

Three Localized f Electrons in UPd₂Al₃ and in UGe₂ Intermetallics

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We have calculated the strength of the excitations between the crystal-field states which are in agreement with inelastic-neutron-scattering results. This agreement confirms the existence in the heavy-fermion superconductor UPd₂Al₃ the crystal-field electronic structure being the finger-print of the U³⁺ ions with three localized f electrons forming strongly-correlated atomic-like quantum system $5f^3$. The ionic integrity and the low-energy crystal-field electronic structure is preserved in this metallic system in the meV scale as has been postulated in the Quantum Atomistic Solid State theory (QUASST). We provide preliminary results with the U³⁺ ion in UGe₂ showing the ground-state eigenfunction which reproduces the ordered magnetic-moment value of 1.48 μ_B . This moment is composed from the dominant orbital contribution (2.6 μ_B) and the opposite spin moment (1.12 μ_B).

DOI: [10.12693/APhysPolA.130.545](https://doi.org/10.12693/APhysPolA.130.545)

PACS/topics: 71.70.Ej, 75.10.Dg, 75.30.Gw

1. Introduction

UPd₂Al₃, discovered already more than 20 years ago [1], is still under hot debate due to exhibiting both the heavy-fermion behavior and the superconductivity coexisting with a rather large magnetic moment, of 0.85–1.5 μ_B below $T_N = 14.3$ K [2, 3]. The main point of the debate is related to the understanding of the role played by f electrons — they are localized or itinerant, or more exactly how many f electrons are localized or itinerant [2–11]. The itinerant nature of the U $5f$ electrons in UGe₂ has been arguing, basing on, apart of photoemission experiments, the relatively small ordered moment in the ferromagnetic phase in comparison to the almost full effective moment seen in the paramagnetic phase, say above 150 K [4, 10, 11].

In the theoretical band calculations the $5f$ electrons are considered as itinerant [4–11] whereas surprisingly nice reproduction of many experimental results can be obtained within the crystal-field (CEF) approach [1, 2, 12–14], i.e. treating well-defined integer number f electrons as localized. However, within the CEF approach there is presently still going on long-time discussion about the tetravalent [1, 2, 13] or trivalent [12, 14] uranium state in UPd₂Al₃. This f^2 or f^3 controversy has started just after the discovery of UPd₂Al₃. In fact, one of us (R.J.R.) from the very beginning, i.e. from 1992 [13], was claiming the U³⁺ configuration being alone against the all magnetic community. Actually at that time a use of the single-ion crystal field to conducting compounds was treated as erroneous and the crystal-field theory was largely discriminated.

For a scientific historical reason we add that in order to reconcile contrasting experimental results, in particu-

lar the inelastic-neutron-scattering (INS) experiment of Krimmel et al. from 1996 [15]. Zwicky et al. [6, 7] came out with a model with a dual nature of f electrons, in which they consider *two* f electrons as localized and the third f electron to be largely itinerant.

UGe₂ is a unique uranium compound indeed, becoming superconductor from the ferromagnetic state formed below T_c of 52 K [4]. For this intermetallic compound there is also going on a debate on the role played by f electrons [16]. Recently Spalek et al. in series of *Phys. Rev. B* publications [8, 9] claim that in UGe₂ *all* f electrons are itinerant — in fact these publications were a direct motivation for this paper and its title, in particular. When we have been preparing this publication we have learnt about studies of Troc et al. [17] with analysis of magnetic properties of UGe₂ with the $5f^2$ (U⁴⁺) configuration. A basic idea of studies of Troc et al. for searching for the crystal-field-based explanation of magnetic properties is quite similar to the main idea of studies of Radwanski undertaken almost 30 years ago for rare-earth and actinide intermetallics (later for $3d$ oxides): try to understand as much as possible by conventional physical interactions. The crystal field is surely conventional physical interaction, but often ignored in the modern solid-state physics. For instance, in a new nice textbook of Spalek devoted to the solid-state physics and magnetism the crystal field is even not mentioned [18].

In this paper we will calculate the strength of the excitations between the crystal-field states and compare them with inelastic-neutron-scattering results in order to get further support for the U³⁺ configuration in the metallic heavy-fermion superconductor UPd₂Al₃. We will provide preliminary results with the U³⁺ ion in UGe₂ showing the ground-state eigenfunction which reproduces the ordered magnetic moment and the overall temperature dependence of the specific heat including the λ -type at T_N . We will calculate the orbital and spin momenta in UGe₂.

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2. Theoretical outline

For the description of electronic and magnetic properties of a compound containing rare-earth or actinide atom (in fact, also of a $3d$ atom) we start from a detailed analysis of conventional interactions. Such analysis starts with a trial attributing the integer valency to the involved $4f/5f/3d$ atoms and subsequently we assume that the $4f/5f/3d$ ions keep their atomic-like integrity also being a part of the crystal lattice (thus we call our approach — the quantum atomistic solid-state theory (QUASST)) [19]. Being specific, we think that Er atoms, for instance, in ErNi_5 become Er^{3+} ions with the strongly-correlated $4f^{11}$ quantum system maintaining its characteristics known from the atomic physics [20]. In a crystal such ionic system experiences multipolar charge interactions and spin-dependent interactions. So, we have applied a single-ion like Hamiltonian for the ground multiplet $^4I_{9/2}$ ($J = 9/2$ in the LS coupling for the Nd^{3+} and U^{3+} ions) completed with inter-site spin interactions [12, 20, 21]:

$$H = H_{\text{CF}} + H_{f-f} = \sum \sum B_N^m O_N^m + n_{f-f} g^2 \mu_B^2 \left(-J \langle J \rangle + \frac{1}{2} \langle J \rangle^2 \right). \quad (1)$$

The first term is the crystal-field Hamiltonian written in the well-known Stevens notation (the relevant Stevens coefficients for the U^{3+} ion are: $\alpha = -6.428 \times 10^{-3}$, $\beta = -291.1 \times 10^{-6}$ and $\gamma = -37.99 \times 10^{-6}$). The second term takes into account intersite spin-dependent interactions, avoiding the double counting that produces the magnetic order below T_N (T_c). Note that the magnetic moment of the involved U ion is given as $m_U = -gJ\mu_B$ ($g = 8/11$ for the U^{3+} ion) and that the internal molecular field is given as usually $B_{\text{mol}} = n_{f-f} m_U$, with n_{f-f} being the molecular-field coefficient. The work with the ground multiplet, with 10 states instead of 364 states, makes calculations handy but still troublesome. It is justified because the higher multiplets are so high in energy that their population even in the room and ambient temperatures is negligibly small. Calculations of Troc et al. [17] show that higher states make less than 10 percents contribution to the ground-state eigenfunction — the four terms of the ground-state eigenfunction associated with the 3H_4 multiplet, shown in Table III in Ref. [17] gives 90% of the whole ground-state eigenfunction. The strong intra-atomic relativistic spin-orbit interactions assure that the total J becomes the good quantum number. Thus we think that the used simplification, i.e. the work within the ground multiplet, does not change the physics of the obtained results.

The crystal-field Hamiltonian for the hexagonal symmetry relevant for UPd_2Al_3 and UGa_2 compounds takes the form in the Stevens notation for the lowest multiplet

$$H_{\text{CF}} = B_2^0 O_2^0 + B_4^0 O_4^0 + B_6^0 O_6^0 + B_6^6 O_6^6. \quad (2)$$

Under the action of these multipolar crystal-field interactions the 10-fold degenerated $^4I_{9/2}$ multiplet is split into 5 Kramers doublets, denoted as Γ_7 , $\Gamma_8^{(1)}$, $\Gamma_8^{(2)}$, $\Gamma_9^{(1)}$

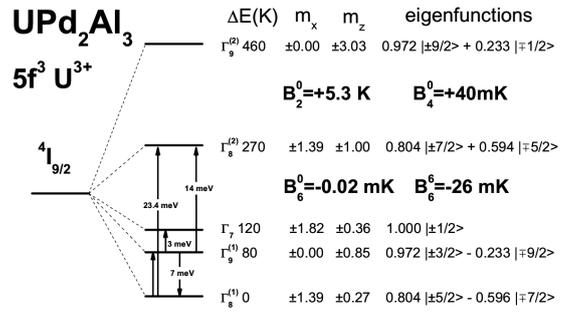


Fig. 1. Energy level scheme of the U^{3+} ion in UPd_2Al_3 derived in [12]. Arrows indicate transitions that we have attributed to excitations revealed by the inelastic-neutron-scattering experiment [15].

and $\Gamma_9^{(2)}$. These Kramers doublets become split in the magnetically ordered state. The appearance of this splitting manifests usually in the specific heat experiment as the λ -peak in the temperature dependence of the heat capacity at T_N (T_c).

3. Results and discussion

3.1. UPd_2Al_3

A milestone in the atomic-scale theoretical description of UPd_2Al_3 was the INS experiment in 1996 of Krimmel et al. [15] which revealed at 25 K the existence of the crystal-field excitations with energies of 7 and 23.4 meV. This experiment at 150 K has revealed further excitations at 3 and 14 meV at the energy-loss side and at 7 meV at the energy-gain side. Although a discrete electronic structure was theoretically found by one of us (R.J.R.) in 1992 (0, 100, 150, 350, and 600 K) by analysis of the magnetic specific heat [14] but the real observation of the discrete electronic structure by INS is extremely important as an unambiguous proof for the discrete electronic structure. According to us this discrete electronic structure is the fine electronic structure associated with the $5f^3$ state. Authors of Ref. [15] from the Steglich group have tried to describe (unsuccessfully) these excitations as due to the $5f^2$ configuration. Fulde and Zwicky [6, 7], after this experimental result, have changed their mind coming out then with a theoretical model with the dual nature of f electrons in UPd_2Al_3 on basis of the $5f^2$ configuration. In their theoretical considerations they use only two (of all 9 levels of the lowest 3H_4 multiplet) lowest singlet-states, Γ_3 and Γ_4 , separated by 7 meV. Despite of 12 years they did not develop further the dual model for UPd_2Al_3 to account for its magnetism. This 7 meV has been taken in order to fit to the energy value revealed in INS experiment of Krimmel et al. [15].

We have interpreted the INS excitations as related to the energy level scheme: 0, 7 meV (80 K), 10 meV (116 K) and 23.4 meV (271 K) and we have ascribed this scheme to the f^3 (U^{3+}) scheme. We have described this fine electronic structure by a set of CEF parameters of the

hexagonal symmetry (z quantization axis is taken along c -axis of the hexagonal structure): $B_2^0 = +5.3$ K, $B_4^0 = +40$ mK, $B_6^0 = -0.02$ mK and $B_6^6 = -26$ mK which yields the $\Gamma_8^{(1)}$ ground-state doublet and excited states at 80, 120, 270, and 460 K, Fig. 1. The ground-state doublet has the form

$$\Gamma_8^{(1)} = 0.804|\pm 5/2\rangle - 0.596|\mp 7/2\rangle,$$

where only M values of the $|JM\rangle$ functions are shown.

For a deeper proving this U^{3+} energy level scheme with the attributed eigenfunctions we now calculate probabilities for the transitions between the CEF levels in the inelastic-neutron-scattering, Table I, on powder sample of UPd_2Al_3 . For it we have calculated values of J_z , J_+ and J_- (these values come out in the BIREC computer program) occurring in the INS intensity relation.

TABLE I

Calculated probabilities for the transitions between the CEF levels $|\langle \Gamma_i | J_\perp | \Gamma_j \rangle|^2$ in the inelastic-neutron-scattering on powder sample of UPd_2Al_3 . The values at the diagonal refer to intra-doublet transitions which occur without a change of the energy in the paramagnetic state and with a change of the energy in the magnetically ordered state. The off-diagonal matrix refers to inter-doublet transitions with a change of the energy.

	$\Gamma_9^{(2)}$	$\Gamma_8^{(2)}$	Γ_7	$\Gamma_9^{(1)}$	$\Gamma_8^{(1)}$
$\Gamma_9^{(2)}$ (39.6 meV)	23.22				
$\Gamma_8^{(2)}$ (23.3)	5.93	12.29			
Γ_7 (10.3)	0.87	0	17.00		
$\Gamma_9^{(1)}$ (7.0)	1.23	2.89	15.11	1.83	
$\Gamma_8^{(1)}$ (0)	0	11.88	0	10.67	9.94

The scattering law for unpolarized neutrons in the dipole approximation for the system of N non-interacting ions is given in [22]. The probability of the INS transition between two CEF levels Γ_i and Γ_j with the energies E_i and E_j depends critically on the matrix element $|\langle \Gamma_j | J_\perp | \Gamma_i \rangle|^2$ where J_\perp is the component of the total angular momentum operator perpendicular to the scattering vector Q . In the case of the polycrystalline sample the matrix element takes a form:

$$|\langle \Gamma_j | J_\perp | \Gamma_i \rangle|^2 = (2|\langle \Gamma_j | J_z | \Gamma_i \rangle|^2 + |\langle \Gamma_j | J_+ | \Gamma_i \rangle|^2 + |\langle \Gamma_j | J_- | \Gamma_i \rangle|^2)/3, \quad (3)$$

where J_+ , J_- , J_z are the total angular momentum operators. The population factor $\rho_i(T)$ is given by the Boltzmann statistics.

The calculated strengths of INS transitions are collected in Table I. One can see that from the ground state there should be only two transitions, to the first ($\Gamma_9^{(1)}$) and to the third ($\Gamma_8^{(2)}$) doublet levels — other two transitions have the zero probability to occur. This prediction is in full agreement with experimental observation at low temperatures, i.e. at 25 K in the experiment of Krimmel et al. [15]. Next, from the first excited level there should be very strong transition to the third level, with

TABLE II

Comparison of the crystal-field parameters for hexagonal intermetallics: UPd_2Al_3 [12], UGa_2 [21], $NdPd_2Al_3$ [23] and $NdNi_5$ (= $NdNi_2Ni_3$) [24]. The ‘‘CF size’’ denotes the total splitting of the lowest $^4I_{9/2}$ multiplet of the $5f^3/4f^3$ configuration.

	UPd_2Al_3	UGa_2	$NdPd_2Al_3$	$NdNi_5$
B_2^0 (K)	+5.3	+8.38	+2.48	+3.35
B_4^0 (mK)	+40	+36.25	-12.2	+14.5
B_6^0 (mK)	-0.02	-0.5	-0.26	-0.35
B_6^6 (mK)	-26	-52	+5.09	-13.5
CF size (K)	460	713	142	242

the energy of 3.4 meV. Again this theoretical prediction is in nice agreement with the experimental observation at 150 K. According to our calculations at this temperature the first excited level is already populated in 26%. So we conclude that the derived U^{3+} electronic structure, with the attributed eigenfunctions, is correct with the high certainty.

At zero temperature we have calculated the appearance of a spin gap of 2.1 meV. According to us, this splitting of the ground-state Kramers doublet $\Gamma_8^{(1)}$ in the antiferromagnetic state has been observed by a low-energy INS experiment of Sato et al. at the energy of 1.6–1.8 meV [3]. The moment experiences the internal magnetic field of 14.5 T. This value is very close to the metamagnetic field of 18 T.

Finally, in Table II we have compared the derived CEF parameters of UPd_2Al_3 with those derived for hexagonal (isostructural) compounds UGa_2 [21], $NdPd_2Al_3$ [23], and $NdNi_5$ [24]. This latter compound can be considered as $NdNi_2Ni_3$. The derived-at-present CEF parameters of these different compounds are quite close. In fact, a systematically larger total splitting for the U compounds compared to Nd compounds is expected owing to the larger radial distribution of the $5f$ shell. One may would expect the perfect similarities but we consider already these similarities as revealing the beauty of the nature at the atomic level taking into account that we work here with the electronic structure in the meV energy scale. Note that standard *ab initio*/first-principles band structure calculations provide the electronic structure in the eV energy scale, i.e. with 1000 times smaller accuracy [25]. Moreover, the obtained band results depend drastically on the used approximation, see Fig. 9 of Ref. [25]. It reveals a substantial shortage of the present *ab initio*/first-principles band-structure methods for calculations of the electronic structure at the vicinity of the Fermi level for uranium intermetallics, for UPd_2Al_3 in particular, where strong electron correlations play a cardinal role. In the used by us many-electron crystal field approach we take into account the strong electron correlations.

It is useful to recalculate CEF parameters to the Wybourne notation. The obtained CEF parameters for UPd_2Al_3 are: $B_{20} = -1145$ cm $^{-1}$, $B_{40} = -764$ cm $^{-1}$,

$B_{60} = +5.9 \text{ cm}^{-1}$ and $B_{66} = +500 \text{ cm}^{-1}$. They enable an easy calculation of the multipolar CEF strengths S_k^2 [26]. With known from the atomic physics the ion aspherities of the rank k of the multiplet $^4I_{9/2}$, given in Refs. [26, 27] as -0.4954 , -0.4904 and -1.1085 , one can calculate the second moment of the CEF splitting which can be compared with the second moment of the derived level scheme. The calculated square of the second moment with these values for aspherities gives a value of $12\,725 \text{ (cm}^{-1}\text{)}^2$ in very good agreement with a value of $12\,762 \text{ (cm}^{-1}\text{)}^2$ obtained for the second moment of the level scheme shown in Fig. 1. Such good agreement, according to Refs. [26, 27], is strong argument for the localization of $5f$ electrons.

3.2. UGe_2

UGe_2 has the orthorhombic symmetry of the space group No. 65 ($Cmmm$) [28]. Then extra terms B_2^2 , B_4^2 , B_4^4 , B_6^2 , B_6^4 will appear in the hexagonal-symmetry crystal-field Hamiltonian. Due to the low symmetry and nine parameters involved in the single-ion description the derivation of the crystal-field parameters is a hard job. We present here a preliminary set of the CEF parameters for the U^{3+} ion with the strongly-correlated $5f^3$ quantum system — it reproduces some properties of UGe_2 , the magnetic-moment value and its direction, in particular. The set of CEF parameters (z quantization axis is taken along the c -axis of the orthorhombic cell defined in Ref. [28]): $B_2^0 = +16 \text{ K}$, $B_2^2 = +8 \text{ K}$, $B_4^0 = +70 \text{ mK}$, $B_6^0 = -1.0 \text{ mK}$ and $B_6^6 = -100 \text{ mK}$ yields five doublet states at 0, 172, 442, 1092, and 1392 K with the doublet ground-state function:

$$\Gamma_{GS} = 0.800 | \pm 5/2 \rangle + 0.560 | \mp 7/2 \rangle - 0.154 | \mp 3/2 \rangle - 0.148 | \pm 1/2 \rangle. \quad (4)$$

where only M values of the $|JM\rangle$ functions are shown.

The formation of the magnetic state is shown in Fig. 2. With decreasing temperature there is increasing mag-

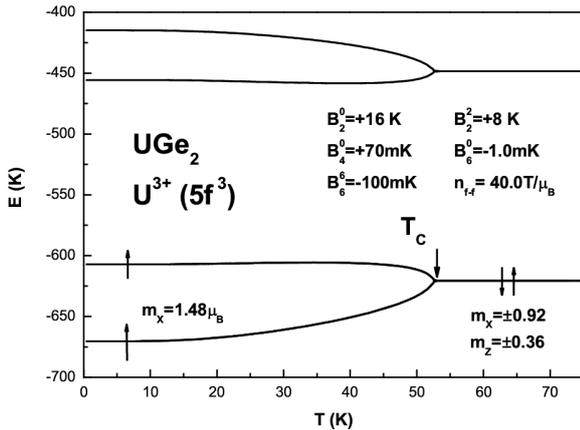


Fig. 2. The splitting of the two lowest Kramers doublets in the ferromagnetic state of UGe_2 . CEF parameters are shown as well as values of the magnetic moment in the paramagnetic and ferromagnetic states.

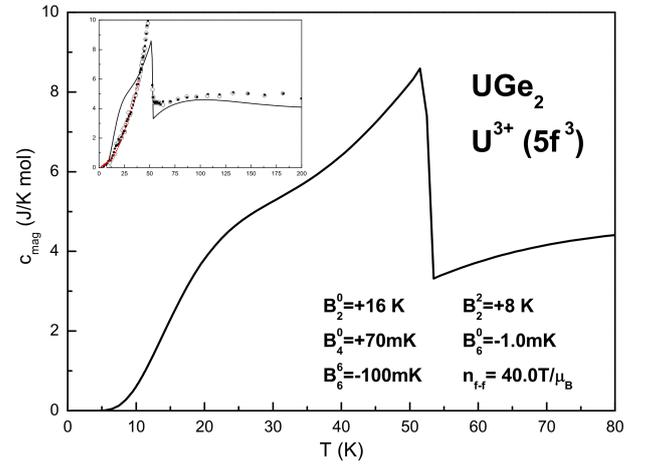


Fig. 3. Temperature dependence of the magnetic specific heat $c_{\text{mag}}(T)$ in temperature range 0–75 K with λ -type peak at the transition to the ferromagnetic state at 52 K. CEF parameters are shown. Inset shows $c_{\text{mag}}(T)$ in the wider temperature range.

netic polarization of the lowest state forming a local U moment with value of $1.48 \mu_B$ in perfect agreement with the experimental value. This moment is oriented along the a axis in agreement with the experimental observation. The moment is built from the orbital moment of $2.60 \mu_B$ ($14/11 \times J_x$ of the magnetic ground state) and the spin moment of $-1.12 \mu_B$ ($-6/11 \times J_x$). At zero temperature the moment experiences internal field of 59 T and there exists a spin gap of 5.4 meV.

The calculated temperature dependence of the magnetic specific heat $c_{\text{mag}}(T)$ (in fact it should be named as c_f as coming from f electrons), in temperature range 0–75 K, Fig. 3, reveals the λ -type peak at the transition to the ferromagnetic state at 52 K. For details of our calculations of the magnetic specific heat please see our earlier papers [12, 20, 21, 23, 24, 29, 30]. It is worth noting that we describe $c_{\text{mag}}(T)$ in the wide temperature range in the same consistent approach. A peculiar hump in the c_{mag} vs. T dependence at 20–25 K results from typical behavior of the ground Kramers-doublet seen in Fig. 2 — the excited conjugated state has the positive magnetic moment, of $0.32 \mu_B$, like the magnetic ground state. In the typical case two Kramers conjugate states have the opposite magnetic moments — such the situation occurs in UPd_2Al_3 [12]. It means that thermal excitations do not cause the reversal of the magnetic moment. We could speculate if such anomalous behavior is responsible for the formation of the ferromagnetic state as we have found the same situation in case of UGa_2 [21], which is ferromagnet with T_c of 125 K.

Temperature dependence of the paramagnetic susceptibility of UGe_2 for the $5f^3$ configuration calculated from the derived CEF parameters with taking into account the ferromagnetic interactions, of $40 \text{ T}/\mu_B$, reveals very large ferromagnetic anisotropy, by factor 2.5 at 400 K, similar to experimental observation, see Fig. 2 of Ref. [17], though our present parameters do not describe details of $\chi(T)$ so well

as that in Ref. [17]. The linear parts of the paramagnetic susceptibility above 200 K yields an effective moment of $3.30 \mu_B$ ($\parallel a$) and $3.95 \mu_B$ ($\parallel c$) being very close to the atomic effective moment of $3.62 \mu_B$. We would like to note that we have obtained this full effective moment despite of the low ordered moment of $1.48 \mu_B$. It means that an argument of Saxena et al. [4] and Pfeleiderer [10], recalled by Fujimori et al. [11], that a small ordered moment indicates the itinerant behavior of $5f$ electrons of U is erroneous.

We would like to point out that magnetic properties are calculated really self-consistently. From the experimental value of T_c we derive the strength of ferromagnetic interactions from $\chi_{CF}^{-1}(T_c)$ ($= n_{f-f}$) (obtained as $7.8 \text{ T}/\mu_B$ in the case of UGe_2), which is calculated from the set of CEF parameters. This value of n_{f-f} enters to the Hamiltonian (1) of which the free-energy vs. T is calculated for para- and ferromagnetic states. As always the lower free energy indicates the realized magnetic state.

Commenting the dual model for $5f$ electrons we say, that in any uranium intermetallics, the U atoms donors some, say three, electrons, denoted in the U atom as $7s^2$ and $6d^1$, to the conduction band. So, in $4f/5f$ intermetallics there are two electron systems, band/itinerant and localized electrons systems [19, 20]. According to us, the excitations over crystal-field states behave like quasi-particles lying at the Fermi energy of the conduction electrons. It is just the effect of strong electron correlations. In specific heat experiments we exactly see this situation. The observation of $f-f$ neutral excitations ($=$ crystal-field excitations) can be, and is indeed observed at low temperatures. Similarly at super-low temperatures the nuclear excitations are observed though obviously nuclear states are very deeply in the energy scale.

4. Conclusions

We have calculated the strength of the excitations between the crystal-field states which are in agreement with inelastic-neutron-scattering results. This agreement confirms the existence in the heavy-fermion superconductor UPd_2Al_3 of the crystal-field electronic structure originating from the U^{3+} ions with three localized f electrons forming the strongly-correlated atomic-like quantum system $5f^3$. The ionic integrity and the low-energy crystal-field electronic structure is preserved in metallic systems in the meV scale as has been postulated in the QUASST theory [12, 20][†]. The derived set of crystal-field parameters consistently describes both the paramagnetic and the

magnetic state. We provide preliminary results with the U^{3+} ion in UGe_2 showing the ground-state eigenfunction which reproduces the ordered magnetic-moment value of $1.48 \mu_B$. It is built from the orbital moment of $2.60 \mu_B$ and the opposite spin-moment of $1.12 \mu_B$. The used by us many-electron crystal-field approach takes into account the strong electron correlations. According to us heavy-fermion phenomena are related with difficulties in the removal of the on-site Kramers degeneracy undergoing at temperatures lower than, say, 4 K [32]. Our studies indicate that any physically adequate theoretical description of $4f/5f/3d$ compounds, both intermetallic and ionic, has to be made at the atomic scale and at the meV energy scale.

The present studies as well as of Troc et al. [17] prove that any theoretical description of any $4f/5f$ compound without the crystal-field states is incomplete. Simply any theoretical approach neglecting the many-electron atomic-like crystal-field states is unable to describe magnetic properties of uranium compounds, of UPd_2Al_3 and UGe_2 in particular. Thanks different studies of $5f^2$ and $5f^3$ configurations, and other models, the best description of UGe_2 can be found in the open scientific discussion.

References

- [1] A. Geibel, C. Schank, S. Thies, H. Kitazawa, C.D. Bredl, A. Bohm, M. Rau, A. Grauel, R. Caspary, R. Helfrich, U. Ahlheim, G. Weber, F. Steglich, *Z. Phys. B Condens. Matter* **84**, 1 (1991).
- [2] C. Grauel, A. Bohm, H. Fischer, C. Geibel, R. Kohler, R. Modler, C. Schank, F. Steglich, G. Weber, T. Komatsubara, N. Sato, *Phys. Rev. B* **46**, 5818 (1992).
- [3] N.K. Sato, N. Aso, K. Miyako, R. Shiina, P. Thalmeier, G. Vareloglannis, C. Geibel, F. Steglich, P. Fulde, T. Komatsubara, *Nature* **410**, 340 (2001).
- [4] S.S. Saxena, P. Agarwal, K. Ahilan, F.M. Grosche, R.K.W. Haselwimmer, M.J. Steiner, E. Pugh, I.R. Walker, S.R. Julian, P. Monthoux, G.G. Lonzarich, A. Huxley, I. Shelkin, D. Braithwaite, J. Flouquet, *Nature* **406**, 587 (2000).
- [5] A.B. Shick, W.E. Pickett, *Phys. Rev. Lett.* **86**, 300 (2001).
- [6] G. Zwicknagl, A. Yaresko, P. Fulde, *Phys. Rev. B* **68**, 052508 (2003); G. Zwicknagl, A.N. Yaresko, P. Fulde, *Phys. Rev. B* **65**, 081103(R) (2002).
- [7] G. Zwicknagl, P. Fulde, *J. Phys. Condens. Matter* **15**, S1911 (2003); P. McHale, P. Fulde, P. Thalmeier, *Phys. Rev. B* **70**, 014513 (2004).

[†]QUASST has been formulated in times when the use of the crystal-field theory for description of magnetic and electronic properties of compounds containing transition-metal $4f/5f/3d$ atoms, the existence of the localized crystal-field states in metallic compounds in particular, was regarded as erroneous, see for instance *Acta Physica* **12-13**, (2007) and *Acta Physica* **16-17** (2008). Radwanski has pointed out the substantial physical adequacy of the atomic-like localized crystal-field states, the detailed local symme-

try, importance of the intra-atomic spin-orbit coupling and the orbital magnetism both for $4f/5f$ intermetallics and $3d$ -ion compounds. Such QUASST-like approach starting from the single-ion description of the magnetocrystalline anisotropy of an intermetallic Ho_2Co_{17} [31], based on the crystal-field states, was later criticized with arguments like “a use of the single-ion crystal field to conducting compounds is erroneous”, “the usual parametrization of crystal field provides only very limited physical information” and “the crystal-field model used by Radwanski is oversimplified and the obtained agreement of calculations with experiments is accidental”.

- [8] M.M. Wysokinski, M. Abram, J. Spalek, *Phys. Rev. B* **90**, 081114(R) (2014); M.M. Wysokinski, M. Abram, J. Spalek, *Phys. Rev. B* **91**, 081108(R) (2015).
- [9] M.M. Wysokinski, J. Kaczmarczyk, J. Spalek, *Phys. Rev. B* **92**, 125135 (2015).
- [10] C. Pfeleiderer, *Rev. Mod. Phys.* **81**, 1551 (2009).
- [11] S.-I. Fujimori, T. Ohkochi, I. Kawasaki, A. Yasui, Y. Takeda, T. Okane, Y. Saitoh, A. Fujimori, H. Yamagami, Y. Haga, E. Yamamoto, Y. Onuki, *Phys. Rev. B* **91**, 174503 (2015).
- [12] R.J. Radwanski, R. Michalski, Z. Ropka, *Physica B* **276-278**, 803 (2000).
- [13] A. Bohm, A. Grauel, N. Sato, C. Schank, C. Geibel, T. Komatsubara, G. Weber, F. Steglich, *Int. J. Mod. Phys.* **7**, 34 (2003).
- [14] R.J. Radwanski, J.J.M. Franse, *Int. J. Mod. Phys.* **7**, 38 (2003).
- [15] A. Krimmel, A. Loidl, R. Eccleston, C. Geibel, F. Steglich, *J. Phys. Condens. Matter* **8**, 1677 (1996).
- [16] M. Samsel-Czekala, M. Werwinski, A. Szajek, G. Chelkowska, R. Troc, *Intermetallics* **19**, 1411 (2011).
- [17] R. Troc, Z. Gajek, A. Pikul, *Phys. Rev. B* **86**, 224403 (2012).
- [18] J. Spalek, *Introduction to Condensed Matter Physics*, PWN, Warszawa 2015 (in Polish).
- [19] R.J. Radwanski, R. Michalski, Z. Ropka, *Acta Phys. Pol. B* **31**, 3079 (2000).
- [20] R.J. Radwanski, N.H. Kim-Ngan, F.E. Kayzel, J.J.M. Franse, D. Gignoux, D. Schmitt, F.Y. Zhang, *J. Phys. Condens. Matter* **4**, 8853 (1992).
- [21] R.J. Radwanski, N.-T.H. Kim-Ngan, *J. Magn. Magn. Mater.* **140-144**, 1373 (1995).
- [22] A. Donni, A. Furrer, H. Kitazawa, M. Zolliker, *J. Phys. Condens. Matter* **9**, 5921 (1997).
- [23] R.J. Radwanski, D.M. Nalecz, Z. Ropka, *Acta Phys. Pol. A* **126**, A-32 (2014).
- [24] R.J. Radwanski, *J. Phys. Condens. Matter* **6**, L15 (1994).
- [25] A.N. Yaresko, V.N. Antonov, B.N. Harmon, *Phys. Rev. B* **68**, 214426 (2003).
- [26] J. Mulak, M. Mulak, *J. Mod. Phys.* **2**, 1373 (2011).
- [27] J. Mulak, M. Mulak, *Phys. Status Solidi B* **248**, 2159 (2011).
- [28] P. Boulet, A. Daoudi, M. Potel, H. Noel, G.M. Gross, G. Andre, F. Bouree, *J. Alloys Comp.* **247**, 104 (1997).
- [29] Z. Ropka, R. Michalski, R.J. Radwanski, *Phys. Rev. B* **63**, 172404 (2001).
- [30] R.J. Radwanski, R. Michalski, Z. Ropka, A. Blaut, *Physica B* **319**, 78 (2002).
- [31] J.J.M. Franse, F.R. de Boer, P.H. Frings, R. Gersdorf, A. Menovsky, F.A. Muller, R.J. Radwanski, S. Sinema, *Phys. Rev. B* **31**, 4347 (1985).
- [32] R.J. Radwanski, *Acta Physica* **7-8**, 1 (2007).