

Research Article

Are there crystal field levels in UPd<sub>2</sub>Al<sub>3</sub>?  
We answer, THERE ARE♣

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We claim that crystal field (CEF) levels exist in UPd<sub>2</sub>Al<sub>3</sub> in contrary to a recent claim of Hiess *et al.* (cond-mat/0411041) and Bernhoeft *et al.* (cond-mat/0411042), that there is no experimental evidence for discrete crystal field levels in this superconducting heavy-fermion antiferromagnet. We claim that excitations revealed by Krimmel *et al.* (J. Phys.: Condens. Matter **8** (1996) 1677) in inelastic-neutron-scattering (INS) studies are i) crystal-field excitations described by us within ii) the  $5f^3$  (U<sup>3+</sup>) configuration. Moreover, our  $5f^3$  (U<sup>3+</sup>) scheme, presented in Physica B **276-278** (2000) 803 and in Czech. J. Phys. **54** (2004) D295, provides a clear physical explanation for the 1.7 meV excitation (magnetic exciton) as associated to the removal of the Kramers-doublet ground state degeneracy in the antiferromagnetic state. The crystal-field theory completed by strong intra-atomic correlations and intersite spin-dependent interactions to the Quantum Atomistic Solid State Theory (QUASST), offers the meV energy scale needed for description of magnetic and electronic properties of compounds containing open-shell  $3d$ ,  $4f$ ,  $5f$  atoms. The derived set of CEF parameters for the U<sup>3+</sup> state reproduces both the INS excitations, temperature dependence of the heat capacity, large uranium magnetic moment as well as its direction.

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## I. INTRODUCTION

The description of electronic and magnetic properties of  $\text{UPd}_2\text{Al}_3$  is still under hot debate though its exotic properties has been discovered already more than 10 years ago [1, 2]. The uniqueness of  $\text{UPd}_2\text{Al}_3$  relies in the coexistence of the heavy-fermion (h-f) phenomena, the large magnetic moment of about  $0.85\text{-}1.5 \mu_B$  in the antiferromagnetic state below  $T_N = 14.3$  K and the superconductivity below 2 K. The main point of this debate is related to the understanding of the role played by  $f$  electrons.

In band calculations  $f$  electrons are considered as itinerant [3–7] whereas surprisingly nice reproduction of many experimental results can be obtained within the crystal-field (CEF) approach [2, 8–12], i.e. treating  $f$  electrons as localized. Within the CEF approach there is presently discussion about the tetravalent [2, 8, 10, 11] or trivalent [9, 12] uranium state in  $\text{UPd}_2\text{Al}_3$ . When it have seemed that the problem of the existence of the crystal field levels in  $\text{UPd}_2\text{Al}_3$  has come to the positive conclusion (Fulde and Zwignagl by years propagating the itinerant behaviour of  $f$  electrons as the origin of heavy-fermion phenomena have admitted in 2002 [13, 14]) the existence in  $\text{UPt}_3$  and  $\text{UPd}_2\text{Al}_3$  of two localized  $f$  electrons with the  $f^2$  multiplet structure and after seemed to unambiguous experimental evidence of crystal field excitations due to Krimmel *et al.* [15] recently appear two papers by Hiess *et al.* cond-mat/0411041 [16] and by Bernhoeft *et al.* cond-mat/0411042 [17] with a clear and sharp negative statement about crystal field levels in  $\text{UPd}_2\text{Al}_3$ . The appearance of these two papers is the direct motivation for this paper. Here we would like to clarify a problem, "Are there crystal-field levels in  $\text{UPd}_2\text{Al}_3$ ?" Our answer is, THERE ARE. However, as we do not think that so prominent physicists like Prof. Prof. N. Aso, N. Bernhoeft, Y. Haga, A. Hiess, Y. Koike, T. Komatsubara, G.H. Lander, N. Metoki, Y. Onuki, B. Roessli, and N.K. Sato do not know what crystal-field levels are by this paper we would like to start an open discussion about the crystal field theory and its physical adequacy. The biggest problem in this discussion is related to a fact that the crystal-field theory has within the magnetic community in last 30 years a special place - being continuously rejected from the scientific life permanently appears as a unavoids approach for explanation of properties of real compounds. The crystal-field theory is in the modern solid-state theory like a unwilling child, 75 years old already.

## II. $f$ STATES IN $\text{UPd}_2\text{Al}_3$ - A HISTORICAL OUTLINE

We are, in particular R.J. Radwanski, by years formulating the fundamental controversy in the theoretical understanding of compounds containing open-electron shells ( $3d$ ,  $4f$  and  $5f$ ), including those exhibiting heavy-fermion phenomena, as related to the treatment of  $f$  electrons (as localized or itinerant) and subsequently the formation by them a wide 2-5 eV band or low-energy discrete crystal-field states, in the meV scale. In particular, in 1992, when the itinerant  $f$  electron picture, with the  $f$  electrons put at the Fermi level, was fully dominating I (RJR) came out with a view that the heavy-fermion phenomena are related to crystal-field interactions of a strongly-correlated

odd-number electron system [18, 19] like the  $Ce^{3+}$  ( $5f^1$ ),  $Yb^{3+}$  ( $4f^{13}$ ) and  $U^{3+}$  ( $5f^3$ ) configuration (all are Kramers ions; it is worth noting, that in fact, the  $U^{3+}$  ion itself is a heavy fermion system with strongly-correlated atomic-like 89 electrons) and, for  $UPd_2Al_3$  we have explained the overall temperature dependence of the heat capacity, including the  $\lambda$ -type peak at  $T_N$ , as the crystal-field contribution of the  $U^{3+}$  ( $5f^3$ ) configuration [9]. At the same Conference Frank Steglich's group have presented CEF calculations for the  $U^{4+}$  ( $5f^2$ ) configuration [8]. The CEF approach, completed with inter-site spin-dependent interactions, we have successfully applied for description of electronic and magnetic properties of conventional rare-earth intermetallics ( $ErNi_5$ ,  $NdNi_5$ ,  $Ho_2Co_{17}$ ,  $Nd_2Fe_{14}B$ , ...) [20–22], though up to now there is strong opposition for the use of the crystal-field approach [23] to intermetallic compounds. At that time we have extended the crystal-field theory to an individualized electron model for rare-earth intermetallics, which subsequently has been extended to Quantum Atomistic Solid State Theory (QUASST) for transition-metal  $3d$ -/ $4f$ -/ $5f$ -atom containing compounds [24]. In 1996 Krimmel *et al.* have presented their results of "Search for crystal-field excitations in  $UPd_2Al_3$ " [15] undertaken with an aim to confirm their  $5f^2$  configuration. They did not manage to attribute the observed excitations to the  $5f^2$  scheme similarly as a later paper by Schenck *et al.* [11]. In a year of 2000, in our paper, Ref. 12, we took experimental results of Krimmel *et al.* as confirmation of the existence of crystal-field states in  $UPd_2Al_3$ , exactly in the same line as Krimmel *et al.* have presented own results (despite a fact, that there is the controversy about a detailed description of the observed states: are they related to  $5f^2$  or  $5f^3$  configuration, what we treat as an important but a minor problem within the localized-electron paradigm) and we have attributed the observed excitations to the strongly-correlated electron  $f^3$  scheme proposed by us already in 1992. According to us, and we thought up to now that for everybody, the INS experiment of Krimmel *et al.* [15] has revealed at 25 K 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.

Sato, Steglich *et al.* in 2001 [25, 26] and later Fulde and Zwicky [13, 14] accepting the existence of the localized levels of Krimmel *et al.* came out with a concept of a fragmentation of the  $5f$  shell into a local moment and itinerant state known at present as the dual nature of  $f$  electrons. The local  $5f^2$  subshell should manifest in the multiplet structure with the  $\Gamma_4$  ground state originating from:

$$\Gamma_{3,4} = \frac{1}{\sqrt{2}} (|J=4; J_z=+3\rangle \pm |J=4; J_z=-3\rangle)$$

split in energy, of  $\sim 7$  meV, by the crystalline electric field [14]. Although these authors did not present a detailed analysis for the confirmation of the  $5f^2$  multiplet structure they obviously think of in terms of the crystal field theory. The recalled energy splitting is the excitation observed by Krimmel *et al.* [15]. In Ref. [25] an extra low-energy excitation of 1.7 meV, at  $T = 0$  K, has been established to exist in the magnetically ordered state and

this excitation has been ascribed by authors of Ref. [25] to a magnetic exciton, with the value determined by uniaxial exchange anisotropies. In the superconducting state an ultra-low energy excitation of 0.35 meV at  $T = 0$  K appears.

In such experimental and theoretical situation we found with a really big surprise a recent claim of Hiess *et al.* and Bernhoeft *et al.* that in  $\text{UPd}_2\text{Al}_3$  there is no evidence for CEF levels. Below we cite in the full extension their statements:

Hiess *et al.* [16] - in Resume on p. 4 write: "Work on polycrystalline material at the ISIS spallation source by Krimmel *et al.* then followed giving an overview of the inelastic response function up to 20 meV. This study gives no evidence for a discrete crystal field level scheme and ..."

Bernhoeft *et al.* [17] on p. 12: "Furthermore, the fundamental assumption of strongly localised 5f levels is difficult to reconcile with the lack of observation of CEF levels in the paramagnetic state [here citation to Krimmel *et al.*, Ref. [15]] and the success of ab-initio band-structure calculations using the delocalised LSDA approach to reproduce experimental Fermi surface areas, as measured by dHvA effect, which often are taken as an indication that the 5f levels are largely delocalised." (here is citation to Knopfle *et al.*, Ref. [5] and to Inada *et al.*, Ref. [7].)

Thus, according to authors of the cond-mat/0411041 paper: A. Hiess, N. Bernhoeft, N. Metoki, G.H. Lander, B. Roessli, N.K. Sato, N. Aso, Y. Haga, Y. Koike, T. Komatsubara, and Y. Onuki, and the cond-mat/0411042 paper: N. Bernhoeft, A. Hiess, N. Metoki, G.H. Lander and B. Roessli, there is no evidence for discrete crystal field levels in  $\text{UPd}_2\text{Al}_3$ . We claim that such a sharp negative statement is completely wrong. It is also, as we have shown above, in the sharp contrast with the interpretation of Krimmel *et al.*'s results of previously recalled researchers. We do not agree that there is "the lack of observation of CEF levels in the paramagnetic state" as the INS experiments conducted at 25 K and 150 K are both in the paramagnetic state.

Such opinion of prominent physicists introduces an enormous chaos in the general understanding of the magnetism and the electronic structure of open-shell compounds, here of  $\text{UPd}_2\text{Al}_3$ . We claim that properties of  $\text{UPd}_2\text{Al}_3$  can be only understood starting from localized states of f electrons, in a number of 2 or 3. We prefer 3 f electrons owing to the intrinsic dynamics of the Kramers system, states of which are established by the atomic physics (in particular the number of states and their many-electron atomic-like nature). The detailed electronic structure is predominantly determined by conventional interactions in a solid: the Stark-like effect by the crystalline electric field potential due to 3-dimensional array of charges in a crystal acting on the aspherical incomplete shell, and the Zeeman-like effect due to spin-dependent interactions of the incomplete-shell spin (atomic-like moment) with self-consistently induced spin surroundings. These states can become broaden in energy by different interactions (lowering of the local symmetry, thermal expansion, appearance of a few inequivalent sites, lattice imperfection, surface effects and other solid-state effects). Obviously, we should not think that discrete crys-

tal field states mean that they are extremely thin lines. 3 or even 10 meV broad lines are still of the crystal-field origin. Underlying by us by many, many years the importance of the crystal field we have treated as an opposite view to the overwhelmed band structure view yielding the spreading of the f-electron spectrum by 2-5 eV.

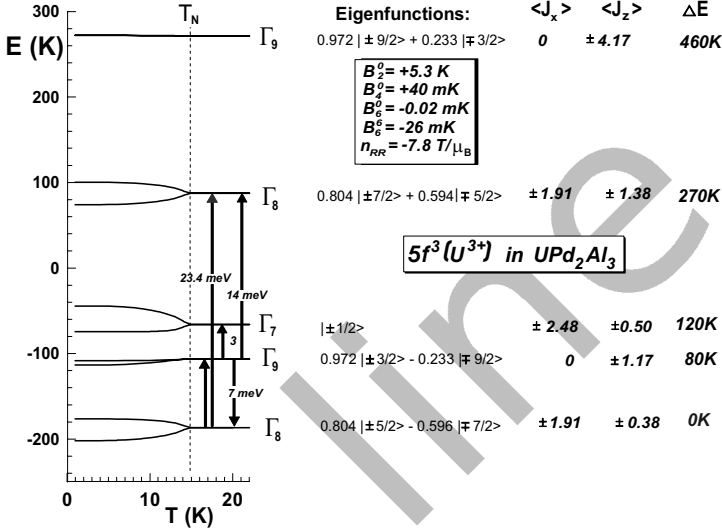


FIG. 1: Energy level scheme of the  $U^{3+}$  ion in  $UPd_2Al_3$  taken from our papers [12, 27]. Arrows indicate transitions which we have attributed to excitations revealed by inelastic-neutron-scattering experiments of Krimmel *et al.* [15].

In Ref. [12] (and recently in Ref. [27]; a direct motivation for this paper was a claim [13, 14, 25] for the  $5f^2$  configuration) we have interpreted the excitations of Krimmel *et al.* as related to the energy level scheme: 0, 7 meV (81 K), 10 meV (116 K) and 23.4 meV (271 K) and we have ascribed this scheme to the  $f^3$  ( $U^{3+}$ ) scheme. The fine electronic structure of the  $f^3$  ( $U^{3+}$  consists of five Kramers doublets split by multipolar charge interactions (CEF interactions). The fine electronic structure originates from the lowest multiplet  $^4I_{9/2}$ , higher multiplets are at least 0.2 eV above and do not affect practically the ground-multiplet properties. These 5 doublets are further split in the antiferromagnetic state, i.e. below  $T_N$  of 14 K in case of  $UPd_2Al_3$  as is shown in Fig. 1. A derived set of CEF parameters of the hexagonal symmetry:  $B_2^0 = +5.3$  K,  $B_4^0 = +40$  mK,  $B_6^0 = -0.02$  mK and  $B_6^6 = -26$  mK yields states at 81 K, 120 K, 270 K and 460 K, energies of which are in perfect agreement with the experimentally observed excitations. The highest state was not observed in INS experiment. This electronic structure accounts also surprisingly well for the overall temperature dependence of the heat capacity, the substantial

uranium magnetic moment and its direction if we make use of a single-ion like Hamiltonian for the ground multiplet  $J=9/2$  [12]:

$$H = H_{CF} + H_{f-f} = \sum_n \sum B_n^m O_n^m + n_{RR} g^2 \mu_B^2 \left( -J \langle J \rangle + \frac{1}{2} \langle J \rangle^2 \right)$$

The first term is the crystal-field Hamiltonian. The second term takes into account intersite spin-dependent interactions ( $n_{RR}$  - molecular field coefficient,  $g=11/8$  - Lande factor) that produce the magnetic order below  $T_N$  what is seen in Fig. 1 as the appearance of the splitting of the Kramers doublets and in experiment as the  $\lambda$ -peak in the heat capacity at  $T_N$ . In Fig. 2 we have shown temperature dependence of the splitting energy between two conjugate Kramers ground state. Surprisingly, both the value of the energy and its temperature dependence is in close agreement to a low-energy excitation of 1.7 meV at  $T=0$  K observed by Sato *et al.* [25] which has been attributed by them to a magnetic exciton. The agreement is very remarkable taking into account that Fig. 2 has been made by the simple subtraction of two lowest states shown in our published papers [12, 27]. Thus, we are convinced that the  $5f^3(U^{3+})$  scheme provides a clear physical explanation for the 1.7 meV excitation (magnetic exciton) - this excitation is associated to the removal of the Kramers-doublet ground state degeneracy in the antiferromagnetic state.

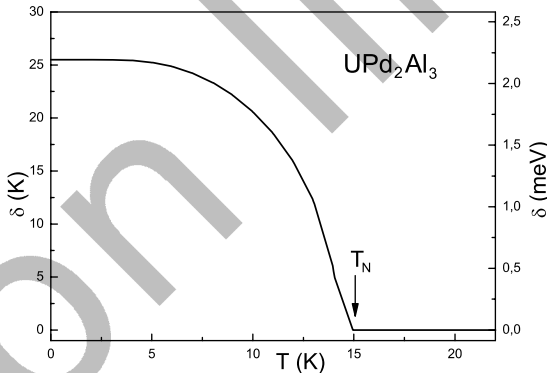


FIG. 2: Temperature dependence of the excitation energy  $\delta$  to the Kramers conjugate state of the  $f^3 (U^{3+})$  configuration in  $UPd_2Al_3$ . This energy is obtained as the difference between energies of two lowest states seen in Fig. 1, taken from Ref. [27]. Both the value of the energy and its temperature dependence is in close agreement to a low-energy excitation of 1.7 meV at  $T = 0$  K observed by Sato *et al.* [25] and attributed by them to a magnetic exciton.

### III. CRYSTAL-FIELD THEORY - SOME REMARKS

A strange scientific climate about the crystal-field theory in the modern solid-state paradigm comes from a widely-spread view within the magnetic community that it does not have the proper theoretical justification. An oversimplified point charge model is treated as an essence of the crystal-field

theory. An indication in some cases that the point charge model is not sufficient to account for the crystal-field splittings was taken as the conclusive proof for the incorrectness of the crystal-field concept at all. According to us the theoretical background for the crystal-field theory is the atomic construction of matter. Simply, atoms constituting a solid preserve much of their atomic properties. One can say that the atomic-like integrity is preserved, after giving up partly or fully some electrons, and then the atomic identity serves as the good quantum number of the electron system. We are quite satisfied that the point-charge model provides the proper variation the ground states going on from one to another  $3d/4f/5f$  atom. Different ionic states we consider as different states of the atom, though it is better instead of the ionic state to say about the electron configurations and their different contributions to magnetic, electronic, spectroscopic and optical properties. For instance, in metallic  $\text{ErNi}_5$  there exists  $4f^{11}$  electron configuration, often written as the  $\text{Er}^{3+}$  ion, that is found to be predominantly responsible for the magnetism and the electronic structure of the whole compound [20–22]; the other electrons of Er and Ni are responsible for the metallic behavior. We point out the multipolar character of the electric potential in a solid. It is very fortunate situation when a solid, with milliard of milliard of atoms, can be described with the single electronic structure. It is true, that the crystal-field theory being itself a single-ion theory cannot describe a solid with collective interactions. For this reason we came out with the Quantum Atomistic Solid State Theory and completed the crystal-field theory with strong intra-atomic correlations and intersite spin-dependent interactions. By pointing out the importance of the CEF theory we would like to put attention to the fundamental importance of the atomic physics (Hund's rules, spin-orbit coupling, ...) and local single-ion effects. It is worth remind that the source of a collective phenomenon, the magnetism of a solid, are atoms constituting this solid. Properties of these potentially-active atoms (open-shell atoms) are determined by local surroundings and local symmetry. Subsequently, these atomic moments, with spin and orbital parts, enter to the collective game in a solid.

Another wrong conviction about the crystal-field theory is that it was exploited already completely. In order to shown that this thinking is wrong we turn the reader's attention that the crystal-field approach used within the rare-earth and actinide community ( $4f$  and  $5f$  systems) fundamentally differs from that used within the  $3d$  community. The  $4f/5f$  community works with  $J$  as the good quantum number whereas the  $3d$  community "quenches" the orbital moment and works with only the spin  $S$ . Our description of a  $3d$ -atom compound like  $\text{FeBr}_2$  and  $\text{LaCoO}_3$  one can find in Refs [28] and [29]. In case of the strongly-correlated crystal-field approach we work with many-electron states of the whole  $4f^n, 5f^n, 3d^n$  configuration in contrary to single-electron states used in  $3d$  magnetism and LDA, LSDA, and many other so-called *ab initio* approaches. Technically, strong correlations are put within the CEF theory, and in QUASST, by application of two Hund's rules. The *ab initio* calculations will meet with the CEF (QUASST) theory in the evaluation of the detailed charge distribution within the unit cell and after taking into ac-

count strong intra-atomic correlations among electrons of incomplete shells and the spin-orbit coupling in order to reproduce the CEF conditions (two Hund's rules, also the third one for rare-earths and actinides).

We would like to mention that we are fully aware that used by us the Russell-Saunders LS coupling can show some shortages in case of actinides related to the growing importance of the j-j coupling. We are aware of many other physical problems which we could not mention here due to the length problem - finally we mention only that we can reverse scientific problem in the solid state physics and use  $4f/5f/3d$  compounds as a laboratory for the atomic physics for study  $3d/4f/5f$  atoms in extremal electric and magnetic fields. In the solid-state physics we study the lowest part of the atomic structure but extremely exactly.

We call for an open and honest scientific discussion on the strongly-correlated crystal-field theory being convinced that it is the fundamental ingredient of the modern solid-state paradigm. There is a hope that such discussion can proceed in Phys. Rev. B where the Hiess *et al.* (cond-mat/0411041) and Bernhoeft *et al.* (cond-mat/0411042) have submitted their papers and after being printed we intend to comment them.

#### IV. CONCLUSIONS

We claim that excitations revealed by Krimmel *et al.* (J. Phys.: Condens. Matter **8** (1996) 1677) in inelastic-neutron-scattering (INS) studies of superconducting heavy-fermion antiferromagnet  $UPd_2Al_3$  are i) crystal-field (CEF) excitations described by us within ii) the  $5f^3$  ( $U^{3+}$ ) configuration, in contrary to a recent claim of Hiess *et al.* (cond-mat/0411041) and Bernhoeft *et al.* (cond-mat/0411042), that there is no experimental evidence for discrete crystal-field levels. Moreover, our  $U^{3+}$  scheme, presented in Refs [12] and [27], provides a clear physical explanation for the 1.7 meV excitation (a magnetic exciton) as associated to the removal of the Kramers-doublet ground state degeneracy in the antiferromagnetic state. The crystal-field view completed by strong intra-atomic correlations and intersite spin-dependent interactions to the Quantum Atomistic Solid State Theory (QUASST), offers the meV energy scale needed for description of magnetic and electronic properties of compounds containing open-shell  $3d$ ,  $4f$ ,  $5f$  atoms. The derived set of CEF parameters for the  $U^{3+}$  state in  $UPd_2Al_3$  reproduces both spectroscopic and thermodynamical properties and a substantial uranium magnetic moment as well as its direction. In metallic  $UPd_2Al_3$  three localized f electrons coexist with conduction electrons originating from outer  $7s^2/6d$  electrons of uranium as well as from Pd and Al outer electrons [27]. Our studies point out the necessity of discussion of electronic and magnetic properties on the atomic scale, where the local symmetry, crystal field and strong electron correlations, of the intra-atomic and inter-site origin, play the fundamental role in the formation of the low-energy discrete energy spectrum. Our localized view is supported by results of Fujimori *et al.* [30] who found that the resonant photoemission spectrum of  $UPd_2Al_3$  reflects the single site effect of uranium sites. We are convinced that strongly-correlated crystal-field theory is a fundamental



ingredient of the modern solid-state paradigm.

We do not claim to (re-)invent crystal-field theory but we advocate for its high physical adequacy as a basic ingredient of the modern solid-state paradigm. We call for an open and honest scientific discussion on the strongly-correlated crystal-field theory as the fundamental ingredient of the modern solid-state paradigm. We understand our scientific work as continuation of works of J. H. Van Vleck, H. A. Kramers, K. W. H. Stevens and many, many others started in 1929 by Hans Bethe.

♣Dedicated to H. Bethe, J. H. Van Vleck and K. W. H. Stevens on the 75<sup>th</sup> anniversary of the crystal-field theory.

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- [23] let mention only that in May 2004 the Polish Government Scientific Council undertook a decision, with the help of Polish theoreticians, that the use of the CEF approach to metallic magnetic compounds, even conventional rare-earths, is erroneous. More information: H. Szymczak, J. Klamut, A. M. Oles and J. Sznajd. About this inquisition decision the Chairman of SCES-04 Conference, Prof. H. von Lohneysen has been informed in July 2004 with a kind request to inform the whole Intern. Advisory Board about such curious interference of the high-level administration to the scientific life and to undertake the relevant steps.
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