Magnetic frustration and quantum criticality in 4*f* materials

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In contrast to magnetic insulators, geometrical frustration in magnetic intermetallic compounds is quite uncommon. Its presence can however lead to new exciting ground states and phases. It might even influence quantum critical behavior. We have focused our work on local-moment compounds with stable magnetic moments as well as on heavy-fermion systems carrying moments of more itinerant character and being located close to quantum criticality. Combining spectroscopic measurements, mainly neutron scattering, with macroscopic measurements, such as heat capacity and magnetization or susceptibility, allowed us to gain new insight in to the intriguing properties of these fascinating materials. We have obtained our results in part with well-renowned external research groups and also by carrying out experiments at several large-scale facilities.

The influence of magnetic frustration on the physical low-temperature properties of compounds is highly interesting and in the focus of current research in solid state physics. As a result, magnetic frustration can lead to exotic ground states such as spin-liquid or spin-glass states to just mention a few. While in two dimensions geometrical frustration is closely connected to triangular arrangements of magnetic moments, as e.g., in triangular or kagome lattices, in three dimensions pyrochlore or face-centered cubic (fcc) lattices are candidates for geometrical frustration. However, with their lower coordination number fcc lattices are generally less prone to frustration in comparison to kagome compounds. In the past, mainly insulating, frustrated systems have been studied. In contrast, magnetic frustration in intermetallics occurs only in very rare cases because of the more itinerant character of the magnetic moments and the conduction electrons mediating the long-range interactions. Especially interesting is the case of rare-earth based intermetallic compounds with their partially (de)localized 4f electrons. In particular, heavy-fermion compounds which easily can be tuned in their ground states, allow for a detailed study of magnetic frustration and competing interactions to tune their ground states and hence the influence on quantum critical behavior. An important issue in our recent studies is the effect of geometrical frustration on the magnetism in holmium based intermetallic materials, but also cerium and ytterbium based heavy-fermion compounds which are located in the vicinity to a quantum critical point, i.e., a continuous $T = 0$ phase transition. As a continuation of our previous work on CePdAl [1, 2, 3, 4] we now performed a detailed characterization of magnetically frustrated $HoInCu_4$ [5]. In addition, we investigated $YbNi_4P_2$ [6] and CeCoSi [7], but also continued our studies on $CeCu_{6-x}Au_x$ [8, 9, 10, 11] and $CeCu_2Si_2$ [12]. We addressed the above-mentioned issues by using microscopic as well as macroscopic techniques including elastic and inelastic neutron scattering, heat capacity and magnetic susceptibility measurements and also collaborated in time-resolved THz spectroscopy.

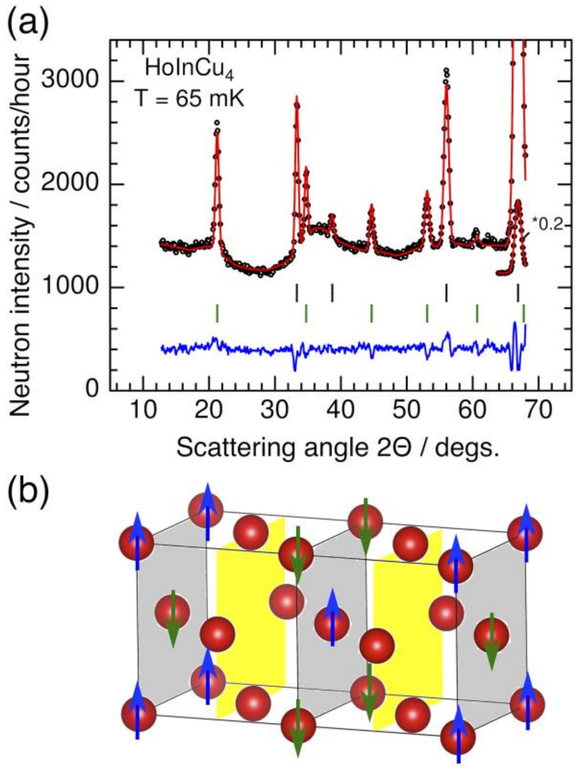

Fig. 1: (a) Neutron powder diffraction pattern of $HoInCu_4$ recorded at lowest temperature of $T = 65$ mK. Data points (black circles) and the fit to the data (solid red line) are displayed together with the deviation between fit and data (blue line) and the positions of nuclear and magnetic peaks (black and green vertical bars). Data taken on E2 at HZB. (b) Schematic view of the partially ordered magnetic structure of $HoInCu_4$ (only Ho atoms are shown) with the disordered Ho planes (in yellow) [5].

Magnetic frustration in cubic $HoInCu_4$ [5]

In the intermetallic, cubic compound $HoInCu_4$ all holmium atoms reside on an fcc lattice leading to partial magnetic frustration with a ground state where only half of the Ho moments exhibits long-range antiferromagnetic order below the quite low ordering temperature T_N = 0.76 K as evidenced by our neutron scattering experiments [5]. Rietveld refinement of our neutron powder diffraction pattern displayed in Fig. 1(a) confirms the cubic crystal structure and reveals a partially ordered type-III antiferromagnetic order as shown in Fig. 1(b). Planes of long-range ordered Ho 4f moments are separated by planes of frustrated, disordered Ho moments. From the intensities of the magnetic Bragg peaks the ordered magnetic moment is determined to 4.6 μ_B per Ho atom and is given by the ground state of the crystalline electric field (CEF) levels of the 4f electrons. Combining our inelastic neutron scattering experiments on $HoInCu_4$ with heat capacity and magnetic susceptibility allowed us to get a consistent set of CEF parameters [5]. All these results agree well with each other. The good description of the experimental data can be seen, e.g., in the CEF contribution to the magnetic heat capacity as visible in Fig. 2. Furthermore, the small ordered Ho 4f moment is also corroborated by the heat capacity measurements (cf. Fig. 2) which indicate an anomaly at the ordering temperature of largely reduced size and a Schottky anomaly at very low temperatures given by the splitting of the nuclear Ho moments in the magnetic field of the ordered 4f moments. The tail in the

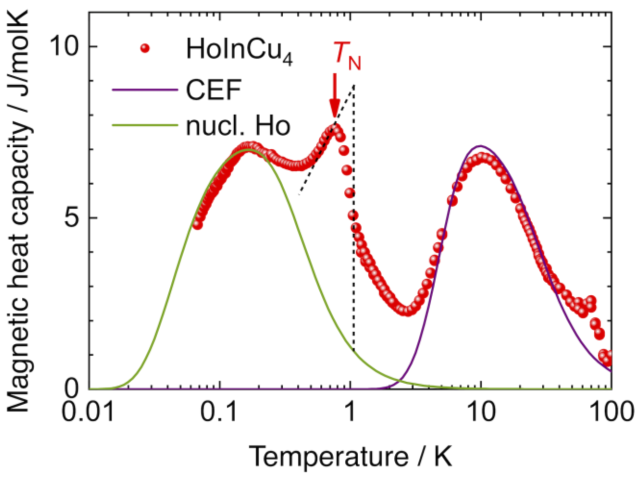

Fig. 2: Magnetic heat capacity of $HoInCu_4$ versus temperature. Solid lines indicate the CEF contribution to the heat capacity (purple line) as well as the Schottky anomaly due to the splitting of the nuclear magnetic moments of Ho (green line). An idealized mean-field anomaly at T_N is denoted by the dotted line [5].

magnetic heat capacity above T_N is a consequence of strong magnetic fluctuations due to the frustration in $HoInCu_4$. We obtained direct evidence for frustration persisting down to lowest temperatures inside the ordered state through single crystal neutron diffraction which revealed a strong diffuse magnetic signal well below T_N. Replacing In by Cd results in a complete breakdown of magnetic frustration in $HoCdCu_4$. Antiferromagnetic order sets in at a much higher temperature T_N = 7 K. In consequence, we observed in our neutron diffraction experiments a fully ordered type-II antiferromagnetic structure in $HoCdCu_4$ with an ordered 4f moment of 9.5 μ_B per Ho atom [5]. No traces of frustration have been detected in $HoCdCu_4$. As a result of our theoretical calculations, the density of states for the itinerant electron bands seems to act as a tuning parameter for the ratio between nearest-neighbor and next-nearest-neighbor interactions and thus determines the degree of magnetic frustration [5]. In the future, we will continue our work on $HoInCu_4$ to look for an influence of frustration on the spin-wave excitations, but we will also study other 4f-based intermetallics with an fcc arrangement of magnetic moments.

Characteristic energy scales in $YbNi_4P_2$ [6] and CeCoSi [7]

Special structural motifs, e.g., geometrical frustration or low-dimensional coordination, and competing interactions are also quite important in determining the properties of heavy-fermion compounds. Even in well-known $CeCu_2Si_2$ the body-centered tetragonal crystal structure can give rise to geometrical frustration. We focused on $YbNi_4P_2$, a rare case of a heavy-fermion ferromagnet with a very low ordering temperature T_C = 0.15 K (i.e., close to a ferromagnetic quantum critical point) and a quasi-1D arrangement of the Yb atoms and on antiferromagnetically ordered CeCoSi with Ce double layers stacked along the tetragonal *c*-axis suggesting reduced dimensionality. In both compounds we studied the characteristic energy scales, namely CEF and spin-wave excitations, by inelastic powder neutron scattering.

Based on experimental data from inelastic neutron scattering, heat capacity, susceptibility and NMR measurements, we determined the CEF level scheme of $YbNi_4P_2$ [6]. Despite its tetragonal crystal structure, the local site symmetry of the Yb ions is orthorhombic, increasing the relevant CEF parameters to nine. A large basal plane anisotropy is found by powder NMR measurements which probe the local symmetry. Our analysis yields a CEF level scheme with excitation energies of 8.5, 12.5 and roughly 30 meV. The ground-state wave function is dominated by the 5/2 state. Additional calculations using density-functional theory confirm the large basal plane anisotropy observed experimentally [6]. Since the first excited CEF state is well separated from the ground state, the low-temperature behavior of $YbNi_4P_2$ will be entirely determined by the ground state properties. It will be highly interesting to search for spin-wave excitations at very low temperatures in order to understand this peculiar compound close to ferromagnetic quantum criticality.

The strongly correlated electron compound CeCoSi exhibits a quite unusual temperature-pressure phase diagram: at ambient pressure it orders antiferromagnetically below T_N = 8.8 K, while the application of hydrostatic pressure induces a new magnetically ordered phase with an exceptionally high transition temperature of ~40 K at 1.5 GPa. We studied the magnetic properties and the pressure-induced magnetic phase of CeCoSi by means of neutron diffraction and inelastic neutron scattering (INS) complemented by heat-capacity measurements [7]. At ambient pressure we found a simple commensurate antiferromagnetic structure in CeCoSi with a highly reduced ordered moment of 0.37 μ_B/Ce. Specific heat as well as low-energy INS indicate a significant gap in the low-energy magnon excitation spectrum in the antiferromagnetic phase, with all CEF levels located above 10 meV as seen in Fig. 3. Hydrostatic pressure gradually shifts the energy of the magnon band from around 2.5 meV towards higher energies up to 4 meV. The temperature variation of the magnon intensity measured at 1.5 GPa

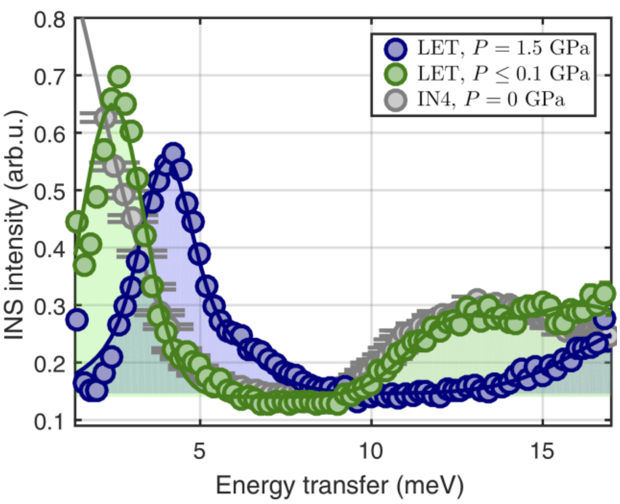

Fig. 3: Inelastic neutron scattering (INS) signal as function of energy transfer in CeCoSi at different hydrostatic pressures. Data taken on LET at ISIS and IN4 at ILL [7].

follows an order-parameter dependence and is consistent with the phase diagram. In addition, the CEF excitations are also drastically modified under pressure. Single-crystal neutron scattering measurements are currently in progress to get further insight into the spin dynamics of this interesting rare-earth compound.

Studies on other quantum critical compounds [8, 9, 10, 11, 12]

We were also involved into time-resolved Terahertz spectroscopy measurements on $CeCu_{6-x}Au_x$ and $YbRh_2Si_2$, which allowed us not only to extract information on the CEF excitations, but also on the dynamics of the Kondo state [8, 9, 10]. These measurements yielded evidence that the Kondo temperature remains finite at the quantum critical point, while the Kondo weight vanishes. Furthermore, through detailed elastic neutron scattering on $CeCu_{6-x}Au_x$ we could demonstrate that the unusual quantum criticality in $CeCu_{6-x}Au_x$ is not due to structural fluctuations around the transition to a monoclinic low-temperature structure, but solely determined by magnetic fluctuations [11]. Finally, in inelastic neutron measurements on $CeCu_2Si_2$ we found that the spin excitations in superconducting samples closely resemble the magnons in magnetically ordered samples [12], a behavior quite often observed, e.g., in high-Tc cuprate or pnictide superconductors. This strongly suggests the idea of a common, magnetic driving mechanism for the unconventional superconductivity in these compounds.

External Cooperation Partners

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References

[1] *Signature of frustrated moments in quantum critical $CePd_{1-x}Ni_xAl$, A. Sakai, S. Lucas, P. Gegenwart, O. Stockert, H. v. Loehneysen, and V. Fritsch, Phys. Rev. B* ***94*** *(2016) 220405,* doi.org/10.1103/PhysRevB.94.220405

[2] *Evolution of the partially frustrated magnetic order in $CePd_{1-x}Ni_xAl$, Z. Huesges, S. Lucas, S. Wunderlich, F. Yokaichiya, K. Prokes, K. Schmalzl, M.-H. Lemee-Cailleau, B. Pedersen, V. Fritsch, H. v. Loehneysen, and O. Stockert, Phys. Rev. B* ***96*** *(2017) 144405,* doi.org/10.1103/PhysRevB.96.144405

[3] *Entropy Evolution in the Magnetic Phases of Partially Frustrated CePdAl, S. Lucas, K. Grube, C.-L. Huang, A. Sakai, S. Wunderlich, E. L. Green, J. Wosnitza, V. Fritsch, P. Gegenwart, O. Stockert, and H. v. Loehneysen, Phys. Rev. Lett.* ***118*** *(2017) 107204,* doi.org/10.1103/PhysRevLett.118.107204

[4] *Multidimensional entropy landscape of quantum criticality, K. Grube, S. Zaum, O. Stockert, Q. Si, and H. v. Loehneysen, Nature Physics* ***13****, (2017) 742,* doi.org/10.1038/nphys4113

[5]* *Magnetic frustration in a metallic fcc lattice, O. Stockert, J.-U. Hoffmann, M. Mühlbauer, A. Senyshyn, M. M. Koza, A. A. Tsirlin, F. M. Wolf, S. Bachus, P. Gegenwart, R. Movshovich, S. Bobev, and V. Fritsch, Phys. Rev. Research* ***2*** *(2020) 013183,* doi.org/10.1103/PhysRevResearch.2.013183

[6]* *Analysis of the crystal electric field parameters of $YbNi_4P_2$, Z. Huesges, K. Kliemt, C. Krellner, R. Sarkar, H.-H. Klauß, C. Geibel, M. Rotter, P. Novak, J. Kunes, and O. Stockert, New. J. Phys.* ***20*** *(2018) 073021,* doi.org/10.1088/1367-2630/aace35

[7]* *Gradual pressure-induced enhancement of magnon excitations in CeCoSi, S. E. Nikitin, D. G. Franco, J. Kwon, R. Bewley, A. Podlesnyak, A. Hoser, M. M. Koza, C. Geibel, and O. Stockert, Phys. Rev. B* ***101*** *(2020) 214426,* doi.org/10.1103/PhysRevB.101.214426

[8]* *Time-resolved collapse and revival of the Kondo state near a quantum phase transition, C. Wetli, S. Pal, J. Kroha, K. Kliemt, C. Krellner, O. Stockert, H. v. Loehneysen, and M. Fiebig, Nature Physics* ***14*** *(2018) 1103,* doi.org/10.1038/s41567-018-0228-3

[9]* *Fermi Volume Evolution and Crystal-Field Excitations in Heavy-Fermion Compounds Probed by Time-Domain Terahertz Spectroscopy, S. Pal, C. Wetli, F. Zamani, O. Stockert, H. v. Loehneysen, M. Fiebig, and J. Kroha, Phys. Rev. Lett.* ***122*** *(2019) 096401,* doi.org/10.1103/PhysRevLett.122.096401

[10]* *Terahertz conductivity of heavy-fermion systems from time-resolved spectroscopy***,** *C.-J. Yang, S. Pal, F. Zamani, K. Kliemt, C. Krellner, O. Stockert, H. v. Löhneysen, and M. Fiebig, Phys. Rev. Research* ***2*** *(2020) 033296,* doi.org/10.1103/PhysRevResearch.2.033296

[11]* *Magnetic and structural quantum phase transitions in $CeCu_{6-x}Au_x$ are independent, K. Grube, L. Pintschovius, F. Weber, J.-P. Castellan, S. Zaum, S. Kuntz, P. Schweiss, O. Stockert, S. Bachus, Y. Shimura, V. Fritsch, and H. v. Löhneysen, Phys. Rev. Lett.* ***121*** *(2018) 087203,* doi.org/10.1103/PhysRevLett.121.087203

[12]* *Robustness of magnons near the quantum critical point in the heavy-fermion superconductor $CeCu_2Si_2$, Z. Huesges, K. Schmalzl, C. Geibel, M. Brando, F. Steglich, and O. Stockert, Phys. Rev. B* ***98*** *(2018) 134425,* doi.org/10.1103/PhysRevB.98.134425

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