Superconductivity, Quantum Phase Transitions and Fermi Surface of Strongly Correlated *f*-Electron Systems

This research project is built upon our long-standing, successful collaboration with Prof. D. Aoki. We have investigated two families of Ce-based heavy-fermion materials, in which quantum criticality can be induced by doping. To this end, several new samples were grown. Some of these studies yielded interesting preliminary results, and will be continued, also in high magnetic fields. In addition, we have finished the analysis and discussion of some of the previously obtained data. In conjunction with new results, this gave rise to two articles submitted to high-level journals. One of them was recently published [1].

Quantum critical points (QCPs), i.e., continuous phase transitions at zero temperature, play a key role in the physics of heavy-fermion (HF) compounds and other materials. Recent theoretical attempts to classify QCPs in HF systems rely on the knowledge of whether the f electrons are itinerant or localized on both sides of a QCP, i.e. whether or not they contribute to the Fermi surface (FS). This can be achieved by comparing experimentally established FS topology with the results of band structure calculations performed for both itinerant and localized f electrons. Magnetic quantum oscillations, such as the de Haas-van Alphen (dHvA) effect, are the most direct way to establish the FS topology of a metal.

CeRhIn₅ is one of the best-studied HF materials. This tetragonal antiferromagnetic (AFM) compound with $T_{\rm N} = 3.8$ K can be tuned to a QCP by pressure, chemical substitution, and magnetic field. Several dHvA experiments evidence that the f electrons of CeRhIn₅ are localized at ambient pressure, although some of the theoretically predicted dHvA frequencies were not experimentally observed. As the critical pressure for the suppression of antiferromagnetism, $P_{\rm c} = 2.3$ GPa, is reached, all dHvA frequencies observed at $P < P_{c}$ change discontinuously, signaling an abrupt FS reconstruction as a consequence of the *f*-electron delocalization. In addition, the effective masses diverge at $P_{\rm c}$. A similar discontinuous change of the dHvA frequencies was observed upon substituting Rh by Co in CeRh_{1-x}Co_xIn₅. However, the FS reconstruction does not occur at the critical concentration $x_c \approx 0.8$, where the AFM order is suppressed, but deep inside the AFM state, at $x \approx 0.4$, where the AFM order alters its character and superconductivity emerges to coexist with antiferromagnetism. Furthermore, the effective masses do not diverge here.

Another way to tune CeRhIn₅ to quantum criticality is to substitute Rh by Ir [2], as shown in Fig. 1(a). Upon doping CeRhIn₅ with Ir, the antiferromagnetic order is suppressed at a critical concentration $x_c = 0.6$ giving rise to a QCP. In addition, a commensurate magnetic order develops above the Ir concentration of about 0.3, and co-exists with the incommensurate structure up to x_c . Finally, superconductivity was observed over a wide concentration range 0.3 < x < 1.





The FSs and effective masses of the end compounds, CeRhIn₅ and CeIrIn₅, are well known. CeRhIn₅ is characterized by FSs with localized f electrons and only slightly enhanced effective masses. On the contrary, CeIrIn₅ posses Fermi surfaces with itinerant f electrons and strongly enhanced effective masses change as a function of Ir concentration is one of the important open questions, which we addressed within

this project. To this end, high quality single crystals of CeRh_{1-x}Ir_xIn₅ with several different concentrations of Ir were grown. Figure 1(b) shows the Fourier spectra of the dHvA oscillations in CeRh_{0.8}Ir_{0.2}In₅ and CeRh_{0.4}Ir_{0.6}. For both compositions, the observed dHvA frequencies and effective masses are the same as in pure CeRhIn₅. This suggests that the FSs and effective masses in CeRh_{1-x}Ir_xIn₅ do not change up to at least the critical concentration of Ir. This conclusion should, however, be treated with caution, as a more recent microprobe analysis revealed a significant difference between the real and nominal Ir concentrations in some of the samples. Therefore, new samples of CeRh1-xIrxIn5 are being grown. All the new crystals will be tested by microprobe and specific heat measurements, prior to dHvA studies, which will be performed, if necessary, also in high magnetic fields up to 36 T in Grenoble.

In pure CeRhIn₅, the analysis of our previous dHvA results in magnetic fields up to 70 T revealed that several dHvA frequencies gradually emerge at high fields as a result of magnetic breakdown. Among them is the thermodynamically important β_1 branch, which has not been observed so far. Comparison of our angular-dependent dHvA spectra with those of the non-4f compound LaRhIn₅ and with band-structure calculations evidences that the Ce 4f electrons in CeRhIn₅ remain localized over the whole field range. This rules out any FS reconstruction. either at the suggested nematic phase transition at B^* ≈ 30 T or at the putative quantum critical point at $B_{\rm c}$ \approx 50 T. Our results rather demonstrate the robustness of the FS and the localized nature of the 4f electrons in- and outside of the antiferromagnetic phase. A manuscript based on these findings was recently submitted to Physical Review Letters.

In non-magnetic CeCoIn₅ and CeIrIn₅, a QCP can be induced by doping, e.g. by Cd substitution into In sites. Introduction of Cd into CeCoIn₅ creates initially a two phase region above nominal x = 0.075, where $T_N > T_c$, followed by only antiferromagnetism for x >0.12. The phase diagram of CeIr(In_{1-x}Cd_x)₅ seems to be strikingly different. Only superconductivity was observed for $x \le 0.025$, while only a magnetic ground state was observed for $x \ge 0.075$. This ambiguity motivated us to re-examine the low-temperature properties of CeIrIn₅ doped with 5% of Cd, CeIr(In_{0.95}Cd_{0.05})₅.



Fig. 2 Superconducting transition in $CeIr(In_{0.95}Cd_{0.05})_5$ observed in specific heat (left axis) and resistivity (right axis).

As shown in Fig. 2, we observed a superconducting transition both in resistivity and specific heat. Similar to pure CeIrIn₅, the transition temperature in resistivity is much higher than that determined from specific heat measurements. On the other hand, we have not observed any signature of an AFM transition. These results allowed us to revise the previously suggested temperature-concentration phase diagram of CeIr(In_{1-x}Cd_x)₅. Furthermore, we have confirmed beyond any doubt that superconductivity does not co-exist with antiferromagnetism in Cd-doped CeIrIn₅. An article based on these results together with those of our previous neutron diffraction study of CeIr(In_{0.9}Cd_{0.1})₅ was recently published in Physical Review B [1].

Finally, several new samples of the recently discovered spin-triplet superconductor UTe_2 were grown and tested. This allowed us to further master the growing technique of this fascinating material. Consequently, high quality single crystals of UTe_2 were grown in the CEA-Grenoble. One of these crystals was chosen for the future dHvA experiments in high magnetic fields.

References

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