the other takes place. The purpose of this Rapid Communication is to get an insight into the processes taking place under the influence of H on the basis of microscopic theoretical treatment. Our main tool will be the combination of the first-principles calculations and symmetry analysis.

The calculations are performed with the augmented spherical waves method [27,28] generalized to deal with noncollinear magnetism and spin-orbit coupling [29]. The generalized gradient approximation (GGA) to the energy functional [30] is employed in the calculations. To examine the influence of the on-site correlation of the 5*f* electrons on the calculated quantities, we applied the GGA + *U* method [31].

First, we performed calculations for the H-free system with lattice parameters reported in [25]. The calculations were carried out for the following magnetic configurations:

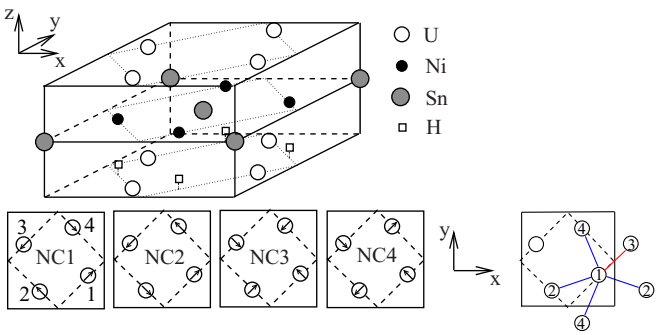

FIG. 1. Unit cell of U_2Ni_2Sn. The positions of the H atoms are schematically shown as well as the four noncollinear magnetic structures suggested on the basis of group theory analysis [24]. In the figure of the NC1 structure the numbering of the U sublattices is given. The small figure at the bottom right shows the near neighbors of atom 1 in the adjacent unit cells.

ferromagnetic states FM_z and FM_x, where the subscript shows the direction of the magnetic moments, and four noncollinear magnetic states depicted in Fig. 1. The precise form of the magnetic structure of UNS-H is not known. The experimental information [26] is restricted to the statement of an in-plane antiferromagnetic configuration. In U_2Pd_2In and U_2Pd_2Sn compounds with the same crystal structure and related properties of the magnetic state, the four noncollinear configurations shown in Fig. 1 were considered as candidates for the ground state magnetic structure with the NC1 structure treated as the most probable one [24,32].

The energies of the magnetic states counted from the energies of corresponding FM_z states are collected in column I of Table I. The out-of-plane FM_z configuration is distinctly lower in energy than the in-plane configurations that confirm the experimental finding of the strong uniaxial MA in the system. Among the in-plane configurations the lowest in energy is FM_x. The energy of the noncollinear states increases in the sequence NC1,NC2,NC3,NC4 similar to the results obtained for other representatives of the uranium 2-2-1 compounds [33,34].

The calculated U moment of the FM_z structure $0.72\mu_B$ is obtained as the vector sum of spin moment $1.62\mu_B$ and oppositely directed orbital moment $2.34\mu_B$. The value of the atomic moment is in good correlation with the experimental estimate of $0.87\mu_B$ [25]. The application of the GGA + U method leads, in general, to increased calculated atomic moments. In the case of UNS the value of $U = 0.05$ Ry already results in the atomic moments of $1.35\mu_B$ considerably exceeding the experimental value. On the other hand, the smaller values of parameter U preserving good agreement of the calculated and experimental atomic moments do not lead to important changes in the picture developed in the Rapid Communication. Therefore we present the results obtained with the GGA exchange-correlation potential [35].

TABLE I. Energies of the magnetic structures in mRy per U atom. The energies are counted from the energies of corresponding FM_z states. Column I corresponds to UNS with the lattice parameters of the H-free crystal; column II corresponds to UNS with the lattice parameters of UNS-H; column III corresponds to UNS-H with the positions of H atoms in the U planes.

	I	II	III
FM_z	0.00	0.00	0.00
FM_x	1.19	1.44	0.73
NC1	1.46	1.36	0.58
NC2	2.17	2.06	1.37
NC3	2.31	2.60	1.89
NC4	2.90	3.12	2.39

The study of the influence of the hydrogenation begins with an examination of the assumption that H influences MA solely through changing the distances between U atoms. The calculations were performed for UNS with the lattice constants and atomic positions reported in Ref. [26] for the hydrogenated system. At this stage, the presence of H was not explicitly taken into account. The calculated energies are collected in column II of Table I. The easy-axis character of the MA is preserved and the calculated change of the strength of the MA is relatively small. The lowest-energy in-plane structure is still FM_x, although the energies of FM_x and NC1 are now very close. We conclude that the anisotropy change cannot be ascribed to the change of the geometry of the U sublattice only.

In the next step, the presence of the H atoms is explicitly included into consideration and the calculations are performed for $U_2Ni_2SnH_2$ (UNS-H). The experiment [26] suggests that the positions of the H atoms deviate by 0.127 Å from the planes of the U atoms. To achieve deeper insight into the role of different factors we first performed calculations with the H atoms occupying the positions in the U plane. The explicit inclusion of the H atoms in the calculation changes quantitatively the relation between energies of the magnetic states (see column III of Table I). The lowest-energy in-plane structure is now NC1 and the MA is considerably decreased. However, also in this case the z axis remains the easy axis and the process of the change of the magnetic anisotropy remains undisclosed.

A qualitatively new situation was obtained after taking into account the experimentally observed shift of the H atoms from the plane of the U atoms. Very importantly, the calculations with the shifted H atoms showed that for this atomic coordination neither FM_z nor NC1 structure remains stable: for both structures already after the first iteration the U atomic moments deviate from their assumed directions manifesting the instability of the FM and NC1 magnetic configurations. The analysis of the symmetry of the magnetic crystals reveals the origin of the instability caused by the shift of the H atoms.

A general principle governing the relation between symmetry and stability of the magnetic structures was discussed in Refs. [29,33,36]. Let us assume that magnetic structure of the system possesses a specific regular feature. In the cases important for us, this regular feature is the collinearity of all atomic moments to either the z axis for the FM_z structure or to the xy plane for the NC1 structure. This assumed regularity in the system can be stable only if there is a symmetry operation that is responsible for this regularity. This means that for the stable regular feature in the magnetic structure there exists a symmetry operation that is destroyed if the moments deviate from the directions defined by the regular structure.

TABLE II. Symmetry operations $\{\alpha|\boldsymbol{\tau}_\alpha\}$ of the atomic lattice of $U_2Ni_2SnH_2$. First column: point operation α: E is the unity operation; C_{nz} are proper rotations by angle $2\pi/n$ about the z axis; σ_β is the reflexion in the β plane, $\beta = x, y$ correspond to the $x = 0$, $y = 0$ planes, $\beta = a$ corresponds to the $y = -x$ plane, and $\beta = b$ corresponds to the $y = x$ plane. Second column: if nonzero, gives the vector of nonprimitive translation $\boldsymbol{\tau} = (0.5, 0.5, 0)$. Third column gives the number of atom 1_α in which atom 1 is transformed by the given symmetry operation. + in the fourth and sixth columns shows, respectively for structures FM_z and NC1, that the space operation is combined with the time inversion. Fifth and seventh columns: the result of the action of the operation corresponding to, respectively, FM_z and NC1 structures on axial vector (m_x, m_y, m_z). $\overline{m}$ means $-m$.

			FM_z		NC1	
α	$\boldsymbol{\tau}_\alpha$	1_α	T	**m**	T	**m**
E	0	1	−	(m_x, m_y, m_z)	−	(m_x, m_y, m_z)
σ_b	$\boldsymbol{\tau}$	1	+	(m_y, m_x, m_z)	+	(m_y, m_x, m_z)
C_{4z}^-	0	2	−	$(m_y, \overline{m}_x, m_z)$	+	$(\overline{m}_y, m_x, \overline{m}_z)$
σ_y	$\boldsymbol{\tau}$	2	+	$(m_x, \overline{m}_y, m_z)$	−	$(\overline{m}_x, m_y, \overline{m}_z)$
C_{2z}	0	3	−	$(\overline{m}_x, \overline{m}_y, m_z)$	−	$(\overline{m}_x, \overline{m}_y, m_z)$
σ_a	$\boldsymbol{\tau}$	3	+	$(\overline{m}_y, \overline{m}_x, m_z)$	+	$(\overline{m}_y, \overline{m}_x, m_z)$
C_{4z}^+	0	4	−	$(\overline{m}_y, m_x, m_z)$	+	$(m_y, \overline{m}_x, \overline{m}_z)$
σ_x	$\boldsymbol{\tau}$	4	+	$(\overline{m}_x, m_y, m_z)$	−	$(m_x, \overline{m}_y, \overline{m}_z)$

The atomic lattice of UNS-H with shifted H atoms is characterized by the space group with eight point operations collected in Table II. In the third column we show the atoms 1_α in which atom 1 is transformed by corresponding symmetry operation. Since the U atoms are equivalent, the information involving atom 1 is sufficient to restore the whole picture. The symmetry operations of a magnetic structure must leave invariant not only the atomic lattice but also the directions of the atomic moments. To obtain adequate description of the symmetry of the magnetic structure it is very important to include into consideration the operation of time inversion. Both FM_z and NC1 magnetic structures are invariant with respect to eight operations. The space parts of the operations are identical to each other and coincide with the symmetry operations of the atomic lattice. However, half of the operations must be combined with time inversion to be the symmetry operations of the magnetic structures. The combination of the space transformations with time inversion is different for the two magnetic structures. This results in different constraints imposed by the symmetry on the magnetic moments. Columns 4 and 6 of Table II show which of the space transformations are combined with time inversion in the cases of FM_z and NC1 structures, respectively. In columns 5 and 7 we show how the symmetry operation transforms the atomic moment of atom 1.

We remark that the symmetry constraint imposed on the atomic moment of atom 1 is, for both magnetic structures, $m_x = m_y$.

Let us consider the symmetry constraint on the atomic moments of the FM_z structure. Importantly, the symmetry operations do not request that the m_x and m_y projections of the atomic moments are zero. If all four U moments deviate from the z axis by the same angle θ and the projections of the moments on the xy plane form the structure of the NC2 type (Fig. 1), all symmetry operations remain intact. This means that the collinear FM_z structure is not distinguished by symmetry with respect to this type of noncollinear structures. Since there are no symmetry operations responsible for the collinearity of the moments to the z axis and the FM_z state appears to be just one of the continuums of the states that are equivalent from the symmetry point of view, the probability that the energy minimum corresponds to the collinear state with $\theta = 0$ is negligible and the deviations of the atomic moments from the z axis must take place. This is exactly what our calculations give: the U atomic moments deviate from the z axis by the same angle θ and the xy projections form the NC2 configuration [Fig. 2(a)]. The self-consistent state is given by the magnetic structure with $\theta = 36.6^\circ$.

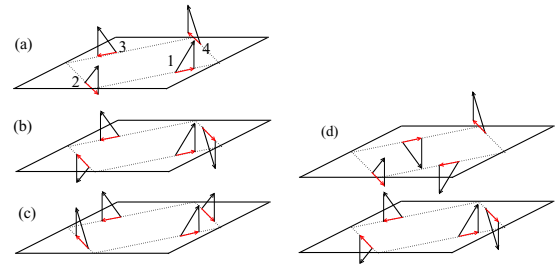

FIG. 2. Schematic presentation of noncollinear noncoplanar magnetic structures of hydrogenated UNS. (a) The z projections of all atomic moments are positive; the projections on the xy plane are of the NC2 type. (b) The z projections of the moments of atoms 1 and 3 and atoms 2 and 4 have opposite signs; the projections on the xy plane are of the NC1 type. (c) The z projections of all atomic moments are positive; the projections on the xy plane are of the NC1 type. (d) The structure obtained starting with the AFM-GS state containing eight U atoms per magnetic unit cell.

In the case of the planar NC1 structure the symmetry operations and corresponding transformations of the atoms and atomic moments do not request that the m_z projections of the U moments are zero. The $m_z = 0$ is, however, the characteristic feature of the coplanar structure. Indeed our calculations result in the deviation of the moments from the initial in-plane directions: the moments keep the NC1-type projection on the xy plane but assume nonzero z components that are equal to one another in the absolute value and opposite in sign for atoms 1 and 3 and atoms 2 and 4 as predicted by the symmetry analysis [Fig. 2(b)]. Here the deviation angle is relatively small and takes the value of 9.5°.

An additional important observation we obtained by partly releasing the symmetry constraint on the relaxation of the NC1 structure. We introduced very small vertical moment on the Sn atoms. This weak "noise" decreased the number of constraining symmetry operations from 8 to 4. The remaining operations are $E, C_{2z}, \sigma_b, \sigma_a$. This symmetry breaking was sufficient to strongly change the behavior of the system. We again began iterations with the coplanar NC1 structure. At first, the change in the magnetic structure during iterations followed closely the behavior obtained under the full symmetry constraint for the NC1 structure. Then, however, the behavior changed strongly and the resulting self-consistent magnetic structure had the angles $\theta = 57^\circ$ for all four U atoms and the xy projection of the NC1 type [Fig. 2(c)].

We also obtained the same type of behavior for the antiferromagnetic ground state AFM-GS of H-free UNS [25]. The atomic moments of AFM-GS are collinear to the z axis. The structure is characterized by doubling of the magnetic unit cell along the z axis. The hydrogenation lowers the symmetry of the magnetic state and leads to its instability. The self-consistent noncollinear configuration obtained by relaxation of AFM-GS is depicted in Fig. 2(d). The U atomic moments deviate from the z axis by angle 46° and the projections of the moments on the xy plane are of the NC1 type. The energy of the relaxed AFM-GS structure is the lowest. The energies of the structures presented in Figs. 2(a), 2(b), and 2(c) are higher by 0.20, 0.45, and 0.14 mRy, respectively.

We emphasize that the instability of the collinear and coplanar structures is the consequence of the shift of the H atoms from the U plane. Indeed, the atomic lattices of both UNS and UNS-H with the H atoms in the U planes have 16 symmetry operations instead of 8 operations collected in Table II. These additional operations can be obtained by combining the operations from Table II with space inversion. Also the number of symmetry operations of the magnetic structures doubles. It is crucial that these additional symmetry operations request that $m_x = m_y = 0$ in the case of magnetic states collinear to the z axis and $m_z = 0$ in the case of planar state. Therefore these symmetry operations prevent the deviations of the atomic moments obtained for the UNS-H with shifted H atoms. The magnetic states remain stable, which is confirmed by our calculations.

Our findings show that the apparent change of the magnetic anisotropy takes place not in the discontinuous scenario of the competition between the energies of two magnetic configurations, one collinear to the z axis and the other parallel to the xy plane. Instead, both types of configurations become unstable after adding H because of the decreased symmetry of the atomic lattice. The self-consistent configurations are the canted states that combine features of both limiting magnetic states. This reveals a continuous scenario of changing the magnetic state under the influence of H.

The results we obtained raise a new question of high importance for the understanding of the nature of the revealed processes. It is well known that the canting of the atomic moments from their directions in an expected symmetric magnetic structure can be the consequence of the Dzyaloshinskii-Moriya interaction (DMI) [37–39]. Recently the DMI experienced a strong revival of research interest because of its role in the stabilization of skyrmions [40–43] and and the formation of highly mobile chiral domain walls [44–46]. In this respect, it is of high interest to investigate the role of the DMI in the case of magnetic $5f$ systems. In the case of UNS, our concrete task is to understand if the DMI is responsible for the destabilization of the FM and NC1 structures or the destabilization is connected with the change of the MA as was expected at the beginning of the study.

To address this problem we consider the properties of the magnetic interactions in terms of the bilinear Hamiltonian of interacting atomic moments $H = \sum_{ij} \hat{\mathbf{S}}_i A^{(i,j)} \hat{\mathbf{S}}_j^T$ where $A^{(i,j)}$ are 3×3 matrices, T means matrix transposition, and $\hat{\mathbf{S}}_i$ is the unit vector in the direction of the ith atomic moment [47].

An arbitrary matrix can be represented as the sum of a symmetric and an antisymmetric matrix $A = B + C$. The energy contribution due to the antisymmetric part can be recast as $\hat{\mathbf{S}}_i C^{ij} \hat{\mathbf{S}}_j^T = \boldsymbol{D}_{ij} \cdot [\hat{\mathbf{S}}_i \times \hat{\mathbf{S}}_j]$ and corresponds to DMI. The DMI vector $\boldsymbol{D}_{ij}$ is defined by the elements of the antisymmetric matrix $\boldsymbol{D}_{ij} = (C_{xy}^{ij}, -C_{xz}^{ij}, C_{yz}^{ij})$. The symmetric parts B of the matrices A supply energy contributions of the isotropic Heisenberg's exchange and magnetic anisotropy. The on-site matrices $A^{(i,i)}$ are always symmetric and reflect the on-site anisotropy of atom i.

The symmetry constraint on matrices $A^{(i,j)}$ imposed by the symmetry operation $\{\alpha|\boldsymbol{\tau}_\alpha\}$ is given by the expression $A^{(i,j)} = \alpha^T A^{(i_\alpha, j_\alpha, \boldsymbol{R})} \alpha$ where the uranium sublattices i_α, j_α and lattice vector $\boldsymbol{R}$ are defined by the action of operation $\{\alpha|\boldsymbol{\tau}_\alpha\}$ on atoms i and j [34]. First we remark that the account for the 16 symmetry operations of the UNS or UNS-H with H atoms in the U plane results in the A matrices of the block-diagonal form for any pair of atoms: $A^{(i,j)} = \begin{pmatrix} * & * & 0 \\ * & * & 0 \\ 0 & 0 & * \end{pmatrix}$. Since the matrix elements (xz), (yz), (zx), (zy) responsible for the interaction of the in-plane and out-of-plane components of the atomic moments are zero, both the structure collinear to the z axis and the structures parallel to the xy plane are quasistable and their relative energies determine the ground magnetic state.

In the UNS-H case with shifted H atoms the interaction matrices lose their block-diagonal form. For the $A^{(1,j)}$ matrices describing the on-site MA of atom 1 and the interaction of atom 1 with atom 3_{100} we obtain

$$A^{(1,1)} = \begin{pmatrix} * & * & b \\ * & * & b \\ b & b & * \end{pmatrix}, \quad A^{(1,3_{100})} = \begin{pmatrix} * & * & c \\ * & * & c \\ -c & -c & * \end{pmatrix}. \tag{1}$$

The notation 3_{100} means the U atom of the third sublattice in the unit cell shifted by the lattice vector (100) (Fig. 1); the asterisk, $*$, replaces the matrix elements whose values are not important for the present discussion and b and c are nonzero numbers. Since the nonzero matrix elements b are symmetric in the matrix they correspond to the on-site anisotropy. In this case the easy axis will be neither parallel to the z axis nor lie in the xy plane. Instead, both the out-of-plane and in-plane components of the orientation vector of the easy axis are nonzero. On the other hand, since $A_{xz} = -A_{zx} = c$ and $A_{yz} = -A_{zy} = c$ for atoms 1 and 3_{100} we deal in this case with the DMI interaction between these atoms. The corresponding components of the DMI vector are D_y and D_x whose absolute values are equal to each other.

The interactions of atom 1 with atoms 2 and 2_{100} and with atoms 4 and $4_{0\bar{1}0}$ are given by the matrices

$$A^{(1,2+2_{100})} = \begin{pmatrix} * & * & d \\ * & * & e \\ d & -e & * \end{pmatrix}, \quad A^{(1,4+4_{0\bar{1}0})} = \begin{pmatrix} * & * & e \\ * & * & d \\ -e & d & * \end{pmatrix}. \tag{2}$$

Here there are both symmetric and antisymmetric contributions to the off-diagonal blocks, which means that both MA and the DMI contribute to the deviations of the moments from the z axis in the FM_z structure and from the xy plane in the NC1 structure. This analysis of the interatomic interaction

matrices shows that the DMI contributes to the change of the magnetic properties due to the H absorption that was first assumed to be the consequence of MA only [48].

To summarize, the transformation from the very strong uniaxial anisotropy to the apparent easy-plane anisotropy by absorption of hydrogen takes place through the formation of noncollinear magnetic states with atomic moments deviating from both the z axis and the xy plane. Since the deviation of H atoms up or down off the U planes is random in the real samples [26], we expect that the local canting of the moments towards the positive and negative directions of the z axis is also random leading to the compensation of the out-of-plane components of the magnetic moments over the sample. However, despite the global compensation of the z components of the magnetic moments it is the local canting that governs the energetics of the hydrogenated system. Although the discussion was started in terms of the giant MA change our microscopic study shows that both the MA and DMI contribute to the underlying physical processes.

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