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NOVEL STATES IN QUANTUM MATTER

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Abstract. Understanding the collective electronic properties of emergent materials remains a great challenge to condensed-matter theory. The quantum mechanical ground states are determined by subtle compromises between varoius interactions. The resulting high sensitivity with respect to external fields will allow for new functionalities and novel technical applications. The present paper discusses typical emergent phenomena including the formation of heavy fermions in Kondo lattices, quantum phase transitions and quantum criticality as well as orbital-selective localization transitions.

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1. INTRODUCTION

Quantum materials are a topic at the forefront of research in condensed matter physics. Important examples are transition metal oxides, metals containing lanthanide or actinide atoms and organic conductors. At low temperatures, these materials exhibit novel phenomena like metal-to-insulator transitions, heavy fermions, unconventional superconductivity and unusual magnetism. As the properties of quantum materials are highly sensitive with respect to changes in external parameters like pressure, magnetic and electric fields they are important candidates for technological applications. The electronic properties of these materials, however, are very difficult to predict despite complete knowledge of the underlying interactions.

The electronic structure problem for a solid is one of many interacting fermions (electrons) moving in a lattice. The properties of these systems are generally determined by the interplay of two types of influences as schematically summarized in Fig. 1. First, there is a tendency towards delocalization which is a consequence of the fact that wave functions at neighboring lattice sites overlap. It favors the formation of a conventional band spectrum and one-electron Bloch states in which the itinerant electrons are distributed throughout the entire crystal. On the other hand, the electrons interact via the Coulomb interaction which leads to localization. The Coulomb repulsion necessarily restricts the hopping of an electron from one site to another. The reason is that a second electron feels a strong repulsion at a given atomic site when another electron is already present. As a consequence, the hopping probability will depend on whether the final site is occupied or empty and therefore introduce correlations.

A strong Coulomb interaction between two electrons at the same lattice site would favor the formation of local magnetic moments which can be observed in the magnetic susceptibility. Interacting electrons in a lattice experience both trends and the relative importance of both determines the electronic properties of a given material (for textbooks see e.g.[1, 2]).

The general theoretical problem has proved too difficult for an exact analysis. Approximate solutions can be found only in limiting cases.

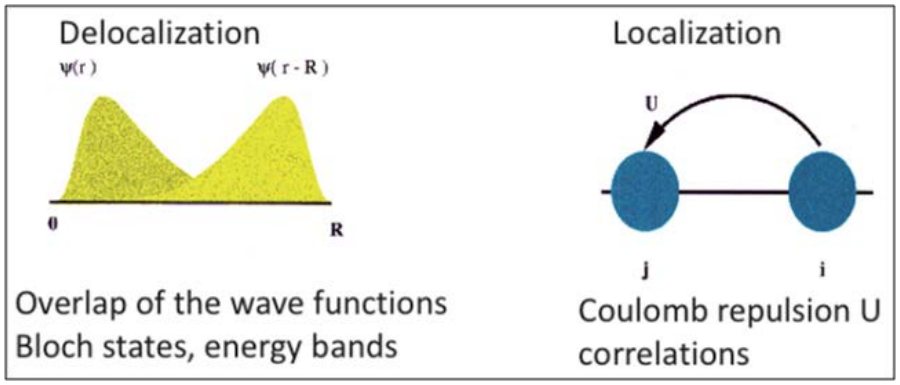


Fig. 1: Electrons in a crystal experience both a trend towards delocalization through the formation of extended Bloch states and a trend towards localization resulting from the local Coulomb repulsion.

2. HISTORIC REVIEW

The traditional electron theory of metals which is based on the Sommerfeld-Bethe model [3] emphasizes the aspect of delocalization. Starting from the picture of free electrons the model suceessfully describes a great variety of metallic properties. The ground state of the non-interacting spin1/2 fermions is obtained by successively filling the single-particle states with the lowest energies in accordance with the Pauli principle. The characteristic feature is the Fermi surface in momentum space separating occupied and empty states. The chemical potantial defines the characteristic energy, the Fermi energy E_F which is of the order of 10 eV in metals. The low-energy excitations are particle-hole excitations where electrons are promoted across the Fermi surface. For low temperatures the model predicts a linear variation with temperature of the electronic contribution to the specific heat $C \approx \gamma T + \ldots$ with a proportionality factor γ of order $1 mJ / moleK^2$ and a temperature independent magnetic susceptibility reflecting the non-magnetic character of the ground state. The success of this theory which completely neglects electron-electron interactions is at first glance rather surprising since one knows that the Coulomb interaction between the electrons must produce correlations.

The “free” electron picture was justified by Landau in his famous theory of Fermi liquids [4] which provides a highly useful conceptual framework for describing interacting fermi systems. The fundamental conceptual basis is the notion of “adiabatic continuity” which should prevail if the interaction is gradually turned on [5]. As states of simular symmetry do not cross during this adiabatic process there should result a one-to-one

correspondence between the well-known free fermion states and their unknown complex counterparts in the interacting system. In particular, the character of the ground states are the same, i. e., filled Fermi seas. The low-energy excitations can be described to good approximation in terms of a single-particle spectrum. This important fact is a direct consequence of the Pauli principle. The single-particle excitations or "quasi-particles" are characterized by momentum and spin and obey Fermi statistics. They can be visualized as composite objects consisting of the bare particle (electron) and some kind of polarization cloud which results from the Coulomb correlations. The many-body effects modify the energy dispersion relation and the interactions of the quasi-particles. The one-to-one correspondence implies in general that the interactions do not change the volume enclosed by the Fermi surface. This statement is commonly referred to as Luttinger's theorem [6]. Today, Landau's Fermi liquid theory is the "standard model" for the modern electron theory of metals.

Density Functional Theory (DFT) introduced by Hohenberg, Kohn and Sham [7, 8] provides an efficient and rather accurate method to explicitly construct effective potentials required for material specific calculations. The fundamental quantity of the theory is the (inhomogeneous) electron density in the ground state which is to be determined selfconsistently. Working with densities which depend on a single spatial variable instead of the full many-body wave function of an N particle system results in a tremendous simplification which makes electronic structure calculations possible for rather complex materials. The theory allows for parameter-free calculations of the ground state properties [9]. The method yields ficticious energy eigenvalues which are usually very good approximations to the physical energy levels. For a review see [10].

The quasiparticles successfully describe the low-energy excitations of metals in their normal state. David Bohm and David Pines [11] realized that in interactions can lead to collective oscillations of the electron gas, the plasmons.

The break-down of the Landau Fermi liquid theory at the transition from the normal into the superfluid or superconducting state can be described by introducing the concept of broken symmetry[12]. According to this ansatz, phase transitions occur via symmetry reduction which is described in terms of an order parameter. The latter characterizes the appearance of long-range correlations - the pair-correlations in superconductors. Similar examples are the formation of a Charge Density Wave (CDW) or a Spin Density Wave (SDW).

The framework described above consistently and successfuly describes the behavior of "ordinary" metals. In the spirit of Landau, the starting point is the picture of non-interacting electrons moving in effective potentials which also account for electron-electron interaction effects. The DFT provides a highly sophisticated scheme for explicitly constructing the effective potentials. The electron states provide the basis for understanding the collective dynamics which affects the response to external perturbations and phase transitions where long-range correlations are accounted for by symmetry-breaking.

3. STRONG LOCAL CORRELATIONS

The framework as described in the preceding section was seriously challenged by an unexpected series of discoveries starting from 1976. Important examples are the Heavy Fermion (HF) superconductivity in lanthanide and actinide compounds, the Fractional Quantum Hall Effect (FQHE), superconductivity in cuprates, colossal magnetoresistence, Quantum Criticality (see e. g. [1] and references therein). The materials have in common that their low-energy properties are determined by strong interactions among the electrons. The interactions, however, cannot be accounted for by effective potentials as in DFT nor by considering collective excitations or by symmetry breaking. The consequences of electron interactions which cannot be captured by the standard framework are termed “correlation effects”. Electron correlations lead to novel, often highly complex ground states and anomalies in numerous observables.

Electron correlations are strongly evident in the cuprate superconductors which may have transition temperatures ranging up to 100K. The superconducting systems are obtained by doping a small number of electrons or holes into a parent compound whose insulating state is a direct consequence of electron correlations. The key is the number of valence electrons in the unit cell. The independent electron picture for the ground state has the following implication for periodic crystals: For N electrons, we have to fill the N/2 lowest lying band states with two electrons. We immediately see that with an odd number of (valence) electrons in the unit cell, there must be a partially filled band. As a consequence, band theory predicts materials with an odd number of (valence) electrons per unit cell to be metallic.

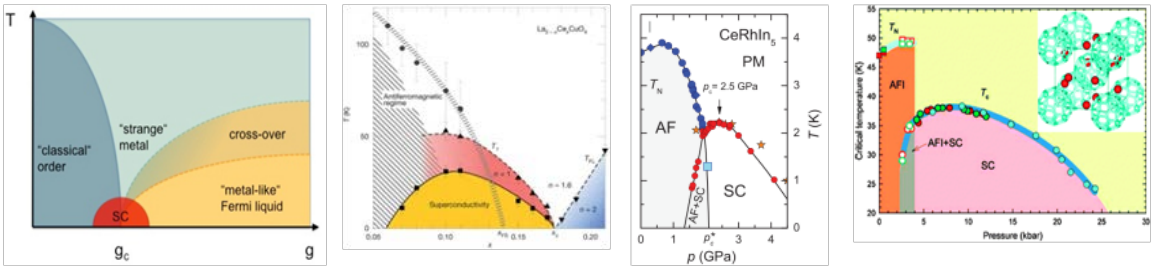


Fig. 2: Generic phase diagram of materials with strongly correlated electrons From left to right: Schematic phase diagram denoting the various low-temperature phases. Experimental phase diagrams of the cuprate superconductor $La_{2-x}Ce_xCuO_4$, the heavy fermion system $CeRhIn_5$, and the doped fulleride Cs_3C_{60} (from [35]).

This very general rule derived from independent electron band theory fails e. g. in the undoped parent material La_2CuO_4 of the cuprate superconductor $La_{2-x}Sr_xCuO_4$. Although there is an odd number of electrons in the unit cell containing two La-, four O- and one Cu-atom in $3d^9 4s^2$ configuration the material is an insulator. The counting argument refers to the chemical unit cell. Breaking translational symmetry by a (magnetic) superstructure can lead to a (magnetic) supercell with an even number of electrons. This, however, can be ruled out for the antiferromagnet La_2CuO_4 which remains an insulator also in the translational invariant state above the Néel temperature. We therefore conclude that the insulating ground state of La_2CuO_4 is a consequence of the strong electron-electron interactions. The correlation-driven metal-to-

insulator transition is called Mott transition, materials like La_2CuO_4 are called Mott insulators[13, 14].

The failure of independent electron theory in the case of Mott insulators highlights the necessity to incorporate electron correlations into electronic structure calculations. This can be achieved technically by the LDA+U method where the Coulomb repulsion U is introduced in an ad-hoc fashion. A more sophisticated treatment of strong local correlations is provided by the Dynamical Mean Field Theory (DMFT) [15, 16].

The Mott insulating state, however, has an extremely high degeneracy resulting from the spins of the localized electrons. This degeneracy is usually lifted by forming "classical" long-range magnetic order. Reducing the distance between the lattice sites by application of pressure leads to an increase in the energy due to delocalization. Many Mott systems can hence be driven through a an insulator-to-metal transition to a metallic phase by changes in a non-thermal parameter like pressure, composition etc. Fig. 2 displays the generic phase diagram which schematically describes the behavior of a wide variety of strongly correlated fermion systems including transition metal oxides, organic conductors, and heavy fermion materials.

4. HEAVY FERMIONS IN KONDO LATTICES

In emergent materials, the correlation effects, i. e., the deviations from the anticipated behavior of independent electrons result from partially filled inner d- or f-shells. From a theorist's point of view, these materials lie at the intersection of a large number of long-standing problems in the physics of metals. We immediately face the fundamental question which picture provides the better starting point for theoretical models, a delocalized description in terms of energy bands or a localized representation which properly accounts for the atomic properties. This question turns out to be ill-posed in the case of lanthanide-based heavy fermion compounds where the character of the 4f electrons seems to change with temperature, pressure and magnetic field. Some of the best known examples are the Ce-based compounds $CeCu_2Si_2$, $CeRu_2Si_2$, $CeTIn_5$ with T=Co, Rh, Ir as well as the Yb-systems $YbRh_2Si_2$ and $YbIr_2Si_2$. For recent reviews see e. g. [17–19] and references therein.

The 4f electrons are rather well localized, the corresponding charges are more or less confined within spheres of the ionic radii. As a consequence, the f states are expected to preserve their atomic character. The strong Coulomb interaction at each atomic site is rather strong and can lead to a variety of magnetic effects. The novel phenomena we encounter in these systems is the appearance of a new energy scale $k_B T^* \approx 1meV$ which result from the strong correlations within the f-shells. As a consequence of the strong correlations, it is not possible to describe the influence of the f-states in terms of a unique simple model. In the high-temperature regime, the degrees of freedom associated with the partially filled f-shells are conveniently modelled by local magnetic moments which weakly interact with the conduction electrons. The f-electrons can be considered as part of the ion cores in viewing the band structures of heavy fermion compounds. The number of quasiparticles which determines the volume enclosed by the Fermi surface is given by the number of conduction electrons and, concomittantly, equals the nominal

chemical valence. The Fermi surface is “small”. At low temperatures, the local spin-degrees of freedom at the f-sites are strongly coupled to the conduction electrons forming (local) singlets with them. The specific heat varies approximately linearly with temperature (that is, $C \approx \gamma T + \ldots$), and the magnetic susceptibilty, χ, approaches a Pauli-like form, becoming almost independent of temperature. The values of the coefficients are of the order of $1J/moleK^2$ and consequently several order of magnitude larger than those of ordinary metals which are of the order of $1mJ/moleK^2$. Due to the large Sommerfeld coefficient of the electronic specific heat which can be written in terms of an effective mass m^* of the quasiparticles these materials have been termed heavy fermion compounds. The magnetic susceptibility χ, is enhanced by a factor of comparable magnitude. These findings suggest that Heavy Fermions compounds form Fermi liquids at sufficiently low temperatures. Many well-known relations derived for independent electrons are satisfied provided the huge effective mass of the quasiparticles is properly accounted for. This is achieved by replacing the Fermi energy E_F which is typically of order 10 eV in a conventional metal by the characteristic temperature T^* which is of order 1meV. The central assumption is that the low-energy excitations reflected in the low-temperature properties are long-lived fermionic quasiparticles with predominantly f-character. The f-degrees of freedom are modelled by itinerant ”heavy” quasiparticles which form narrow bands in the vicinity of the Fermi surface. The number of quasiparticles which determines the volume of the Fermi surface includes the f-states, the Fermi surface is “large”. Comparing the high and the low temperature regime we see that the f-degrees of freedom seem to change their character which is a consequence of the strong correlations[20]. The latter lead to a clear deviation from Luttinger’s theorem according to which the volume enclosed by the Fermi surface should not change.

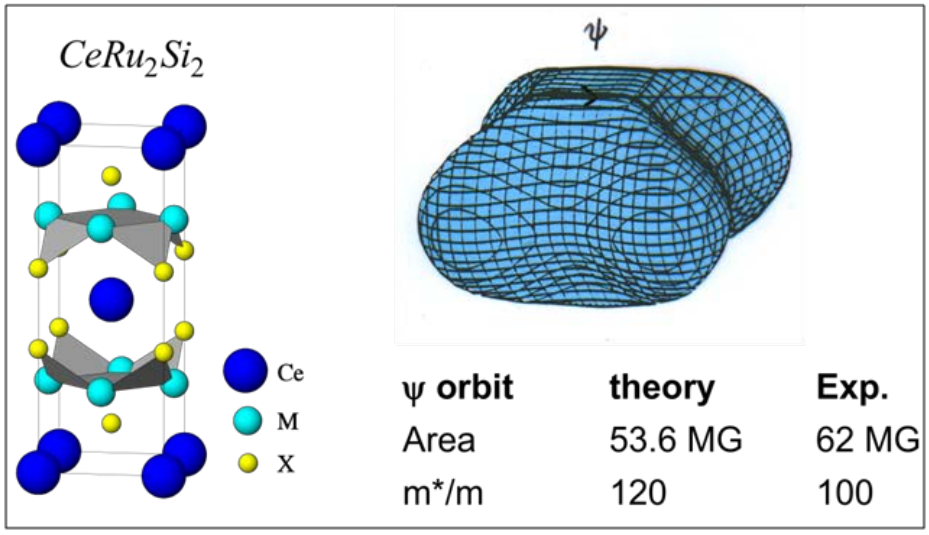


Fig. 3: Heavy quasiparticles in $CeRu_2Si_2$: Comparison of theory [21] and experiment [22]. The heavy quasiparticles explain the high specific heat ar low temperatures, i. e., they exhaust the low-energy excitations.

The hypothesis has been convincingly corroborated by experiment[21] as can be seen from Fig. 3. The theoretically predicted quasiparticle dispersions both for the high temperature

state with localized f-states with a “small” Fermi surface as well as for the itinerant f-derived heavy quasiparticles with their “large” Fermi surface were confirmed by deHaas-vanAlphen [22]and Angle Resolved Photo Electron Spectroscopy(ARPES)[23]. The calculations adopted the Renormalized Band (RB) method which is a concept for a theoretical description of the Fermi liquid state. It merges ab-initio-band structure calculations and phenomenological considerations in the spirit of Landau [24] . The central idea is to account for the correlations by introducing a small number of phenomenological parameters which are determined by fitting to appropriate experiments. For heavy fermion systems it is sufficient to adjust a single parameter - the averaged effective mass of the quasiparticles – to obtain a detailed description of the energy dispersion for a dilute gas of quasiparticles.

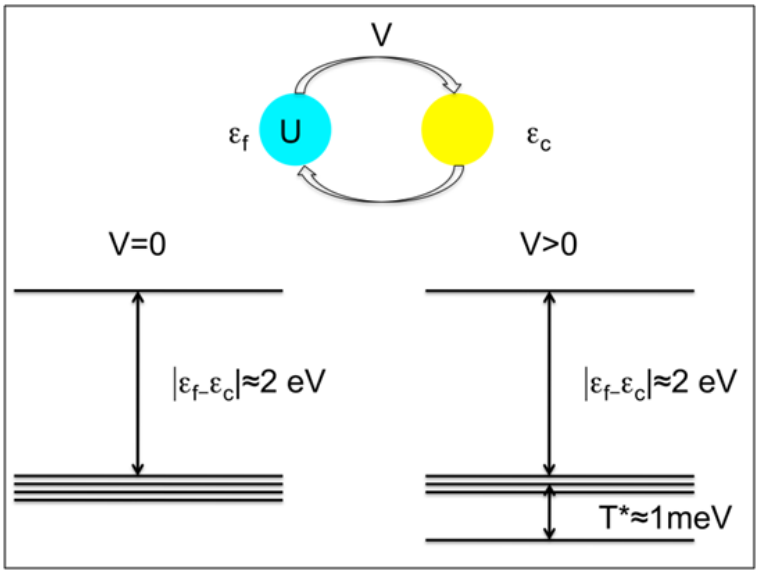


Fig. 4: Simple Molecular Model and eigenvalues of the corresponding two-electron problem for $(\varepsilon_f < \varepsilon_c (<< (V ($ in the limit of strong Coulomb repulsion $U \to \infty$. Due to the strong Coulomb repulsion among the f-electrons, the energetically most favorable states are obtained by occupying one f-state and one ligand state. This results in a fourfold degenerate ground state for vanishing hybridization. The spin degeneracy is lifted by hybridization with the doubly occulpied ligand orbital. The low-energy singlet state is lowered relative to the triplet state by a small amount of energy $k_B T^*$.

The similarities in the behavior of Ce- and Yb-based heavy-fermion systems to that of dilute magnetic alloys have led to the assumption that these systems are ”Kondo lattices” where the observed anomalous behavior can be explained in terms of periodically repeated resonant Kondo scattering[25]. The Kondo effect leads to a characteristic minimum in the variation with temperature of the electrical resistance of dilute magnetic alloys whose position does not depend on the impurity concentration. The unexpected rise of resistance at low temperatures highlights the condensed-matter analogue of quark confinement. Like the quarks, the local momemnts in a metallic host are asymptotically free at high temperatures/energies while at low temperatures/energies, they form compound quasiparticles with the conduction electrons, they become confined. The Kondo lattice model provides a microscopic explanation for the formation of a singlet groundstate and the existence of heavy quasiparticles[26]. In addition, it

explains why there is no magnetic pairbreaking associated with the presence of the f-electrons. The Kondo picture for the Ce- and Yb-based heavy-fermion compounds is supported by photoelectron and tunneling spectroscopy [23, 27].

The characteristic temperature T^* which is much smaller than typical electronic energies associated with single-particle excitations in ordinary metals as well as the non-magnetic ground state each result from many-body effects which are absent in a homogeneous electron gas. The essential physics of heavy fermion systems is already contained in the Molecular Model [28, 29] displayed in Fig. 4 where the weakly correlated conduction states of the metal are replaced by a two-fold degenerate extended ligand state such as an s-orbital. The Coulomb repulsion in this extended orbital is rather weak and can be neglected. The strong correlations are introduced by localized f-states with orbital energy $\varepsilon_f < \varepsilon_c$ which strongly repel one another. We consider only the spin degeneracy neglecting the orbital degrees of freedom. These two subsystems are coupled through a weak hybridization V. For our qualitative considerations we shall assume $U \to \infty$ and consider only states where the f-orbital is empty (f^0-configuration) and where it is singly occupied (f^1-configuration).

Let us consider the two-electron states of the model displayed in Figure 3. In the absence of hybridization (V = 0) the ground state with $E = \varepsilon_f$ is fourfold degenerate which is a direct consequence of the fact that the strong Coulomb repulsion among the f-electrons makes the f^2- configuration energetically unfavorable. The degeneracy is lifted by the weak hybridization between the ligand and the f-states. To leading order in the small ratio $V^2 / (\varepsilon_f ($ the singlet state is lowered in energy by an amount $k_B = 2V^2 / (\varepsilon_f ($ which, in turn, characterizes the low-energy excitations. For the parameter range considered here the f-valence n_f is close to unity, $n_f = 1 - V^2 / (\varepsilon_f ($

5. INSTABILITIES OF THE HEAVY FERMI LIQUID

Going back to the generic phase diagram Fig. 2, a natural question is to ask how does the magnetically ordered ground state at small values of the tuning parameter emerge from the Fermi liquid state realized at large values at T=0? The consequences of such zero temperature phase transitions represent a major unsolved challenge to our understanding of correlated matter. Although a Quantum Critical Point (QCP) strictly exists only at T=0, it can have observable consequences at elevated temperatures through quantum critical fluctuations. A prominent example is the quasi-linear variation with temperature close to quantum criticality where we encounter “strange metals”. Considering the transition, there are two possible scenarios (a) a symmetrybroken state carrying a SDW can form out of the Fermi liquid state or, alternatively, (b) the Fermi liquid state can collapse at the same critical value gc where also the magnetic order disappears (see e. g. [30, 31])

It could be demonstrated the the transition to the A phase in $CeCu_2Si_2$ occurs through a SDW transition as explained in Fig. 5. At low magnetic fields, there is a QCP of type (b) in $YbRh_2Si_2$.

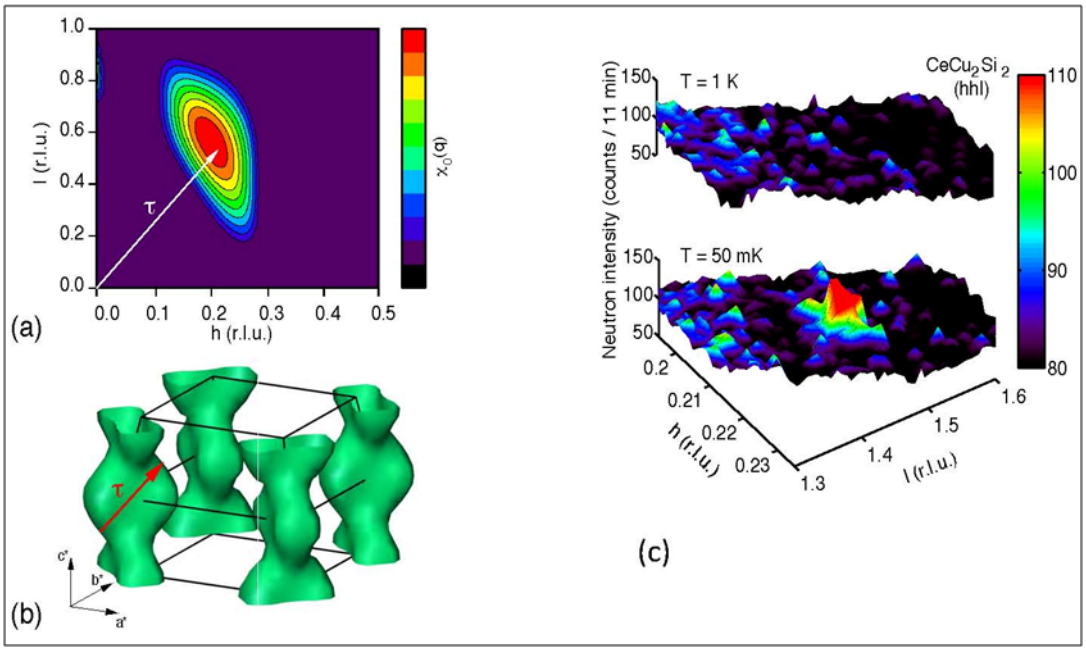


Fig. 5: Left panel: Nesting on Fermi surface of heavy quasiparticles leads to a peak in the static suceptibility at finite wave vector Q as shown by the intensity map (value increasing from dark to bright) in the reciprocal (h,h,l)- plane as calculated for the Renormalized Bands at T = 100 mK. The experimental Q at 50 mK from the right panel shows perfect agreement with the calculated maximum position. Neutron diffraction intensity in $CeCu_2Si_2$ at temperature above and below the A- phase transition temperature T_A.

6. 3=2+1: DUAL NATURE OF 5F ELECTRONS

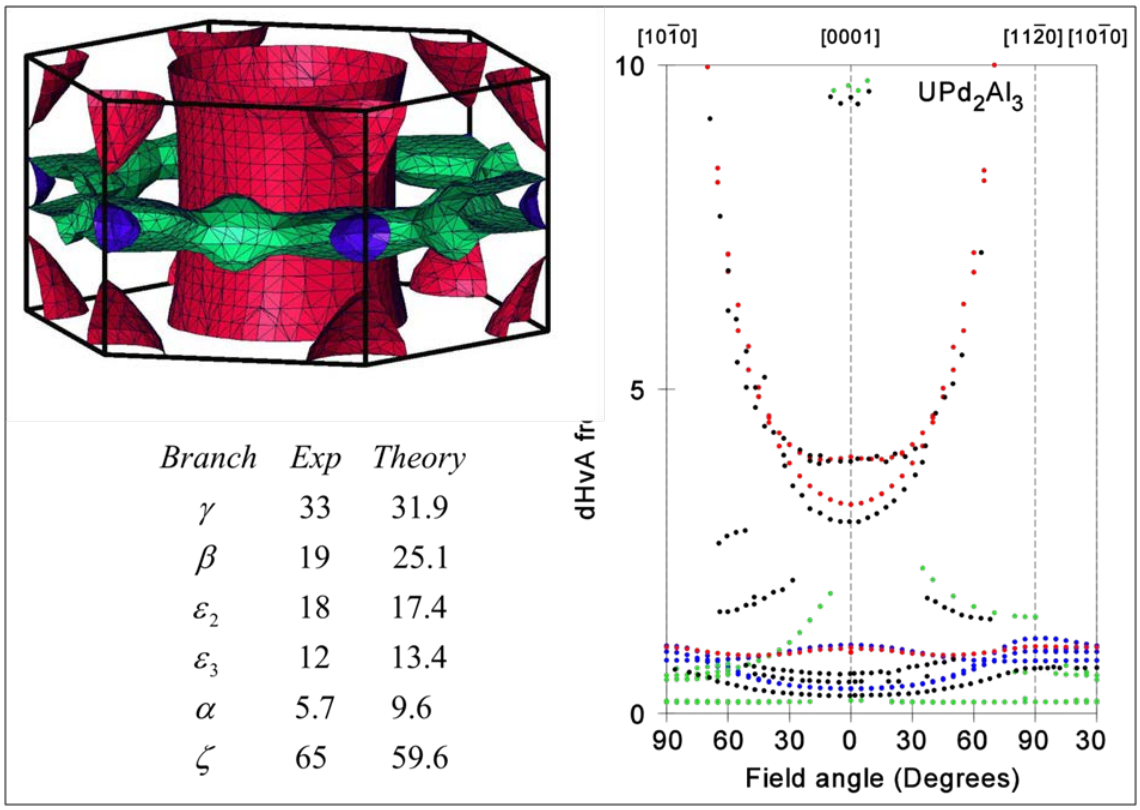
The strongly correlated 5f electrons in actinide compounds may undergo a partial, i. e., orbital-5f moments coexist with itinerant 5f band electrons. The 5f-electrons have “dual” nature[32]. Initially, the dual character has been conjectured for UPd_2Al_3 where the variation with temperature of the magnetic susceptibility points to the existence of CEF-split localized 5f states in a heavy fermion system with 5f-derived itinerant quasiparticles. Concerning the low-energy excitations it has been shown that the dual model allows for a quantitative description of the renormalized quasiparticles - the heavy fermions - in UPd_2Al_3 and UPt_3. The results for UPd_2Al_3 are summarized in Fig. 6. There is clear evidence that the presence of localized 5f states is even responsible for the attractive interaction leading to superconductivity. For a recent review of experimental facts see [17].

Before turning to a discussion of the dual model, its results and their implications we should like to add a few comments. In referring to the dual model one has to keep in mind that the latter provides an effective Hamiltonian designed exclusively for the low-energy dynamics. As such it seems appropriate for typical excitation energies below ~10 meV. In general, effective low-energy models are derived form the underlying microscopic Hamiltonians - to

borrow the language of Wilson's renormalization group - by integrating out processes at higher energies.selective localization .

The dual model conjectures that the complicated excitations of the strongly correlated 5f electron system can be approximately decomposed in the low-energy regime into coherent dispersive quasiparticle bands and incoherent local excitations. Model calculations based on Exact Diagonalization (ED) for small clusters as well as mean-field studies for infinite periodic lattices show that intra-atomic correlations as described by Hund's rules may strongly enhance anisotropies in the effective band widths and thus lead to an orbital-selective Mott transition in 5f-systems[33].

The correlation-induced orbital-selective Mott transition leads to a novel state of matter where localized atomic-like 5f electrons coexist with itinerant 5f band electrons.



Branch	*Exp*	*Theory*
γ	33	31.9
β	19	25.1
ε_2	18	17.4
ε_3	12	13.4
α	5.7	9.6
ζ	65	59.6

Fig. 6: Heavy quasiparticles in the heavy fermion compound UPd_2Al_3. The Fermi surface predicted by the Dual Model correctly reproduces the deHaas-vanAlphen data and provides a parameter-free explanation of the heavy masses (see [33,34]).

7. SUMMARY AND OUTLOOK

A remarkable variety of collective electronic phenomena have been discovered in quantum materials with strongly interacting electrons. Their high sensitivity with respect to changes in external parameters like pressure, electric and magnetic fields is considered as a resource which might eventually provide new functionalities. A comprehensive microscopic

understanding of the novel states with their intriguingly complex behavior remains a major challenge for theoretical condensed-matter physics.

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