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A comparison of the static magnetic field induced magnetisation densities in UPt_3 and UBe_{13}

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The static magnetic field induced magnetisation densities of two uranium based heavy-fermion systems have been determined in spin polarised neutron scattering experiments at temperatures of $T = 5$ K and $T = 10$ K, respectively. Both compounds show a significantly different magnetisation density. The various contributions are identified. In UPt_3 the polarisation of conduction electrons is important, but in contrast the magnetisation density in UBe_{13} is observed to be of a pure uranium 5f-electron character with no indication of any conduction electron contribution due to polarization by the external magnetic field. Both heavy-fermion compounds have in common a small value of the aligned 5f-electron magnetic moment which is well described by a spherically symmetric model. However, UPt_3 and UBe_{13} differ significantly in the contributions of the conduction electrons.

1. Introduction

The various known heavy-fermion systems exhibit a variety of ground states: superconducting, magnetic as well as nonsuperconducting and nonmagnetic states at lowest temperatures. All these ground states are generally believed to be caused by a coherent state of 4f- or 5f-electrons, which, while showing single-ion behaviour at elevated temperatures of ~ 100 K and above, enter into a correlated state when cooled down to temperatures below ~ 10 K. This low-temperature state has been named the heavy-fermion state, and it is evidenced by a large contribution of the f-electrons to the electronic part of the specific heat, resulting in an increase of the effective electron mass of two orders of magnitude.

While specific heat measurements and other macroscopic probes of the heavy-fermion state yield valuable information on the overall electronic properties at low temperatures it is rather difficult to isolate only the f-electron part from

the other electronic contributions and to separate the ‘heavy’ f-electrons from non f-electrons. These non f-electrons which are of interest here are those electrons which are located in the conduction band and which are close to the Fermi surface. These electrons will hereafter be referred to as conduction electrons. The conduction electrons and f-electrons are of fundamental importance in characterising the properties of the low-temperature state.

2. Experimental

In order to experimentally separate the f-electron and conduction electron contributions and to quantify their importance for determining the magnetic properties at low temperatures, detailed spin polarised neutron scattering experiments have been carried out. The magnetic field induced magnetisation densities of the two uranium based heavy-fermion superconductors UPt_3 and UBe_{13} have been determined at temperatures of $T = 5$ K and $T = 10$ K, respectively.

In these spin polarised neutron scattering experiments the flipping ratios of approximately 40 crystallographically independent Bragg reflections were obtained. Together with the knowledge of the nuclear structure factors (which were determined experimentally in a separate experiment) the magnetic structure factors could be deduced from the values of the flipping ratios. A detailed account of the experimental procedures, the data analysis and various model calculations together with their interpretations will be given elsewhere. Here attention will be focused solely on the comparison of the experimentally determined magnetisation densities as observed in UPt_3 and UBe_{13} .

UPt_3 (for a discussion of the properties of UPt_3 and UBe_{13} see refs. [1, 2]) is a supercon-

ducting heavy-fermion system ($T_c \approx 0.5$ K) which crystallises in the hexagonal Ni_3Sn structure. The position of the atoms of one of the two formula units per unit cell is shown in fig. 1a. This figure shows a cut through the nuclear unit cell at a value of $z = \frac{1}{4}$. The magnetisation density as obtained at $T = 5$ K is shown in fig. 1b for the same section. The magnetisation density map displays a complex magnetisation pattern including positive and negative magnetisation contributions.

A significant part of the magnetisation density is located on the uranium atom with a radius of approximately $1 \text{ \AA}$. This magnetisation density originates from the 5f-electrons of the uranium atom. In addition to the uranium 5f-electrons other electrons contribute to the pattern in fig.

Fig. 1. The positions of the atoms in the nuclear unit cell of the UPt_3 are shown in fig. 1a. The unit cell contains two formula units of UPt_3 with the atoms being located in planes given by $x, y, z = \frac{1}{4}$ and $x, y, z = \frac{3}{4}$. The induced magnetisation density for UPt_3 at $T = 5$ K is shown in fig. 1b for a section through the unit cell. The plane corresponds to $x, y, z = \frac{1}{4}$, and within this section three platinum atoms and one uranium atom are located. The positions of the platinum atoms are marked with dots. Around the uranium atom position the magnetisation density of 5f, 6d and 7s electrons is clearly visible. In fig. 1b the dotted lines indicate values of $-15, -7.5, -5$ and -2.5 , while the levels with values of $0, 2.5, 5, 10, 15, 20, 25, 35$ and 45 are plotted with straight lines. The units are given in $10^{-3} \mu_B/\text{\AA}^3$.

1b. Around the uranium atom three regions with a magnetisation oriented parallel to the magnetisation direction of the uranium 5f-electrons is observed. The occurrence of the maximum in the radial part of the wavefunction identifies this magnetisation density as arising from 6d-electrons of the uranium. This assignment is confirmed by the angular dependence of this magnetisation as the 6d-electron wavefunction is observed to be split due to the site symmetry of the uranium position into crystal field states. A p- or s-electron contribution is not expected to show such an angular variation. The magnetisation contribution with an opposite moment compared to the uranium 5f-electron magnetisation direction originates from the polarisation of an uranium 7s-electron.

In addition to the contributions located on the uranium atom some magnetisation density is observed around the platinum atoms. This platinum atom magnetisation is predominately positive and orientated parallel to the magnetisation direction of the uranium 5f-electrons. The distribution of the magnetisation density significantly deviates from a magnetisation pattern which is to be expected on the basis of free atom wavefunctions, thereby indicating the importance of wavefunction overlap between the platinum wavefunctions. The platinum atom magnetisation is anisotropic and located close to the hexagonal plane shown in fig. 1b.

Model calculations have been carried out and the magnetisation pattern has been described using free atom wavefunctions and taking into account the experimental observations, i.e. taking into account those wavefunctions which are experimentally observed to be of importance and allowing for overlap between platinum 5d wavefunctions. The overall magnetisation pattern could satisfactorily be reproduced and as a result of this model the various contributions could be quantified.

The magnetisation contribution of the uranium 5f-electrons could be well accounted for within a dipole approximation, which attempts to describe the magnetisation of the f-electrons as spherically symmetric. The overall aligned magnetic moment of the uranium 5f-electrons at a

temperature of $T = 5$ K and in a field of 4.6 T amounts to only 50% of the total magnetisation within the unit cell. All other contributions arise from the magnetisation of non uranium 5f-electrons. These electrons display a very anisotropic magnetisation pattern around the platinum as well as around the uranium atom.

For UPt_3 these measurements indicate the significance of non 5f-electron contributions to the magnetisation density, and it is to be expected that their importance extends also to the determination of other low-temperature properties in UPt_3 .

In order to determine whether or not these complex magnetisation distributions are a general feature of heavy-fermion systems a similar experiment was carried out on a second uranium based heavy-fermion superconductor UBe_{13} .

UBe_{13} crystallises in the cubic $NaZn_{13}$ structure with the uranium atoms being located on the lattice sites of a simple cubic lattice with half the lattice parameter of UBe_{13} . The structure determination was carried out using the instrument D15 at the ILL in Grenoble and the same single crystal was used on the diffractometer D3 for the determination of the flipping ratios of approximately 40 crystallographically independent Bragg reflections at a temperature of $T = 10$ K. From the flipping ratios and with the knowledge of the nuclear structure factors the magnetic structure factors can be obtained. The large number of magnetic structure factors obtained in this manner allowed a magnetisation density map to be constructed by Fourier inversion of the magnetic structure factors.

The projection onto one of the cubic faces (only $\frac{1}{4}$ of the total unit cell projection is shown) of the magnetisation density in UBe_{13} is shown in fig. 2b together with the projection of the nuclear density for the same proportion of the unit cell. The density maps are averaged over a cube of length $0.5\text{\AA}$ in order to minimize Fourier series termination effects.

The magnetisation density in UBe_{13} has only a single contribution which is characteristic of the uranium 5f-electron magnetisation. This magnetisation is found to be isotropic and it is well described using the dipole approximation for the

Fig. 2. The projections of the nuclear and the magnetic field induced magnetisation density are shown for UBe_{13} at a temperature of $T = 10$ K in fig. 2a and 2b, respectively. The projection was performed onto a face defined by the crystallographic (100) and (010) axis. Only $\frac{1}{4}$ of the total unit cell projection is shown for clarity. The position of the uranium atom is located at the centre of the projection, and two uranium atoms are projected down onto the same position in fig. 2a and 2b. The magnetisation density map of UBe_{13} is shown in fig. 2b and it clearly shows a magnetisation density arising from uranium 5f-electrons with no indication of any additional contributions to the magnetisation. The nuclear and the magnetic density have been averaged over a cube of side length $0.4 \text{ \AA}$ in order to minimise the effect of oscillations due to the finite number of Fourier coefficients. The level lines of fig. 2b indicate values of 10, 35, 60, 85, 110 and 135 in units of $10^{-3} \mu_B/\text{\AA}^2$.

uranium 5f-electron magnetisation. No experimental evidence is found for any additional conduction electron contribution, neither one which is located on any of the Be atoms nor a constant polarisation of the conduction electrons. The conduction electrons in free or almost free electron bands are expected to give rise to a constant background in the magnetisation density map. However, an extrapolation of the experimentally determined magnetic form factors to zero scattering vector yields a value which is in good agreement with the value obtained by using the measured susceptibility [3] to estimate the magnetisation at a temperature of $T = 10$ K and in a field of 4.6 T. Thus the good agreement of the extrapolated value to the susceptibility and also the small deviation of the magnetic form factor of low-angle reflections from the form factor calculated in the dipole approxima-

tion indicates the absence of any significant conduction electron magnetisation in UBe_{13} . Thus the observed contribution to the magnetisation density arises entirely from uranium 5f-electrons.

3. Discussion

In the literature a similar experiment has been reported by Stassis et al. [4]. A limited number of flipping ratios were determined and their temperature dependence has been established. However, the limited number of measured points [4] resulted in an incomplete data set and consequently a pure uranium 5f-electron explanation was put forward by Stassis and coworkers for the magnetisation density in UPt_3 and UBe_{13} . As the more detailed investigations reported here show, such a simplistic model is not adequate for UPt_3 ,

but it works well for the description of the magnetisation density in UPe_{13} .

It is concluded that both sets of experiments are in agreement as far as the observed data points are concerned but they differ in their interpretation. The full data set corrects the simple model as suggested by Stassis and co-workers and it illustrates the full complexity of the magnetisation density in UPt_3 . For UPe_{13} the simple uranium 5f-electron model works remarkably well, and both investigations of Stassis et al. [4] and the one reported here agree in the interpretation of the data.

4. Conclusion

The magnetic field induced magnetisation densities of the two heavy-fermion superconductors UPt_3 and UPe_{13} are presented and the various magnetisation contributions are identified. While in UPt_3 the conduction electrons are found to be of importance, the experimental evidence for UPe_{13} indicates the absence of any non uranium 5f-electron magnetisation.

In conclusion the heavy-fermion superconductors UPt_3 and UPe_{13} present a microscopic magnetic field induced magnetisation distribution which is substantially different for both compounds. While UPt_3 displays a complex magnetisation pattern with substantial contribution arising from non uranium 5f-electrons the magnetisation density in UPe_{13} is found to be only determined by the 5f-electron magnetisation of the uranium atom with no indication of any conduction electron contributions. However, both compounds show a similar behaviour of the uranium 5f-electron magnetisation at low temperatures. In UPt_3 as well as in UPe_{13} the f-electron magnetisation is well described using a spherically symmetric magnetisation model. For

both compounds the absolute magnitude of the aligned magnetic moment is small with values below $0.1\mu_B$ in a field of 4.6 T. This value has to be compared with the paramagnetic effective moment which for both compounds is of the order of $3\mu_B$ per uranium atom. However, as far as the non uranium 5f-electron magnetisation contributions are concerned the two heavy-fermion compounds differ substantially and present a very different picture concerning the role of conduction electrons. While in UPe_{13} the conduction electrons have no appreciable contribution to the magnetisation density at low temperatures their contribution amounts to $\sim 50\%$ of the total static magnetisation in UPt_3 .

Any detailed model for UPt_3 has to take into account the various conduction electron contributions which are identified in the experiment discussed here. It is to be expected that conduction electrons play an important role in UPt_3 and that band structure effects significantly determine the properties of UPt_3 at low temperatures.

In contrast, for UPe_{13} the polarisation of the conduction electrons is found to be substantially smaller than in UPt_3 and equal to zero to within the uncertainty of the experiment. As the magnetisation in UPe_{13} is of pure 5f-electron character this heavy-fermion system is the easier one to model magnetically compared to UPt_3 .

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