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Magnetic Resonance on Correlated Semimetals: the case of U$_2$Ru$_2$Sn, CeRu$_4$Sn$_6$ and FeSb$_2$

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Abstract. A comparative magnetic resonance study on correlated semiconductors is presented. Nuclear spin-lattice relaxation measurements provide an excellent method to gain local information about the gap formation in this new class of materials. In contrast to U$_2$Ru$_2$Sn ($\Delta/k_B \approx 230$ K) in CeRu$_4$Sn$_6$ the gap is slightly reduced ($\Delta/k_B \approx 200$ K) and correlations form out of a residual density of states in the gap. For FeSb$_2$ there is revived interest after classifying this system as the second Fe containing Kondo insulator beside FeSi. Surprisingly, FeSb$_2$ shows a colossal Seebeck coefficient at low temperatures. Using $^{121,123}$Sb nuclei as a local probe, our NMR/NQR investigations strongly support the gap scenario. The spin-lattice relaxation rate $^{123}(1/T_1)$ in the entire investigated temperature range 2-200 K is perfectly fitted with rectangle DOS model with narrow impurity in-gap band. These in-gap states might originate the high thermopower observed. The obtained gap value is ($\Delta/k_B \approx 473$ K).

1. Introduction
Correlated semiconductors and especially Kondo insulators (or heavy fermion semiconductors) are a subset of Kondo lattice systems, in which the lattice of magnetic f- or d- ions interact with conduction electrons giving rise to formation of a small hybridization gap at the Fermi level [1]. The signature of the gap formation is usually found in transport (resistivity, thermopower) and specific heat measurements. Strong experimental evidence often comes from magnetic resonance techniques (NMR and NQR) on various nuclei. To overcome problems in the determination of the gap due to the magnetic anisotropy and/or granularity in polycrystalline samples, this local probe is very suitable. The gap formation in these narrow band systems with high density of states at the Fermi level $N(E_F)$ leads to exotic low-temperature properties, such as a large (or gigantic) thermopower S(T) peak, which makes them promising candidates for the thermoelectric applications [2]. A record value of $|S| \sim 45$ mV/K at around 10 K has been observed recently in FeSb$_2$ [3,4], which is characterized as a strongly correlated narrow gap 3d semiconductor - the second Fe containing Kondo insulator beside FeSi. U$_2$Ru$_2$Sn (hereafter referred to as U221) was classified as the first tetragonal Kondo insulating system [5], while the tetragonal ternary stannide CeRu$_4$Sn$_6$ (hereafter referred to as Ce146) was classified by Das and Sampathkumaran [6] as a dense Kondo system with no evidence for low
temperature magnetic ordering. In this paper we present the results of comparative NMR study of the gap formation in the correlated semiconductors U$_2$Ru$_2$Sn, CeRu$_4$Sn$_6$ and FeSb$_2$.

2. Experimental

Single phase polycrystalline samples U$_2$Ru$_2$Sn, CeRu$_4$Sn$_6$ and FeSb$_2$ were prepared as described in [7], [6] and [3,4], respectively. For U$_2$Ru$_2$Sn we investigated also single crystals [8].

Field sweep $^{119}$Sn ($I = 1/2$) NMR spectra on poly- (Ce146) and single- crystalline (U221) samples were performed at different temperatures. The NMR frequency was about 33 MHz for U221 and 47 MHz for Ce146. Doing field sweep NMR a large number of spin echo experiments with small excitation width are performed as a function of field while the frequency is staying constant. The spectra in the field domain are given by plotting spin echo intensity (integration over echo in time domain) as a function of field. For tetragonal systems like U221 and Ce146 with strong anisotropy the resonance fields $\mu_0H^\|e$ and $\mu_0H^\perp e$ could be identified by either taking the sharp resonance peaks from the oriented single crystal in U221 or by fitting the typical powder pattern for Ce146 [8]. For U221 $T_1$ was determined for both directions, whereas for Ce146 $T_1$ was determined for the $\mu_0H^\perp e$ position.

Because Sb has two NMR active isotopes with spin $I = 5/2$ ($^{121}$Sb) and $I = 7/2$ ($^{123}$Sb ) and the orthorhombic structure of FeSb$_2$ field sweep NMR leads to a very complexes and extremely broadened spectra which is very difficult to analyze. In such cases NQR investigations become very suitable because here only two ($^{121}$Sb) plus three ($^{123}$Sb) distinct NQR lines appear which makes the interpretation much clearer. $^{123}$Sb NQR experiments were performed on the home-build phase-coherent pulsed NQR spectrometer in the temperature range of 2 – 200 K. $^{121,123}$Sb NQR spectra were measured using a frequency step point-by-point spin echo technique. At each frequency point the area under spin echo magnitude was integrated in the time domain and averaged over scan accumulation number, which depends on temperature. No strong anisotropy of the lines could be detected. The nuclear spin-lattice relaxation was obtained using the saturation recovery method and experiments were performed on the $^{123}$Sb NQR $\pm 3/2 \leftrightarrow \pm 5/2$ transition at 55.8 MHz.

3. $^{119}$Sn and $^{123}$Sb magnetic resonance: Spin-lattice relaxation

In order to model the spin-lattice rate in the entire temperature range we tried to fit the data by assuming a single particle excitation spectrum with a gapped density of states. As a first approach we fitted the rate with an exponential curve ($1/T_1 - \exp(-\Delta/k_BT)$, figure 1), known from Spin-Peierls systems or conventional superconductors. The striking deviation from the exponential curve observed at low temperatures has been ascribed to residual states in the gap. For U221 our data could be nicely fitted within the “V-shaped” gap model described in Ref [9] with $\Delta/k_B = 230$ K and a bandwidth of $\Delta/k_B = 2500$ K [9].

For Ce146 and FeSb$_2$ at high temperatures we also observe an activated type of behavior whereas towards lower temperatures in contrast to U221 $1/T_1$ increases again (not shown). Furthermore, for Ce146 strong field (frequency) dependence shows up. This could not be described in the rigid “V-shaped” gap model. Here we use a rectangular DOS model with “in-gap states” having a temperature and field dependency. This model was applied very successful for SmB$_6$ where $^{11}$B NMR results lock very similar to our NMR results [10]. It could be speculated that these in-gap states are responsible for the thermopower peaks observed at low temperatures for FeSb$_2$ [3,4] as well as for SmB$_6$ [11]. From the activated behavior at high temperatures we obtained the gap values of $\Delta/k_B = 473$ K for FeSb2, $\Delta/k_B = 200$ K for Ce146 and $\Delta/k_B = 230$ K for U221. A consistent description within the rectangular DOS model with narrow in-gap peak described in Ref. [10] is possible for all systems and this work is under progress.
Figure 1. Nuclear spin-lattice relaxation rate as a function of inverse temperature in \( \text{U}_2\text{Ru}_2\text{Sn} \), Ce\( \text{Ru}_4\text{Sn}_6 \) and \( \text{FeSb}_2 \). Straight lines indicate the activated type of behavior with \( 1/T_1 \propto \exp(-\Delta/k_B T) \)

4. Conclusion
A comparative magnetic resonance study on correlated semiconductors is presented. The spin-lattice relaxation measurements confirm the gap formation in \( \text{U}_2\text{Ru}_2\text{Sn} \), Ce\( \text{Ru}_4\text{Sn}_6 \) and \( \text{FeSb}_2 \) by using the local magnetic resonance probe. The values obtained by a simple activated type of behaviour above 50 K are consistent with what was determined from susceptibility, resistivity and specific heat measurements. For a consistent description of \( 1/T_1 \) in the full temperature range the implementation of residual “in gap states” is required. These states might be responsible for low temperatures anomalies found in thermopower.

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