Photoexcited electron dynamics in Kondo insulators and heavy fermions

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We have studied the photoexcited carrier relaxation dynamics in the Kondo insulator SmB₆ and the heavy fermion metal YbAgCu₄ as a function of temperature and excitation level. The dynamic response is found to be both strongly temperature dependent and nonlinear. The data are analyzed with a Rothwarf-Taylor bottleneck model, where the dynamics are governed by the presence of a narrow gap in the density of states near the Fermi level. The remarkable agreement with the model suggests that carrier relaxation in a broad class of heavy electron systems (both metals and insulators) is governed by the presence of a (weakly temperature dependent) hybridization gap.

Recent experiments on the photoexcited electron dynamics in heavy fermion metals have shown that the picosecond (ps) relaxation time of the photoinduced (PI) reflectivity increases by more than two orders of magnitude upon cooling from 300 to 10 K [1,2]. In contrast, the dynamics of their non-magnetic analogues were similar to conventional metals like Au and Ag [3], and could be described by the so-called two-temperature model (TTM) [3,4], where the ps recovery is governed by electron-phonon (e-ph) thermalization. The relaxation times in magnetic and non-magnetic compounds were quite similar at high temperatures (T) suggesting the TTM as a starting point for understanding the relaxation processes also in heavy fermions. The TTM analysis of the dynamics showed that the relaxation process in heavy fermions can be accounted for using a simple e-ph thermalization, assuming that there exists a mechanism for e-ph scattering suppression when both the initial and final electronic states lie within the peak in the density of states (DOS) at the Fermi level (E_f) [1,2]. It was argued that the small Fermi velocity compared to the sound velocity may be the origin of this suppression, because, in this case, the energy and momentum conservation law suppresses e-ph thermalization [2].

In the previous work, there were some observations that were not described by the TTM model, such as the anomalous rise-time dynamics observed in YbAgCu₄ at low T [1]. Importantly, similar T-dependent rise-time dynamics have also been observed in superconductors [5]. Furthermore, the T-dependence of the relaxation rate observed in YbAgCu₄ also closely resembles the data on superconductors, both conventional [5,6] and cuprate [6,7]. The very similar behavior of the PI carrier dynamics in superconductors and heavy electron systems suggests that the physics governing the relaxation dynamics in heavy electron compounds may need to be reconsidered. Indeed, if hybridization of the local f-moments with conduction electrons leads to the opening of a well established (indirect) hybridization gap (E_g) near E_f [8,9], similar relaxation bottleneck is expected as observed in superconductors [10]. The analogy with superconductors is straightforward for the case of Kondo insulators, where E_f lies within the gap. However, the same bottleneck physics [10] is expected to be effective even in the case of metallic heavy fermions, if E_f lies close to the hybridization gap edge (if the distance between the gap edge and E_f ≪ E_g).

Relaxation phenomena for nonequilibrium superconductors have revealed some of the most intriguing problems in condensed matter physics since the 1960’s [10]. The presence of the superconducting gap in the single particle excitation spectrum presents a relaxation bottleneck. The minimal model that describes the relaxation of the (photo)excited superconductor was formulated in 1967 by Rothwarf and Taylor [11]. Pointing out that the phonon channel should be considered when discussing relaxation processes, they described the relaxation dynamics by two coupled nonlinear-differential equations [11]. While in the low perturbation limit the equations can be linearized [12], it was shown only recently that approximate analytical solutions can be obtained for all limiting cases [13]. Analytical solutions enable comparison of the experimental data with the model, and have revealed that the Rothwarf-Taylor (RT) model can account for both the rise-time dynamics [13] as well as the superconducting state recovery as a function of excitation fluence (F) and T [1,2].

In this Letter, we present the first detailed study of the carrier relaxation dynamics in heavy electron systems as a function of excitation level. Utilizing a low repetition rate optical parametric amplifier we were able to measure the F-dependence of the reflectivity transients in the Kondo insulator SmB₆ and heavy fermion YbAgCu₄ over more than three orders of magnitude in F. Both the transient amplitude and the relaxation rate were found to be strongly F-dependent. Moreover, both observations are found to be consistent with the RT model implying that the relaxation phenomena in Kondo insulators, as well as in heavy fermion metals, are governed by a phonon bottleneck mechanism arising from the presence of the hybridization gap in the DOS at E_f. The temperature dependence of the transient amplitude and relaxation rate further supports this scenario, and the extracted values of the hybridization gap are in good agreement with the
published data. The data suggest that the hybridization gap is, to a first approximation, T-independent consistent with the periodic Anderson lattice model in the $U = 0$ limit [3].

In order to study the $F$-dependence of the dynamical response at low $T$ while minimizing the continuous heating of the probed spot due to laser excitation, we used an amplified Ti:Al$_2$O$_3$ laser system and an optical parametric amplifier operating at 250 kHz producing sub 100 fs pulses. The experiments were performed using a standard pump-probe set-up. The samples were excited at 3.0 eV with $F$ ranging from 0.5 to 600 $\mu$J/cm$^2$, while the PI changes in reflectivity were measured at a photon energy of 1.67 eV. The lowest $F$ used in this study was only about a factor of 5 higher than $F$ used in an earlier study [4], while the maximum $F$ is more than 1000 times higher. Moreover, since the repetition rate of the amplified system is 3 orders of magnitude lower than in the high repetition rate system [1], the effect of sample heating is minimized.

Figure 1(a) presents the PI reflectivity in YbAgCu$_{4}$ measured at 5 K while $F$ is varied from 0.28 to 580 $\mu$J/cm$^2$. All the reflectivity traces in panel (a) are normalized to $F$, i.e. $\Delta R/R$ is divided by $F$ in units of $\mu$J/cm$^2$. For a linear response, all the traces normalized to $F$ would lie on top of each other; this is clearly not the case. The inset shows the data taken at 0.28 $\mu$J/cm$^2$ on a linear scale. The recovery dynamics is found to be non-exponential, and the relaxation non-universal, which is to be expected in the non-linear regime. Therefore, when performing a quantitative analysis of the data, we analyze the amplitude of the transients, $A$, and the initial relaxation rate, $\tau^{-1}$, extracted by fitting the early relaxation dynamics with exponential decay (see inset to panel (a)).

At the lowest $F$, the data are consistent with the low $T$ data obtained in an earlier study [1]. The initial femtosecond rise-time ($\sim 200$ fs, resolution limited) is followed by a further increase of the signal on a ps timescale. The recovery dynamics proceeds on a 100 ps timescale. As $F$ is increased, the PI transient is changed dramatically. The second-stage risetime becomes faster and above $F \approx 4 \mu$J/cm$^2$ the rising edge is resolution limited. The decay rate is also weakly $F$-dependent up to $\approx 4 \mu$J/cm$^2$ and increases by almost 2 orders of magnitude as $F$ is further increased to the 100 $\mu$J/cm$^2$ range. Moreover, the amplitude $A$ shows linear $F$-dependence up to $F \approx 4 \mu$J/cm$^2$, while at higher fluences $A \approx \sqrt{F}$. Panel (b) shows the $T$-dependence of the reflectivity transient taken at constant $F$. The behavior is similar to earlier low $F$ data [1], revealing two orders of magnitude increase in the relaxation rate upon warming to 300 K, while $A$ gradually decreases.

Figure 2 presents $F$- and $T$-dependence of the PI reflectivity in the Kondo insulator SmB$_6$. The behavior is very similar to YbAgCu$_{4}$. The main difference is that in SmB$_6$ no anomalous two-stage risetime is observed. The fluence at which $A$ departs from $A \propto F$ is near 12 $\mu$J/cm$^2$.

Before analyzing the $F$- and $T$-dependence of the PI reflectivity transients, we briefly review the RT model and its solutions, using language appropriate for a photoexcited narrow gap semiconductor. Following photoexcitation, high energy electrons initially release their energy via $e$-$e$ and $e$-$p$ scattering. After this process, which usually proceeds on a sub-ps timescale [2], the system is characterized by excess densities of electron-hole pairs (EHP) and high frequency phonons (HFP). When an
EHP with an energy $\geq E_g$ ($E_g$ is the gap) recombines a high frequency phonon ($\omega > E_g$) is created. Since HFP can subsequently excite EHP, the relaxation is governed by the decay of the HFP population. The dynamics of EHP and HFP populations is determined by:

$$\frac{dn}{dt} = -n\gamma - Rn^2,$$

$$\frac{dN}{dt} = -\eta N/2 + Rn^2/2 - \gamma (N - N_T).$$  \hspace{1cm} (1)

Here $n$ and $N$ are the concentrations of EHP and HFPs, respectively, $\eta$ is the probability for EHP creation by HFP absorption, and $R$ the rate of electron-hole recombination with the creation of a HFP. $N_T$ is the concentration of HFPs in thermal equilibrium, and $\gamma$ their decay rate (governed either by anharmonic decay or by diffusion out of the excitation volume).

The thermal equilibrium concentrations of HFPs and EHPs ($n_T$) satisfy the detailed balance equation $Rn_T^2 = \beta N_T$. Depending on the initial conditions ($n_0$ and $N_0$, which are concentrations of EHP and HFP after photoexcitation and the initial e-e and e-ph avalanche process) and the ratio of $\gamma/\eta$, several different regimes are realized [13]. It follows from the RT analysis that the ps risetime dynamics is observed at low $F$ when photoexcitation and the initial avalanche processes lead to an excess phonon population with respect to the detailed balance condition; i.e. $Rn_0^2 < \beta N_0$. This is followed by the “thermalization” of EHP and HFP distributions leading to quasi-stationary distributions of $n_s$ and $N_s$ that satisfy [13]

$$Rn_s^2 = \beta N_s ; n_s = \frac{R}{4\eta} \sqrt{1 + \frac{16R}{\eta}} n_0 + \frac{8R}{\eta} N_0 - 1.$$  \hspace{1cm} (2)

Since the PI reflectivity (absorption) is proportional to the PI carrier density, the resulting PI reflectivity shows an initial fast rise-time ($n(t)$ reaches $n_0$) followed by a ps rise where $n(t)$ reaches $n_s$. Recovery proceeds on a much longer timescale and is governed by HFP decay.

There are several important predictions of the model that can be used to determine whether photoexcited carrier dynamics in heavy electron systems indeed follow the RT kinetics. First, we should note that the dynamics at low $T$ should be strongly $F$-dependent. Since the amplitude $A$ is a measure of the PI e-h density, i.e. $A \propto n_s - n_T$, and $n_0$ and $N_0$ are, at low-$T$, proportional to $F$, it follows from Eq. (2) that $A \propto \sqrt{1 + cF} - 1$, where $c$ is a constant. As seen in Fig. 3(a) the $F$-dependence of $A$ for both SmB$_6$ and YbAgCu$_4$ is in excellent agreement with the model.

The initial relaxation rate $\tau^{-1}$ is given by [13]

$$\tau^{-1} = \frac{2R\gamma(n_s + n_T)}{\eta^2(1 + 2\gamma/\eta)}.$$  \hspace{1cm} (3)

It suggests that at low $F$, when $n_s \approx n_T$, the relaxation rate should be $F$-independent, while at high fluences, $\tau^{-1} \propto F$. As shown in Fig. 3(b) the data for SmB$_6$ agree well with the model over the entire range of $F$. In YbAgCu$_4$, on the other hand, at high $F$ the experimental $\tau^{-1}$ increases faster than the theoretical prediction, finally saturating. Saturation is also observed in $A(F)$ for both compounds. This saturation can be attributed to the PI smearing of the hybridization gap structure.

The present $F$-dependence studies clearly support the idea of electron relaxation dynamics in heavy electron compounds being governed by the presence of a hybridization gap in the DOS. Moreover, as was shown in Ref. [13], the $T$-dependencies (at constant $F$) of both $A$ and $\tau^{-1}$ are governed by the $T$-dependence of the number density of thermally excited EHP's, $n_T$. It was shown that $n_T \propto A^{-1} - 1$, where $A(T) = A(T)/A(T \rightarrow 0)$. It is easy to demonstrate that the $T$-dependence of $\tau^{-1}$ is governed by the $T$-dependence of $n_T$ as well. Assuming $R, \gamma, \eta$ to be $T$-independent, Eq. (3) can be rewritten as $\tau^{-1}(T) = C[(n_s - n_T) + 2n_T]$, where $C$ is a proportionality constant. Furthermore, since $A \propto n_s - n_T$ and $n_T \propto A^{-1} - 1$, it follows that

$$\tau^{-1}(T) = C[D(n_T + 1)^{-1} + 2n_T],$$  \hspace{1cm} (4)

where $D$ is a constant that depends only on the photoexcitation intensity. Therefore, for constant $F$, $\tau^{-1}(T)$ is entirely determined by the $T$-dependence of $n_T$.

In a narrow band semiconductor the $T$-dependence of $n_T$ depends on the shape of the DOS in the energy range $\varepsilon \approx kT$ around the chemical potential. Generally, $n_T$ is given by:

$$n_T \simeq T^p \exp(-E_g/2T),$$  \hspace{1cm} (5)

where $p$ is on the order of 1, depending on the exact shape of the DOS near the gap edge. For a hybridization gap...
scenario, the shape of the DOS should be close to that of a BCS superconductor, where $p = 1/2$. Neither the exact shape of the low energy DOS, nor T-dependence of $E_g$ is well known in heavy electron systems, therefore there will be some ambiguity in determining the precise value of the indirect gap $E_g$. However, since the main T-dependence in Eq. (5) comes from the exponential term, a rough estimate of the size and the T-dependence of $E_g$ can be obtained.

In Figure 4(a), we plot the T-dependence of $n_T$ for the two compounds. In both data sets $n_T$ changes by almost 3 orders of magnitude between 20 and 200 K. Moreover, the T-dependence of $n_T$ is far from linear (expected in the TTM scenario [4]) over the entire T-range. Furthermore, the absence of any discontinuity or change of slope in $n_T$ at high-T suggests that the hybridization gap structure persists to high T, as observed, for example, in the photoemission spectroscopy of Kondo insulators [14]. The T-dependence of $\tau^{-1}$ shown in Fig. 4(b) further supports this conclusion: in SmB$_6$, $\tau^{-1}$ increases all the way to the highest temperatures measured, while in YbAgCu$_4$, $\tau^{-1}$ saturates only above $\approx$ 160 K.

![Figure 4: The T-dependence of (a) the density of thermally excited electrons $n_T$ ($\propto A^{-1} - 1$), where $A(T)$ is plotted in the inset, and (b) the initial relaxation rate, $\tau^{-1}$. The data were taken at $F = 8.4$ $\mu$J/cm$^2$. The dashed lines are fits to the data by the RT model (see text).](image)

Based on these qualitative findings, we can apply the RT model to extract the magnitude of the indirect hybridization gap, $E_g$. We consider the hybridization gap to be present at all T, and to a first approximation, to be T-independent. Furthermore, we assume a BCS-like DOS near the gap edge, i.e. $p = 0.5$. Fitting $n_T$ with Eq. (5), we find, for SmB$_6$, an excellent agreement over the entire T-range with $E_g \approx 550$ K. In YbAgCu$_4$, on the other hand, the agreement is good up to $\approx 120$ K, with the extracted value of $E_g \approx 100$ K. At higher T in YbAgCu$_4$, $n_T$ starts to increase faster possibly due to a partial suppression of the gap at high T.

Qualitative agreement between the relaxation rate data and the model described by Eq. (5) is also straightforward. At the intermediate temperatures, $n_s - n_T \ll n_T$, and $\tau^{-1}$ is governed by the T-dependence of $n_T$ exhibiting $\exp(-E_g/2T)$ behavior. At low enough T, however, $n_s \gg n_T$ and the relaxation time saturates, as observed experimentally. We fit $\tau^{-1}(T)$ data with Eq. (11) and find remarkable agreement. The extracted values of the gap are $E_g \approx 85$ K for YbAgCu$_4$ and $E_g \approx 350$ K for SmB$_6$, somewhat lower than the values extracted from the fit to $n_T$. This can be partially attributed to the fact that the T-dependence of $R, \gamma$, and $\eta$ was neglected, as well as to the microscopic details like gap anisotropy. Comparison with literature shows that for SmB$_6$ there is a large spread of published values for $E_g$ [15]; however, our estimate is in close agreement with the T-independent pseudogap energy scale of 290 cm$^{-1}$ (450 K) from a recent Raman scattering study [16]. The value of $E_g \approx 100$ K for YbAgCu$_4$ is also in close agreement with recent optical data [17]. The overall agreement and self-consistency of the data with the RT model presents a strong argument that relaxation kinetics in heavy electron systems is indeed governed by the presence of a weakly T-dependent hybridization gap, as for a periodic Anderson lattice model in the $U = 0$ limit [5, 14].

We have presented studies of photoexcited electron relaxation and recombination dynamics in the Kondo insulator SmB$_6$ and heavy fermion YbAgCu$_4$ as a function of temperature and excitation level. The dynamical response is described well using the phenomenological RT model, suggesting that the carrier relaxation and recombination dynamics in heavy electron systems are governed by the presence of a weakly T-dependent hybridization gap.

We acknowledge discussions with V. Kabanov and S. Trugman. This work was supported by US-Slovenian collaboration grant BI-US/05-06/023 and US DOE LDRD program.

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