The hidden order in URu$_2$Si$_2$: High-symmetry anti-toroidal vortexes

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Introduction.—For more than thirty years, there were many attempts to understand the mysterious Hidden Order (HO) in URu$_2$Si$_2$ crystal (they are surveyed in two detailed reviews [1,2] whereas the first publications appeared in 1985 [3–5]). The main problem is that below the HO transition temperature, $T_{HO} = 17.5$ K, there are practically no obvious physical phenomena associated with the order parameter; the only unequivocal evidence for the order is a rather strong specific heat jump [3–5] at $T_{HO}$. For instance, the observed antiferromagnetic order [6,7] is so weak that it cannot explain the behavior of the specific heat. The observed lattice symmetry breaking [8] and in-plane anisotropy of the magnetic susceptibility [9] are also extremely weak and their relation to HO is not clear [12]. However most of researchers agree that at $T_{HO}$ there is time-reversal symmetry breaking (TRSB) related probably with an exotic type of multipole magnetic order [2,13,14]. Many efforts, theoretical and experimental, were concentrated on searching for possible multipole orders [2,12,10].

In this Letter, we suggest a simple HO model based mainly on the symmetry consideration. Indeed, if we cannot detect any pronounced violation of the spatial symmetry below $T_{HO}$, let us assume that the HO has exactly the same symmetry, namely $4/mmm$, as the high temperature phase of URu$_2$Si$_2$. More precisely, we suppose that HO is a non-collinear intra-atomic magnetization of uranium atoms with $4/mmm$ symmetry so that the only symmetry violation at the transition point is TRSB. Surprisingly, such a simple assumption leads to a non-trivial vortex HO which can be described by the toroidal quadrupole order parameter. First-principles calculations show that the vortex HO is perhaps strong enough to be detected by neutron scattering.

Magnetic symmetry of the HO.—We first remind that the magnetic moment $\mathbf{M}(r)$ is a pseudo-vector and transformations of its components under mirror reflections are just opposite to a usual vector: the component normal to the mirror plane keeps its direction whereas the parallel components invert their directions. For instance, for the $m_z$ mirror plane, $M_z(x, y, -z) = M_z(x, y, z)$, $M_x(x, y, -z) = -M_x(x, y, z)$, and $M_y(x, y, -z) = -M_y(x, y, z)$. The space inversion does not change $\mathbf{M}(r)$: $\mathbf{M}(-r) = \mathbf{M}(r)$. The time reversal symmetry operation, denoted by the prime sign, inverts the direction of the magnetization: $\mathbf{M}'(r) = -\mathbf{M}(r)$.

The principal difference between conventional magnetic atoms and a magnetic atom with $4/mmm$ symmetry is obvious from Fig. [1]. The magnetic point symmetry of conventional atoms would be $4/mm'm''$ and it includes one vertical 4-fold axis, one horizontal mirror plane $m$ and two types of vertical mirror planes $m''$ (normal and diagonal to $x, y$ axes). As a result of this symmetry, $M_z(x, y, 0) = 0$ and $M_y(x, y, 0) = 0$ in the $z = 0$ mirror plane and usually these components remain to be small above and below the mirror plane so that the main magnetization of the atom is $M_z$.

For the case of $4/mmm$ symmetry, $M_z$ and $M_y$ are also zero in the plane of the figure but $M_x$ should be very inhomogeneous, it should change its sign at least eight times when we go around the atom (Fig. [1b]). In the horizontal mirror plane $z = 0$, we have eight similar sectors with alternating $M_z$-component. Then, passing through all vertical mirror planes, shown in Fig. [1b] by black lines, the parallel components of $\mathbf{M}(r)$ become zero and change their signs. In other words, for all $r$ belonging to the mirror planes of $4/mmm$ symmetry, $\mathbf{M}(r)$ is normal to the corresponding plane.

For any general position $x, y, z$ the $4/mmm$ symmetry operations create a pair of eight-vector vortexes with

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head-to-tail arrangement of equivalent moments in the $\pm z$ planes (Fig. 1). The toroidal moments $\mathbf{M}_{ijk}$ of these two eight-vector vortexes are anti-parallel (along $\pm z$). It is a general magnetic arrangement dictated by the $4/mmm$ symmetry and it makes the $\mathbf{M}(\mathbf{r})$ field significantly non-collinear and inhomogeneous simply as a result of the symmetry. Each uranium atom looks like an atomic-size magnetic skyrmion built from two equivalent halves at $z > 0$ and $z < 0$ with opposite toroidal moments. We could refer to this configuration as an Anti-Toroidal Vortex (ATV). It should be emphasized that the $4/mmm$ symmetry induces the ATV structure only for pseudo-vectors like $\mathbf{M}(\mathbf{r})$ and not for true vectors like electric dipole moments, etc. (d) In principle, higher symmetries are also possible, up to $\infty/mmm$, which is the symmetry of the nematic order.

To characterize quantitatively the inhomogeneous atomic magnetization with $4/mmm$ point symmetry we can use the tensor moments of $\mathbf{M}(\mathbf{r})$ relative to the atomic center. The average dipole moment $\langle \mathbf{M}(\mathbf{r}) \rangle$ is zero. Here and below $\langle \ldots \rangle$ means integration $V^{-1} \int \ldots d\mathbf{r}$ over a spherical atomic-size volume $V$ around the atom.

The magnetic quadrupole moment $\langle \mathbf{M}(\mathbf{r}) \mathbf{x}_j \rangle = 0$ because of the inversion center $\mathbf{M}(-\mathbf{r}) = \mathbf{M}(\mathbf{r})$. In particular, the atomic toroidal (anapole) moment $\langle \mathbf{r} \times \mathbf{M}(\mathbf{r}) \rangle$ [17-19], which is an antisymmetric part of this tensor, is zero as well as the monopole moment $\langle \mathbf{r} \cdot \mathbf{M}(\mathbf{r}) \rangle$. For the same reason, all even-rank tensor moments of $\langle \mathbf{M}(\mathbf{r}) \mathbf{x}_j \times \ldots \times x_n \rangle$ type are zero as well.

Thus the first non-zero tensor moment of the $4/mmm$ ATV structure is the third-rank tensor $M_{ijk} = \langle \mathbf{M}(\mathbf{r}) \mathbf{x}_i \mathbf{x}_j \rangle$; it is symmetric under permutation of the last two indices. It is easy to show (or to find in textbooks [20]) that for this symmetry the third-rank pseudo-tensor $M_{ijk}$ has only four non-zero components and all of them are equal up to the sign: $M_{123} = M_{132} = -M_{231} = -M_{213} = M_0$, where $M_0 = \frac{4}{3} V^{-1} \int \langle \mathbf{M}(\mathbf{r}) \mathbf{r} \rangle \cdot d\mathbf{r}$. The time-odd parity-even moment $M_0$ characterizes the strength and sign of the ATV HO (it is called either magnetic octopole or quadrupole toroidal moment).

The sign of $M_0$ is a non-trivial attribute. Indeed, we can change the sign of $M_0$ by reversing the magnetization direction in all points $\mathbf{M}(\mathbf{r}) \rightarrow -\mathbf{M}(\mathbf{r})$ (the time reversal operation). However this way we obtain a new object which cannot be superposed with the old one neither by rotations nor by mirror reflections. Since the only symmetry operation relating these two objects is the time inversion, their energies must be equal. Thus any magnetization arrangement with $4/mmm$ point symmetry can exist in two energetically equivalent variants with $\pm M_0$. It is natural to call them clockwise and anticlockwise vortexes for $M_0 > 0$ and $M_0 < 0$, correspondingly. However, it should be emphasized that the sign of $M_0$ is not topologically stable, it can be changed by deformation of the $\mathbf{M}(\mathbf{r})$ field. The magnetization arrangement with $M_0 = 0$ can correspond to non-zero absolute magnetization $\langle |\mathbf{M}(\mathbf{r})| \rangle \neq 0$.

There are two uranium atoms in the body-centered tetragonal unit cell of URu$_2$Si$_2$. In the simplest magnetic structure, both atoms have the vortexes with the same $M_0$, either both clockwise or both anticlockwise. Such structure has $I4/mmm$ magnetic space group [21] and can be called the ferro-vortex phase. The clockwise and anticlockwise ferro-vortex phases should have equal energies and can be mutually transformed by the time reversal. In real samples of the ferro-vortex phase, the clockwise and anticlockwise domains can coexist.

If those two atoms have opposite magnetization directions (clockwise and anticlockwise) then the lattice is primitive and the magnetic symmetry group is $P14/mmm$ [21]. In this case, the lattice consists of clockwise and anticlockwise layers alternating along the $z$-axis; it can be called the antiferro-vortex phase. The time reversal is equivalent to the $(\frac{1}{2}, \frac{1}{2}, 0)$ shift of the lattice.
Besides ferro-vortex and antiferro-vortex phases many (infinite!) symmetrically different arrangements of the clockwise and anticlockwise vortexes are possible but their consideration should be left for the future work. Then, in principle, ATV with higher symmetries are also possible, up to \( \infty/\text{mm} \), which is the symmetry of the nematic order (see Fig. 11). An open question is whether the vortexes with such a high symmetry can exist in free atoms, molecules or nematic-like liquid crystals. Actually, the toroidal quadrupole moments are discussed for positronium atoms [22] and for deuterons [23] (a survey of related works is given in [24]).

The quantitative characterization of \( 4/\text{mmm} \) vortexes by the third-rank tensor \( M_{ijk} \) has three important complications: (i) \( M_{ijk} \) does not depend on the azimuthal orientation of the vortex in the \( xy \) plane: (ii) it does not distinguish between \( 4/\text{mmm} \) and other uniaxial symmetries (422, 4mm, 42m, 622, 6mm, 62m, 6/\text{mmm}, \( \infty/2 \), \( \infty/\text{mm} \)); (iii) the \( M_z \) component gives no contribution to \( M_s \). Some of these drawbacks disappear for the next non-zero tensor (fifth-rank) and for the magnetoelectric tensor \( \langle M_i(r)E_j(r)x_k \rangle \). All this means that pure symmetrical consideration leaves a lot of freedom for possible scenarios of the HO transition and more work is needed here.

First principle simulations.—The symmetry-based approach is of course reliable, but it cannot say whether and when those exotic anti-toroidal vortexes could be energetically stable, what are the values of \( M(r) \) in different points of the unit cell, etc. To find the magnetization \( M(r) \), the electronic densities \( \rho(r) \) and the energies of possible URu\(_2\)Si\(_2\) phases we have performed “illustrative” ab initio simulations using the QUANTUM ESPRESSO package [25, 26].

We do not fix the spatial and magnetic symmetries of URu\(_2\)Si\(_2\) in the beginning and during the self-consistent minimization procedure. Instead, the procedure starts from crystal structures whose symmetries are subgroups of \( 4/\text{mmm} \). Small initial magnetic moments are assigned to silicon and ruthenium atoms so that uranium magnetic moments are not predetermined. Then during the self-consistent iterations those conventional magnetic moments become smaller and smaller but at the same time new magnetization field \( M(r) \) (with zero average magnetization) is growing mainly around uranium atoms, i.e. the absolute magnetization \( (|M(r)|) \) is progressively growing until an equilibrium structure is reached. Symmetry analysis of the appearing magnetization shows that new symmetry elements initially look like some tendency and then become more and more exact if the iterative self-consistent procedure converges. See Supplementary materials for more details of the simulations.

Both the ferro-vortex and antiferro-vortex phases have been obtained in our simulations starting from different initial structures. Their energies are well below the energy of non-magnetic phase: per formula unit, \( \Delta E_{\text{fer}} = -0.0318 \text{ eV/f.u.} \) and \( \Delta E_{\text{ant}} = -0.0364 \text{ eV/f.u.} \). This energy gain seems to be too strong for the observed value of the specific heat jump corresponding to the internal energy change induced by the hidden order of about 0.00018 eV/f.u. In fact, the energy responsible for the HO phase transition is of about an interaction energy between magnetic atoms, which is “fighting” with entropy for the phase transition. The interaction energy is a very small part of the total magnetic energy and the former is impossible to extract from the latter within the conventional DFT simulations. Quite probably, the antitoroidal vortexes appear as fluctuations well above the HO transition temperature, and they are arranged into ferro-vortex or antiferro-vortex phase at the HO transition temperature owing to very subtle interactions between vortexes.

The calculated magnetization and charge densities are shown in Fig. 2 for the diagonal mirror plane \( x = y \) including two U atoms. The main magnetic and charge features obviously correspond to the 5f uranium orbitals [27] (mean radius 0.76 Å). The uranium vortexes are almost the same for both phases, except that in the antiferro-vortex phase they have opposite signs. And the total absolute magnetization is almost the same for both phases: \( |M(r)|_{\text{av}} = 0.93 \mu_B/\text{f.u.} \) and \( |M(r)_{\text{av}}| = 0.96 \mu_B/\text{f.u.} \). According to Ref. [28], the value of about 1 \( \mu_B/\text{f.u.} \) is needed to explain the observed specific heat jump. The magnetization is concentrated around uranium atoms (Fig. 2a,b): in the ferro-vortex(antiferro-vortex) phase, inside the Slater uranium radii \( (R_S = 1.75 \text{ Å}) \), there is about 0.936 (0.93) of the total |M(r)| and remaining itinerant magnetization is distributed in the unit cells according to their space symmetries. The very strong anisotropy of ATVs could naturally explain the Ising-like behavior of HO [2, 12]. The calculated \( M_z \) for one atom is shown in Fig. 1b which is a 0.5 × 0.5 part of the unit cell \( xy \) plane (i.e. about 2 × 2 Å\(^2\)); see also Supplementary materials.

In fact, it is well known that magnetic intra-atomic non-collinearity is a general effect, arising because of the relativistic spin-orbit coupling not only in actinides [29] but also in other materials [30, 32]. The non-collinear magnetism is very sensitive to the space group symmetry and we have predicted recently [33] the toroidal intra-atomic moments for RhGe crystal with the \( P2_13 \) space group. The case of URu\(_2\)Si\(_2\) is especially interesting because its symmetry is so high that observation of its intra-atomic vortexes is really a non-trivial problem.

Discussion: how to observe the ATV HO.—Our symmetry consideration favors the ATV HO with very unusual distributions of the intra-atomic magnetization that results in unusual form-factors for magnetic neutron scattering (see Fig. 3 for the reflection intensities obtained from calculated \( M(r) \) with). An obvious unusual feature is that high-symmetry reflections \( h00, 0k0, 00l, \) and \( hh0 \) are zero for both phases. The main differ-
FIG. 2: (Color online) The calculated magnetization distribution \( M_x(r) \) within the diagonal mirror plane formed by vectors \([1, 1, 0]\) and \([0, 0, 1]\) in the unit cell of the ferro vortex (a) and antiferro vortex (b) phases; (c) the calculated valence electron density which is almost equal for both phases. In this plane, \( M_y(r) = -M_x(r) \) and \( M_z(r) = 0 \). Two uranium atoms are at \((\frac{1}{4}, \frac{1}{4}, \frac{1}{4})\) and \((\frac{3}{4}, \frac{3}{4}, \frac{3}{4})\) positions with Si atoms surrounding them; Ru atoms are out of the plane. The straight lines are intersections with vertical and horizontal mirror planes where \( M_x \) and \( M_y \) change their signs.

Comparison between two phases is that there are pure magnetic reflections for \( h + k + \ell = 2n + 1 \) in the antiferro vortex phase whereas for the ferro vortex phase all the magnetic reflections coincide with nuclear reflections \( h + k + \ell = 2n \). Comparison of Fig. 3 with the observed intensities of pure magnetic reflections \( h \) (100, 102, 201, 203, 106, and 300) allows us to exclude the antiferro vortex phase from the list of possible candidates for HO.

The situation with the ferro vortex phase is much more intriguing: the magnetic reflections only slightly change the nuclear reflection intensities; the latter have never been measured carefully for URu\(_2\)Si\(_2\) across the HO temperature. Moreover, the interference between magnetic and nuclear contributions should vanish in the case of equal fractions of clockwise and anticlockwise domains. According to our calculations, the magnetic structure factor can reach its maximum \( \approx 0.25 \mu_B \) for reflection 525 at \( T = 0 \). However, this reflection has a large nuclear structure factor. There are many weak nuclear reflections with comparable magnetic factors from 0.15 to 0.2 \( \mu_B \), for instance, 307 and 417; they are more sensitive to magnetic scattering. It seems that accurate measurements of neutron reflections as a function of temperature provide the only way to study ATV HO quantitatively. Similar neutron experiments have revealed an unusual magnetic order preserving translational symmetry in the pseudogap phase of high-temperature superconductors [34–37]. We have found recently a striking similarity between hidden orders in the pseudogap phase and URu\(_2\)Si\(_2\) that will be discussed elsewhere.

In conclusion, it is shown that high magnetic symmetry of URu\(_2\)Si\(_2\) crystal can explain why its “hidden order” remains hidden for many years. There is no spatial symmetry breaking in the HO phase transition and solely the time-reversal symmetry is violated. Owing to their
4/mmm symmetry, uranium atoms have zero dipole and quadrupole moments, and the first non-zero magnetic moment of the uranium vortex is the quadrupole toroidal moment which can be used as an order parameter in the Landau theory of the HO phase. The simulations suggest that the vortex magnetic order of URu$_2$Si$_2$ is indeed energetically favorable and strong enough to be detected by neutron diffraction.

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[1] J. A. Mydosh and P. M. Oppeneer, Rev. Mod. Phys. 83, 1301 (2011).
[2] J. A. Mydosh and P. M. Oppeneer, Phil. Mag. 94, 3642 (2014).
[3] T. T. M. Palstra, A. A. Menovsky, J. van den Berg, A. J. Dirkmaat, P. H. Kes, G. J. Nieuwenhuys, and J. A. Mydosh, Phys. Rev. Lett. 55, 2727 (1985).
[4] M. B. Maple, J. W. Chen, Y. Dalichaouch, T. Kohara, C. Rossel, M. S. Torikachvili, M. W. McElfresh, and J. D. Thompson, Phys. Rev. Lett. 56, 185 (1986).
[5] W. Schlabitz, J. Baumann, B. Pollit, U. Rauchschwalbe, H. M. Mayer, U. Ahlheim, and C. D. Bredl, Z. Phys. B - Cond. Mat. 62, 171 (1986).
[6] C. Broholm, J. K. Kjems, W. J. L. Buyers, P. Matthews, T. T. M. Palstra, A. A. Menovsky, and J. A. Mydosh, Phys. Rev. Lett. 58, 1467 (1987).
[7] E. D. Isaacs, D. B. McWhan, R. N. Kleiman, D. J. Bishop, G. E. Ice, P. Zschack, B. D. Gaulin, T. E. Mason, J. D. Garrett, and W. J. L. Buyers, Phys. Rev. Lett. 65, 3185 (1990).
[8] C. Broholm, H. Lin, P. T. Matthews, T. E. Mason, W. J. L. Buyers, M. F. Collins, A. A. Menovsky, J. A. Mydosh, and J. K. Kjems, Phys. Rev. B 43, 12809 (1991).
[9] H. C. Walker, R. Caciuffo, D. Aoki, F. Bourdarot, G. H. Lander, and J. Flouquet, Phys. Rev. B 83, 193102 (2011).
[10] S. Tonegawa, S. Kasahara, T. Fukuda, K. Sugimoto, N. Yasuda, Y. Tsuruhara, D. Watanabe, Y. Mizukami, Y. Haga, T. D. Matsuda, E. Yamamoto, Y. Onuki, H. Ikeda, Y. Matsuda, and T. Shibauchi, Nat. Commun. 5, 4188 (2014).
[11] R. Okazaki, T. Shibauchi, H. J. Shi, Y. Haga, T. D. Matsuda, E. Yamamoto, Y. Onuki, H. Ikeda, and Y. Matsuda, Science 331, 439 (2011).
[12] J. Trinh, E. Brück, T. Siegrist, R. Flint, P. Chandra, P. Coleman, and A. P. Ramirez, Phys. Rev. Lett. 117, 157201 (2016).
[13] M. B. Walker, W. J. L. Buyers, Z. Tun, W. Que, A. A. Menovsky, and J. D. Garrett, Phys. Rev. Lett. 71, 2630 (1993).
[14] S. Takagi, S. Ishihara, M. Yokoyama, and H. Amitsuka, J. Phys. Soc. Japan 81, 114710 (2012).
[15] D. D. Khalyavin, S. W. Lovesey, A. N. Dobrynin, E. Ressouche, R. Ballou, and J. Flouquet, J. Phys.: Condens. Matter 26, 046003 (2014).
[16] M.-T. Suzuki and H. Ikeda, Phys. Rev. B 90, 184407 (2014); see also the recent topical review: M.-T. Suzuki, H. Ikeda, and P. M. Oppeneer, First-principles theory of magnetic multipoles in condensed matter systems. arXiv:1802.00925 (2018).
[17] V. M. Dubovik and V. V. Tugushev, Phys. Rep. 187, 145 (1990).
[18] N. Spaldin, M. Fiebig, and M. Mostovoy, J. Phys.: Condens. Matter, 20, 434203 (2008).
[19] Yu. V. Kopaev, Phys. Usp. 52, 111 (2009).
[20] Yu. I. Sirotin and M. P. Shaskolskaya, Fundamentals of Crystal Physics, Mir Publisher, Moscow, 1982.
[21] http://www.cryst.ehu.es/}
[22] S. G. Porsev, Phys. Rev. A 49, 5105 (1994).
[23] E. Mereghetti, J. de Vries, R. G. E. Timmermans, and U. van Kolck, Phys. Rev. C 88, 034001 (2013).
[24] C. G. Gray, G. Karl, and V. A. Novikov, Am. J. Phys.
[25] http://www.quantum-espresso.org/

[26] P. Giannozzi, S. Baroni, N. Bonini, M. Calandra, R. Car, C. Cavazzoni, D. Ceresoli, G. L. Chiarotti, M. Cococcioni, I. Dabo, A. Dal Corso, S. de Gironcoli, S. Fabris, G. Fratesi, R. Gebauer, U. Gerstmann, C. Gougoussi, A. Kokalj, M. Lazzeri, L. Martin-Samos, N. Marzari, F. Mauri, R. Mazzarello, S. Paolini, A. Pasquarello, L. Paulatto, C. Sbraccia, S. Scandolo, G. Sclauzero, A. P. Seitsonen, A. Smogunov, P. Umari, and R. M. Wentzcovitch, J. Phys.: Condens. Matter 21, 395502 (2009).

[27] K. O. Kvashnina, H. C. Walker, N. Magnani, G. H. Lander, and R. Caciuffo, Phys. Rev. B 95, 245103 (2017).

[28] H. Amitsuka, M. Yokoyama, S. Miyazaki, K. Tenya, T. Sakakibara, W. Higemoto, K. Nagamine, K. Matsuda, Y. Kohori, and T. Kohara, Physica B 312-313, 390 (2002).

[29] L. Nordström and D. J. Singh, Phys. Rev. Lett. 76, 4420 (1996).

[30] L. M. Sandratskii, Adv. Phys. 47, 91 (1998).

[31] F. Bultmark. Distorted Space and Multipoles in Electronic Structure Calculations. Thesis. Digital Comprehensive Summaries of Uppsala Dissertations from the Faculty of Science and Technology 601. 64 pp. 2009. ISBN: 978-91-554-7407-2

[32] P.-W. Ma and S. L. Dudarev, Phys. Rev. B 91, 054420 (2015).

[33] A. V. Tsvyashchenko, V. A. Sidorov, A. E. Petrova, L. N. Fomicheva, I. P. Zibrov, and V. E. Dmitrienko, J. Alloys Compd. 686, 431 (2016).

[34] B. Fauqué, Y. Sidis, V. Hinkov, S. Pailhès, C. T. Lin, X. Chaud, and P. Bourges, Phys. Rev. Lett. 96, 197001 (2006).

[35] H. A. Mook, Y. Sidis, B. Fauché, V. Balédent, and P. Bourges, Phys. Rev. B 78, 020506 (2008).

[36] Y. Li, V. Balédent, N. Barišić, Y. Cho, B. Fauché, Y. Sidis, G. Yu, X. Zhao, P. Bourges, and M. Greven, Nature (London) 455, 372 (2008).

[37] Y. Li, V. Balédent, N. Barišić, Y. Cho, Y. Sidis, G. Yu, X. Zhao, P. Bourges, and M. Greven, Phys. Rev. B 84, 224508 (2011).