Relativistic effects in the magnetism of UFe_4Al_8

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The magnetic structure of UFe_4Al_8 possesses a number of unique features. There are two magnetic sublattices whose magnetic moments are almost orthogonal. Additionally, there is a canting of the magnetic moments within the Fe sublattice. We show that these features can be treated as inevitable consequences of the properties of a simpler magnetic state of the system. It is shown that the magnetism of the U sublattice is induced by the compensated antiferromagnetism of the Fe sublattice. The orthogonality of the induced and inducing moments is explained. The unusual nature of the magnetic anisotropy is demonstrated. A crucial role of the spin-orbit coupling and the interatomic hybridization is exposed. The symmetry criterion of the magnetic instability of a nonmagnetic sublattice in an antiferromagnetic crystal is formulated. [S0163-1829(99)50434-7]

The uranium compounds form an interesting class of materials with widely varying electronic properties. One of the characteristic features of this class of materials is the noncollinearity of the magnetic structure observed in many uranium compounds.[1] Usually, the noncollinearity of the atomic magnetic moments is observed within the U sublattice. The present paper is devoted to the theoretical study of the magnetism of UFe_4Al_8 which differs essentially from the magnetism of other systems.

For a number of years the magnetic structure of UFe_4Al_8 was a matter of much controversy. The suggestions made on the basis of different experimental investigations ranged from simple one-sublattice ferromagnetism up to unusual spin-glass state.[2] Recent investigation of a single crystal of UFe_4Al_8 with the use of unpolarized and polarized neutron diffraction revealed an ordered magnetic structure with a number of unique features (Fig. 1).[3] Two magnetic sublattices were detected. The magnetic moments of the U sublattice form a collinear ferromagnetic structure. A strong noncollinearity is, however, observed between the U and Fe magnetic moments which are almost orthogonal to one another. The magnetic structure of the Fe sublattice is close to a collinear antiferromagnetic (so-called *G*-type antiferromagnetic) structure. Additionally, there is a canting of the Fe moments which leads to the second type of noncollinearity in UFe_4Al_8. The noncollinearity within the Fe sublattice results in a weak ferromagnetic moment parallel to the U moments. The purpose of the present study is to understand the magnetism of UFe_4Al_8, in particular to reveal the hierarchy of the interactions which lead to the formation of this complex magnetic structure.

The description of the calculational approach can be found in Ref. 4. For a part of the calculations the orbital polarization correction (OPC) in the form[5] $\hat{H}_{OP}=I_{OP},\ L\cdot\hat{l}$ was included into the Hamiltonian of the problem. Here L is the atomic orbital moment, $\hat{l}$ the operator of the angular momentum, and I_{OP} a parameter.

We begin with the study of the instability of the nonmagnetic state of UFe_4Al_8 with respect to the formation of the magnetism of the U and Fe sublattices. At the begining a small spin moment of $0.1\mu_B$ was put on each Fe and U atom. To account for experimental information, the Fe moments were directed collinear to the *a* axis and formed the *G*-antiferromagnetic structure. The U moments were directed parallel to the *b* axis. The directions of the moments were constrained and did not vary during calculations. None of the small initial moments collapsed during calculations. On the contrary, the moments increased and resulted in sizeable values for both sublattices. This result shows that, in agreement with experiment, two orthogonal sublattices can, indeed, coexist in UFe_4Al_8. The lengths of the spin, orbital, and total Fe moments were found to be $1.33\mu_B$, $0.07\mu_B$, and $1.40\mu_B$, respectively, in reasonable agreement with the experimental estimate for the total moment of $1.1\mu_B$. The theoretical values of the spin, orbital, and total U moments are $0.54\mu_B$, $-0.69\mu_B$, and $-0.15\mu_B$. Compared with the experimental estimates[3] of $0.52\mu_B$, $-0.99\mu_B$, and $-0.47\mu_B$, we obtained a close value of the spin moment, but a smaller value of the orbital moment. The underestimation of the U orbital moment is a well-known feature of the local spin-density functional (LSDF) theory. Below we comment on the results of the application of the OPC to this compound.

To investigate the interdependence of the magnetism of the Fe and U sublattices we carried out two model calculations. In the first calculation the Fe atoms were constrained

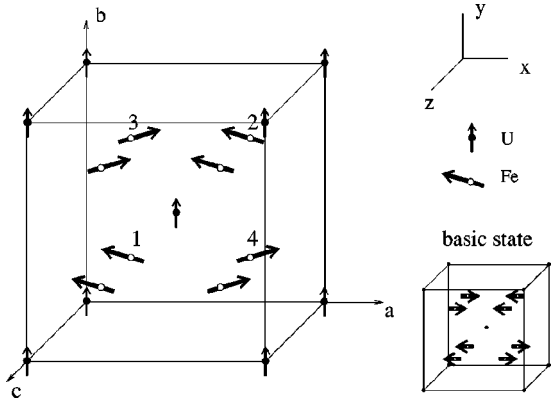

FIG. 1. Experimental magnetic structure (Ref. 3) and basic magnetic state of UFe_4Al_8.

to be nonmagnetic and the U atoms were free to develop their magnetic moments. In this case, a small magnetic moment put on the U atom collapsed and the state of the whole crystal was found to be nonmagnetic. In the second model calculation the U sublattice was constrained to be nonmagnetic. This restriction did not influence noticeably the magnetism of the Fe sublattice.

The results of the two model calculations reveal the principal role played by the Fe sublattice in the magnetism of UFe_4Al_8. In the following, we will show that the complex noncollinear magnetic structure of UFe_4Al_8 is a necessary consequence of the properties of the state of the crystal studied in the second model calculation, that is of the state with the nonmagnetic U sublattice and collinear antiferromagnetic Fe sublattice. To stress the importance of this state for understanding the magnetism of UFe_4Al_8 we will refer to it as the basic state (BS).

To study the influence of the magnetism of the Fe sublattice on the magnetic state of the U atoms the next calculation was started again with the BS. This time, however, no constraint was imposed on the U magnetic moment. Both the length and the direction of the possible U moment were not restricted. Calculations have shown that the U atoms immediately became magnetic, with the magnetic moments directed collinear to the *b* axis and orthogonal to the Fe moments. The value of the U magnetic moments increased during the iterational calculations and resulted in the self-consistent value cited above. The appearance of the magnetic moments at the initially nonmagnetic U sites allows to treat these magnetic moments as *induced* by the magnetism of the Fe sublattice.

Two features are important here. First, the vector sum of the Fe moments is zero. Because of this property and of a symmetrical position of the U atom with respect to the Fe atoms, the Heisenberg's exchange field of the Fe sublattice at the position of the U atom is zero. Therefore the Heisenberg model cannot explain the physics of this compound. The second important feature is the orthogonality of the inducing and induced moments.

These features raise the question about the physical mechanism of breaking the symmetry between two opposite directions of the *b* axis. Indeed, the ferromagnetism of the U sublattice distinguishes one of the two directions. Although the symmetry breaking is a common feature of any ferromagnetism, the present case differs essentially from the usual case of, for instance, the ferromagnetism of bcc Fe.

In bcc Fe, both up and down directions of the parallel atomic moments are equivalent and any of them can be assumed by the moments with an equal probability of 50%. The equivalence of the two ferromagnetic states of bcc Fe can be easily proved by applying the operation of the time reversal to one of the states. In the case of UFe_4Al_8, application of the time-reversal operation changes the directions of both induced and inducing moments and does not help us in studying the properties of the U sublattice. To understand the origin of the symmetry breaking in UFe_4Al_8 we must restrict the consideration to those transformations of the system which leave the inducing magnetic structure invariant.

In Table I we collected the generators of the symmetry group of the BS of UFe_4Al_8. The third column shows the transformation of the coordinates of an axial vector under action of these operations.

TABLE I. Generators of the symmetry group of the basic state of UFe_4Al_8. C_{2x} and C_{2y} are 180° rotations about the *x* and *y* axis, respectively; *I*, inversion; *R*, time reversal.

Operation	Transposition of Fe atoms	Restriction on magnetic moments of U and Fe atoms
C_{2y}	$1\leftrightarrow 3; 2\leftrightarrow 4$	$\begin{pmatrix} m_x \\ m_y \\ m_z \end{pmatrix}_i = \begin{pmatrix} -m_x \\ m_y \\ -m_z \end{pmatrix}_j$
I	no	no
$C_{2x}R$	$1\leftrightarrow 4; 2\leftrightarrow 3$	$\begin{pmatrix} m_x \\ m_y \\ m_z \end{pmatrix}_i = \begin{pmatrix} -m_x \\ m_y \\ m_z \end{pmatrix}_j$

[a]For the U sublattice $i=j$; for the Fe sublattice *i* and *j* according to the column ''Transposition of Fe atoms.''

Two opposite directions of the U magnetic moments can be equivalent only in the case when there is a symmetry operation of the system which reverses the direction of the magnetic moment. In the present case it must be an operation which reverses m_y. Analysis of Table I shows that this condition is not fulfilled. All symmetry transformations leave m_y invariant.

This symmetry breaking is, evidently, a necessary condition for the appearance of the induced moment on the U sites. A stronger statement can, however, be formulated: the magnetic moment on the U sites not only *can* but also *must* appear. Indeed, since the symmetry operations of the BS (Table I) keep m_y unchanged no condition is imposed on the m_y value by the symmetry of the problem. Therefore none of the m_y values is distinguished by symmetry compared to other values. In this situation the probability, of the event that an arbitrary selected state with $m_y=0$ supplies the ground state of the system, is negligible and the magnetic moment must appear at the U site. We can formulate the criterion of this type of magnetic instability. *A nonmagnetic state of a given type of atoms in a magnetic crystal cannot be stable if this state is not distinguished by symmetry when compared to states where these atoms possess an infinitesimal magnetic moment.*

The notion of the symmetry predetermined instability was already introduced by us in an earlier work. In this case, the context was the study of the stability of collinear magnetic configurations.[6,7] The symmetry criterion similar to that given above was formulated. Both criteria can be combined in the following generalized statement. Suppose there is a continuous parameter which describes different magnetic states of the system. Then *the state corresponding to a selected value of the parameter can be stable only in the case when this state is distinguished by symmetry compared to the states obtained with an infinitesimal variation of the parameter*. Depending on the physical problem, the role of the parameter can be played by the length of the atomic moment or by the deviation of the magnetic moments from the collinear directions.

The symmetry predetermined instability of the nonmagnetic state of the U atoms can be illustrated by the properties of the total energy as a function of m_y. The most important feature of the $E(m)$ curve is its asymmetry which leads to an accidental position of the minimum at a nonzero value of m_y.

The orthogonality of the inducing and induced moments is also closely connected with the symmetry properties of the system. In our earlier publications we formulated a symmetry principle according to which *none of the symmetry operations of the initial Kohn-Sham Hamiltonian can be disturbed during the self-consistent calculations*.[8,7] This statement is complementary to the instability criterion formulated above. If we again inspect Table I we see that the only possible direction of the U moments which is invariant with respect to all symmetry operations is the direction collinear to the *b* axis. This direction was obtained both experimentally and theoretically. Thus, we have shown that the magnetism of the U sublattice is a consequence of the symmetry properties of the BS of UFe_4Al_8. No exchange interaction between the U moments is needed for the establishment of the ferromagnetism of the U sublattice.

The next feature of the experimental magnetic structure we want to understand is the noncollinearity within the Fe sublattice. Again, we started the calculation with the BS of the crystal and removed the constraints not only on the U but also on the Fe magnetic moments. After first iteration, additionally the appearance of the magnetic moments on the U sites, we obtained the canting of the Fe moments. The m_z components of different atomic moments have different signs and compensate one another. On the contrary, the m_y components of all atomic moments are equal and, in agreement with experiment, result in a weak ferromagnetic moment along the *b* axis.

The necessity of the canting of the Fe moments follows immediately from the criterion of the instability of the collinear magnetic structures. Indeed, from Table I we see that the canting of the Fe moments in the form obtained in the calculations disturbs none of the symmetry operations of the BS. Therefore, the collinear structure is not distinguished by symmetry and cannot be stable. The simultaneous appearance of the ferromagnetism of the U sublattice and of the canting of the Fe moments shows that none of them can be considered as a consequence of another. They are two interconnected consequences of the properties of the BS of the system.

To reveal the physical interactions which are essential for the appearance of both effects we carried out two further model calculations. In the first calculation, the spin-orbit coupling (SOC) was set to zero. In the second, the hybridization between the U states and states of other atoms was neglected. In each of the calculations both effects disappear. For explanation of this result it is important that in each of the two cases the neglect of the part of interactions increases the symmetry of the system in such a way that the nonmagnetic state of the U sublattice and the collinear configuration of the Fe sublattice are distinguished by symmetry and cannot change during calculations. Thus, the neglect of the SOC leads to the Hamiltonian whose symmetry must be described with the use of the generalized spin-space groups[7] which allow the separate point transformation of spin and space variables. The influence of the intersublattice hybridization becomes evident if we notice that both the U and Fe sublattices taken separately are more symmetrical than their combination in the UFe_4Al_8 crystal structure.

Thus, we have shown that both types of noncollinearity are necessary consequences of the properties of the BS. However, the BS itself is formed by us on the basis of experimental information. To complete the theoretical study we must justify the use of the BS as a starting point in the consideration of the unusual properties of UFe_4Al_8. The magnetic structure of the BS can be characterized by two features: the orientation of the Fe moments with respect to one another and the orientation of the magnetic moments relative to the crystal lattice. Correspondingly, two properties must be verified: first, the different relative orientations of the Fe magnetic moments have higher total energy compared to the total energy of the *G*-antiferromagnetic configuration and, second, the character of the magnetic anisotropy in the crystal, indeed, makes the direction of the Fe moments along the *a* axis energetically preferable compared to the direction parallel to the *c* axis. (Crystallographic *a* and *b* axes are equivalent.)

To verify the first property the calculations were carried out for ferromagnetic and another for antiferromagnetic ($\mathbf{m}_1 = -\mathbf{m}_2 = \mathbf{m}_3 = -\mathbf{m}_4$) configurations of the Fe magnetic moments. In both cases we obtained substantial increase in the total energy compared to the BS.

To verify the second property the calculation was carried out for the state of the system similar to the BS but with the Fe moments collinear to the *c* axis. This calculation, however, did not give the expected result: within the accuracy of the LSDF calculation of about 0.1 mRy per formula unit, the energy of the calculated state was indistinguishable from the total energy of the BS.

The physical origin of the magnetic anisotropy became evident when the constraint on the U atoms was removed. In contrast to the BS, no magnetic moments appeared on the U sites in this case. As the appearance of the induced U moments leads to the decrease of the total energy of the system (Fig. 2), the direction of the Fe moments collinear to the *a* axis became energetically preferable. Thus we obtained an important result that the magnetic anisotropy in UFe_4Al_8 is governed not by the Fe sublattice itself but by the properties of an induced magnetic moment of the U sublattice.

To understand why no induced magnetic moment appeared on the U sites for the Fe magnetic moments collinear to the *c* axis, note that the symmetry of this state is higher than the symmetry of the BS. The increased symmetry is a consequence of the crystallographic equivalence of the *a* and *b* axes. As a result, the nonmagnetic state of the U atoms is distinguished by symmetry and is stable. The calculation of the total energy as a function of the uranium spin moment gave, in contrast to the case of the BS, a symmetrical curve with minimum at $m = 0$ (Fig. 2).

Summarizing, the calculations within the LSDF theory combined with the analysis of the symmetry of the problem allowed us to explain all the important features of the complex magnetic structure of UFe_4Al_8. Quantitative comparison of the experimental and theoretical quantities reveals, however, a noticeable difference. Thus the theoretical canting angle of the Fe moments is about 8° compared with the

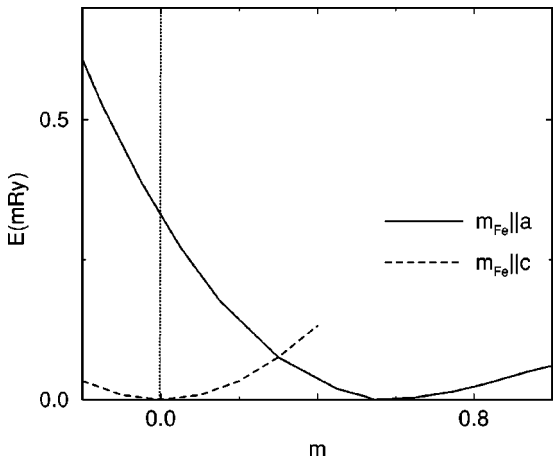

FIG. 2. Total energy as a function of the U spin moment for the Fe moments collinear to the *a* (basic state) and *c* axes.

experimental value of 16°. Also the theoretical value of the U orbital moment is too small (see above). The calculation with the use of the OPC did not lead to the desired improvement of the total U moment. We carried out calculations with different values of I_{OP}. Surprisingly, the U moment decreased rather than increased for small values of the parameter. Simultaneously, the canting of the Fe moments decreased. Therefore the agreement with experiment became worse. Analysis of this unexpected result has shown that the orbital polarization correction leads in this case to the decrease of the spin moment of the U atom. As the value of the U spin moment is important for both the value of the orbital moment and the canting of the Fe moments, these quantities decreased as well. For an orbital parameter of about 1 mRy the value of the U moment jumps to a large value close to $2\mu_B$. Simultaneously, the canting of the Fe moments increases to about 50°. As a result, the obtained value of the magnetic moment per formula unit exceeded the experimental value several times. Presently, we cannot give a detailed explanation of the unexpected influence of the OPC. We note, however, that the situation in UFe_4Al_8 is more peculiar than in the compounds studied so far. As we have shown the magnetism of the U sublattice and canting of the Fe moments are the consequences of the magnetism of the Fe sublattice and of the hybridization of the electron states of different atoms. Obviously, the OPC in the form used in the present calculations influences the important hybridization between the sublattices incorrectly. The unique properties of the intersublattice interaction in UFe_4Al_8 make this compound an important test system for further development of the theoretical schemes (such as the OPC, LDA+U, and self-interaction correction schemes) aiming to improve the standard LSDF theory by taking into account the influence of the strong spatial localization and intra-atomic correlations of the $5f$ electron states. The application of different theoretical schemes to UFe_4Al_8 must be, however, a topic of separate investigation.

A number of results presented in this paper are of general interest. Thus, the approach to the study of the state of the atoms which do not possess an intrinsic magnetic moment is universal and can be applied to any magnetic system. One of the possible examples is the multilayer systems where the magnetic and nonmagnetic layers alternate. Also the phenomenon of the magnetic anisotropy governed by the property of the atoms which do not bear an intrinsic magnetic moment is general and can be important in various magnetic systems.

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