EUROPEAN SPALLATION SOURCE

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Assessment report on the doctoral thesis of Milan Klicpera entitled "Electron properties of the substituted cerium compounds"

Objective of research:

The main goal of this work is to better understand the microscopic origin of the unusual dynamic properties observed in several cerium-based intermetallic compounds which are attributed to magneto-elastic coupling resulting in a so-called 'vibron' state. Starting from the non-cubic parent compounds $CePd_2Al_3$ and $CeCuAl_3$ the influence of doping is investigated by a multitude of experimental methods.

Overview of thesis structure:

The manuscript consists of 108 pages plus a bibliography of 123 references. It is structured in four main chapters framed in an 'introduction' and a 'conclusions' section. The first chapter provides scientific background for the physical description of cerium-based intermetallic compounds. Though correlation effects and magnetic exchange are discussed an emphasis is on single-ion crystal electric field effects (CEF). Details on the experimental techniques used for sample preparation, characterization and investigation are provided in chapter 2. The vibrant state observed in cerium-based compounds is the topic of chapter 3, which motivates the experimental investigation reported in the main chapter 4. This chapter comprises about 60 pages and is split into two parts.

The first part deals with the properties of $CeCuAl_3$ using mainly single crystals. Though the low temperature crystalline structure is consistent with previous results, a different phase has been observed above room temperature. The electronic properties have been investigated by measurements of magnetization, specific heat and electrical resistivity all consistent with a single ion description. Various neutron diffraction experiments were performed to reveal the details of the magnetic order present below T_N = 2.5 K. The magnetic structure is described by a propagation vector of k = (0.4, 0.6, 0) and magnetic moments pointing along the (1 1 0) direction within the basal plane. This magnetic structure is different to previously reported investigations by an other group, but consistent with the anisotropy inferred from the CEF parameters. This finding is put into context with the structure observed for other $CeTX_3$ compounds indicating the magnetic structure depends both on the nearest neighbor distance and the unit cell volume. Finally, inelastic neutron scattering experiments were performed on powder sample changing the Al/Cu ratio in respect to the parent compound e.g. investigating the doping dependence of $CeCu_{(x)}Al_{(4-x)}$. For all but the parent compound no low-lying energy level has been observed but the energy levels around 10meV and 20meV remain present. Implications are discussed very briefly only.

The second part of chapter 4 reports on the other non-cubic compound $CePd_2Al_2$ investigated using single crystals. The bulk data is consistently described when the low-lying energy level is included but fails without this level related to the 'vibron' modelled. Replacing Al by Ga in polycrystalline samples $CePd_2Al_{(2-x)}Ga_{(x)}$ reveals a smooth downwards variation of the magnetic ordering temperature. On the other hand, the temperature of the structural temperature changes for x > 1 only. Using neutron diffraction a different magnetic structure has been determined for the two undoped compounds. Inelastic neutron scattering reveals the low lying energy level depends on the structure e.g. 2meV vs 5meV and only prints in the parent compound $CePd_2Al_2$.

Published papers:
The thesis builds on 12 papers published in refereed journals comprising one published in Physical Review B.

Conclusions:
In his manuscript the candidate Milan Klicpera presents an impressive breadth of experimental methods used to investigate the electronic properties of $CePd_2Al_3$ and $CeCuAl_3$ on doping. His investigations led to a manifold of new experimental results published not only in his thesis but also already on refereed journals e.g. available to the community. His findings are not only relevant for this specific class of cerium compounds but for many colleagues working on the relation of (dynamic) magnetic and structural properties. The manuscript is well written; the background information and the experimental methods are well introduced prior to presenting the experimental results. Though I would like to mention two minor shortcomings in the manuscript. First, the title is very 'generic' e.g. neither the compounds nor the experimental methods are mentioned. Second, experimental results and the discussion are presented within each individual section. Though this is adequate given the breadth of techniques used and results obtained, the reader cannot easily find the main finding inferred from the specific measurement. Here marking (in italic) the relevant sentences would be very valuable. I expect the manuscript will be available in electronic form and 'searchable', so that both topics are less critical than in previous times. I appreciate that the main findings are summarized in the 'conclusions' section, which remains a very high-level discussion only. Finally, I applaud the possibility to expose a candidate to such a breadth of experimental techniques during a single PhD thesis. Having learned to work not only with laboratory methods but also using scattering techniques gives the candidate vast possibilities for further scientific work. I recommend going forward with the PhD examination for which I would expect that the main findings are critically discussed illustrating also the limits of the presented (CEF) analysis.

Lund, Nov 15, 2015

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