THz electrodynamics of mixed-valent YbAl$_3$ and LuAl$_3$ thin films

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Abstract. We present our results from time-domain THz spectroscopy measurements of thin films of mixed-valent YbAl$_3$ and its structural analogue LuAl$_3$. Combined with Fourier transform infrared (FTIR) spectroscopy, the extended Drude formalism is utilized to study the quasiparticle scattering rate and effective masses in YbAl$_3$. We find that LuAl$_3$ demonstrates conventional Drude transport whereas at low temperatures YbAl$_3$ demonstrates a renormalized Drude peak and a mid-infrared (MIR) peak in the conductivity, indicative of the formation of a mass-enhanced Fermi liquid (FL). In YbAl$_3$ the extended Drude analysis demonstrates consistency with FL behavior below the FL coherence temperature $T^\star < 40$ K with the scattering rate following $T^2$ proportionality and a moderate mass enhancement. Despite not observing a clear $\omega^2$ Fermi liquid-like frequency dependence the evidence is consistent with a moderate mass Fermi liquid, albeit one with a smaller mass than observed in single crystals. The extended Drude analysis also demonstrates a slow crossover between the FL state and the normal state above the $T^\star$ in YbAl$_3$, indicative of incoherent hybridization effects persisting to high temperatures.

1 Introduction

Rare-earth intermetallic systems demonstrate a wide variety of low-temperature behavior due to the interplay between electronic correlations and local moment physics. At low temperatures, Kondo physics dominates and the screening of the $f$-electrons leads to the formation of a Fermi-liquid (FL) of heavy quasiparticles below a coherence temperature $T^\star$, giving their name to the heavy fermion (HF) class of materials. These HF materials demonstrate a wide variety of exotic physics including non-conventional superconductivity and quantum phase transitions [1,2]. Related to the class of HF compounds are the family of mixed-valence (MV) materials with non-integer valence on the rare earth site which demonstrate weaker localization of the $f$-orbitals, allowing the interplay of both charge and spin degrees of freedom [3,4]. These materials are characterized by a significantly larger Kondo scale with respect to the Fermi liquid scale, i.e. $T_K \gg T^\star$. While the mixed-valent state is expected to smoothly evolve out of the HF state, there remains many unique phenomena that can arise. Due to their well-separated energy scales, these systems can be tuned to study a variety of emergent phenomena in strongly correlated materials.

Generically, while $f$-orbital systems are expected to exhibit qualitative behaviors consistent with the Anderson single-impurity model formulation, the generalization of the single impurity to the periodic Anderson model (PAM) has remained difficult to treat [5,6]. The single-impurity model has had success in describing the high temperature state of many materials, but as additional coherence effects may appear at the lowest temperatures the periodic Anderson model must be utilized to more accurately describe the emergent phenomena [2,7–9]. Based on the nature of the screening processes occurring, the PAM can give better describe how the crossover between the low-temperature Fermi liquid behavior and the normal state should occur. The concept of a slow crossover arises due to extended screening processes that exist due to the lattice of $f$-orbital ions that are not explicitly described in the single impurity model [10]. In particular, band structure effects and the relative magnitudes of the two energy scales $T^\star$ and $T_K$ play a role in determining the temperature dependence

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of the system and help explain the variation observed in experimental results [11].

Despite extensive studies of lanthanide intermetallic compounds, uncertainty still exists as to why Yb-based correlated materials do not show the same broad range of correlated phenomena as compared to other lanthanide series elements; most notably, there has historically been a stark absence of Yb-based materials that demonstrate unconventional superconductivity. Naively, the commonality between Ce (4f^1) and Yb (4f^13 in Yb^{4+}) compounds as 4f electron-hole counterparts would suggest that they would share a similar range of behaviors [4]; however, experimentally this has not been found to be the case [12–14]. No Yb-based intermetallic material had demonstrated unconventional superconductivity until the discovery in β-YbAlB_4 with T_c = 80 mK by Nakatsuji et al. [15] and more recently in YbRh_2Si_2 with T_c = 2 mK by Schuberth et al. [16]. In β-YbAlB_4 the evidence of a non-FL state transitioning into a superconductivity state at very low temperatures is evidence that the system inherently resides near a quantum critical point (QCP) driven by intrinsic valence fluctuations rather than external pressure [17]. This discovery of historically elusive heavy-fermion superconductivity adjacent to a QCP in Yb-based materials highlights the need for further study of the other mixed-valent Yb-compounds to better understand the role valence fluctuations may play in other strongly correlated systems.

In YbAl_3, the Yb ion has been found to be mixed-valent with a temperature dependence average valence between Yb^{2+} and Yb^{3+} [18,19] and a large separation of energy scales between the heavy Fermi liquid (FL) coherence temperature T^* = 34–40 K and the Kondo temperature T_K ≈ 670 K [20,21]. Consistent with heavy FL quasiparticles forming at low temperatures, there exists a renormalized Drude peak due to the formation of heavy-quasiparticles and a mid-infrared peak corresponding to excitations between the hybridized conduction and f-electron bands [22,23]. The existence of a moderately heavy FL state is supported by experimental results for the specific heat coefficient γ = 40 mJ/mol K and the measured dHvA effective mass below 1.5K of m^* = 15 – 30m_e [21,23–25]. More recently, the growth and subsequent characterization of thin films of YbAl_3 and LuAl_3 using DC transport and ARPES has also uncovered that valence fluctuations drive a Lifshitz transition at 21 K by shifting the chemical potential [26,27]. These recent results suggest that interplay of mixed-valence and Kondo physics can lead to previously unexpected low-temperature behaviors which deserve closer study. In this work we present the optical response of thin films of YbAl_3 and its structural analogue LuAl_3 characterized by time-domain THz spectroscopy (TDTS) and Fourier-transform IR spectroscopy (FTIR). The low-energy scale accessible by our THz spectral range of 0.1–8 meV allows for the direct characterization of low-energy excitations of hybridized states as a function of both frequency and temperature [28,29]. Using the extended Drude formalism, we observe that the quasiparticle scattering rate shows temperature scaling consistent with Fermi liquid theory in YbAl_3 whilst LuAl_3 demonstrates behavior consistent with a simple non-interacting metal. We also observe the formation of a moderately heavy FL in YbAl_3 with a strong temperature-dependent relative mass enhancement from 5 K to room temperature. Additionally, both the quasiparticle scattering rate and effective mass also demonstrate a slow temperature-dependent crossover suggestive of incoherent hybridization effects of Yb f−electrons to the Al conduction band above T^* = 37 K and persisting up to 100 K.

2 Experimental details

The (001)-oriented thin films of YbAl_3 and LuAl_3 used in this study were 15 nm thick and grown on (001) MgO substrates using molecular-beam epitaxy (MBE). For all films, a 0.8 nm thick aluminum buffer layer was first deposited. This allowed the growth of continuous, smooth films of LuAl_3/YbAl_3. In these studies, we investigated a 20 nm thick LuAl_3 film and a 15 nm thick YbAl_3 film. The YbAl_3 was synthesized on top of a 5 nm thick LuAl_3 buffer layer on top of the Al buffer, which improved the quality of the YbAl_3 layer. All films were capped with 2 nm thick layer of Al which was assumed to oxidize and become insulating. Characterization of these films was conducted using x-ray diffraction, low energy electron diffraction and scanning transmission electron spectroscopy. Despite the dramatic advances in growing such films by MBE, it is important to note that the residual resistivity ratio (\(\frac{\rho(300 \text{ K})}{\rho(5 \text{ K})}\)) of the YbAl_3 film is still much less than the approximately 60 found in high-quality single crystals [24]. More details about the thin film growth and characterization can be found elsewhere [26].

The low-energy response of the films were characterized using time-domain THz spectroscopy (TDTS). In TDTS, an approximately single-cycle ps long pulse of radiation is transmitted through the sample. One takes the Fourier transform of the transmitted electric field and references it to the signal through a bare MgO substrate to get the complex transmission of the thin films. An advantage of the technique is that the complex conductivity can be directly extracted from the complex transmission without a Kramers–Kronig transform. The complex conductivity \(\tilde{\sigma}\) can be determined from the transmission by,

\[
\tilde{T}(\omega) = \frac{n_s + 1}{n_s + 1 + Z_0 d \tilde{\sigma}(\omega)} e^{i \xi \Delta L(n_s - 1)},
\]

where \(n_s\) is the substrate index of refraction, \(Z_0\) is the impedance of free space, \(d\) is the sample thickness, \(\tilde{\sigma}\) is the complex conductivity, \(\omega\) is the frequency, \(c\) is the speed of light, and \(\Delta L\) is a correction factor due to the difference in thickness between the substrate and the sample [30]. In order to isolate the conductance of
Fig. 1  DC resistivity as a function of temperature for the YbAl$_3$ and LuAl$_3$ thin films. The inset shows the adherence of the resistivity to $T^2$ behavior for YbAl$_3$ up to $T^* = 40$ K. These dc measurements were performed on similarly prepared, but different films than the THz measurements, so the correspondence with the THz data in Fig. 3 is not precise.

the high quality sample layer of LuAl$_3$/YbAl$_3$ from the buffer and capping layers, a reference film comprised of the buffer and capping layers (without a sample film) was used to correct for the presence of parallel conductance channels. The magnitude of the conductance of the reference film was less than 10% of the conductance of the sample films and displayed no temperature dependence. By subtracting the conductance of the buffer layers from the total conductance of the sample films, the conductivity of the LuAl$_3$/YbAl$_3$ layer could be obtained.

In tandem with the THz measurement, the mid-IR (MIR) response (400–7500 cm$^{-1}$, 0.05–930 meV) was studied using conventional Fourier-transform infrared (FTIR) spectroscopy down to 5 K. Transmission experiments were measured using a commercial FTIR spectrometer (Bruker Vertex 80 V) using a Globar source and a solid-state DTGS detector.

3 Results

In Fig. 1, the DC resistivity of films of the two materials are shown. At the lowest temperatures, the YbAl$_3$ film demonstrates a $\rho \propto T^2$ dependence as shown previously [26]. The resistivity saturates above some temperature $T^*$ that is of order 200 K. The resistivity of the LuAl$_3$ film shows a monotonic decrease with decreasing temperature.

In order to reference out the buffer layer used to grow the sample layers of LuAl$_3$ and YbAl$_3$, the THz transmission of the bare buffer layer and substrate was independently measured and is shown in Fig. 2 alongside the THz transmission and conductance for the two sample films. The conductance of the buffer layer was small (< 10% of the conductance of the sample films) and temperature independent which when subtracted from the sample films would give the isolated conductance of the sample layers. In Fig. 3 we present the real and imaginary parts of the THz conductivity for YbAl$_3$ and LuAl$_3$ at different temperatures. At high temperatures, both films demonstrate large scattering rates indicated by the flat, nearly frequency independent real conductivity. Upon cooling the scattering rate decreases in
Fig. 3  a, b Real and imaginary THz conductivity of YbAl$_3$ as a function of frequency, c, d real and imaginary THz conductivity of LuAl$_3$ as a function of frequency. In the YbAl$_3$ sample, we observe a fairly significant change in the frequency dependence of the conductivity consistent with the scattering rate decreasing as the temperature decreases. In comparison, note how the LuAl$_3$ complex conductivity demonstrates a much lower change from 5 to 300 K

Fig. 4  MIR transmission shown above for YbAl$_3$ (a) and LuAl$_3$ (d). The real and imaginary parts of the optical conductivity modeled using the combined THz complex conductivity and MIR transmission is shown in (b), (c) for YbAl$_3$. The real and imaginary parts of the optical conductivity for LuAl$_3$ are shown in (e) and (f)
both films; however, in YbAl$_3$, the decrease in the scattering rate is observed to be significantly greater from much beyond the THz spectral and can be clearly seen within our spectral range for the lowest temperatures. In the case of YbAl$_3$ sample, at the lowest temperatures there is an increase in the low-frequency spectral weight as seen by the enhancement of the real and imaginary parts of the conductivity compared to the room temperature state. This is consistent with the formation of a narrower Drude peak at the lowest temperatures. This is in contrast to the LuAl$_3$ film, which is characterized by a broader Drude-like contribution to the conductivity without a significant temperature dependence of the scattering rate when compared to the much larger change seen in the YbAl$_3$ film. Additionally, from the real part of the conductivity it is clear that in the case of the LuAl$_3$ film the temperature dependence of the spectral weight is not as large in the YbAl$_3$ film, suggesting no strong renormalization effects.

The MIR transmission of the YbAl$_3$ and LuAl$_3$ thin films are shown in Fig. 4. Using both the THz complex conductivity and the MIR transmission, the optical conductivity was modeled using Drude-Lorentz parameterization in the fitting software refFIT [31]. The LuAl$_3$ data was fit by two Drude terms, whereas the fit to the YbAl$_3$ data used two Drude terms and a single high frequency Drude-Lorentz oscillator. In order to account for the buffer layer present in both sample films, a multi-layer model was generated in refFIT using the parameterization of the bare buffer layer on the substrate to isolate the transmission of the individual sample layers. Due to the strong absorption of the underlying MgO substrate, the transmission below 1000 cm$^{-1}$ was not accessible. Hence, in the FIR region without experimental data, the modeled conductivity determined through the Kramers–Kronig consistent fitting can only give a rough guide, but is not a definitive measure of the conductivity. From the MIR data on the YbAl$_3$ thin film, we see a clear MIR conductivity peak suggestive of a hybridization gap [32], which becomes enhanced as the temperature decreases. The position of the MIR conductivity peak in YbAl$_3$ is centered around $\omega = 2800$ cm$^{-1}$; the corresponding value in single crystals has been found to be centered at $\omega \sim 2000$ cm$^{-1}$ [23,33]. While there is no strain expected to arise from the underlying Al and LuAl$_3$ buffer layers on MgO, this shift in the conduction electron-$4f$ hybridization resonance can be attributed to the effects of strong impurity scattering [34].

4 Discussion

From the comparison of the optical response of LuAl$_3$ to YbAl$_3$, it appears that the full $f$–orbital in the Lu compound prevents the development of the heavy quasiparticles as seen in the YbAl$_3$ film. In order to address the complex quasiparticle behavior, it is useful to use the framework of the extended Drude model by allowing the scattering rate and effective mass to take on both frequency and temperature dependence [35]. These quantities can be arrived at in a model-free fashion by inverting the complex conductivity as,

$$\frac{m^*(\omega)}{m_b} = -\frac{\omega_p^2}{4\pi \omega} \text{Im} \left[ \frac{1}{\sigma(\omega)} \right], \quad (2)$$

$$\frac{1}{\tau(\omega)} = \frac{\omega_p^2}{4\pi} \text{Re} \left[ \frac{1}{\sigma(\omega)} \right], \quad (3)$$

where $m_b$ is the band mass and $\omega_p$ is the intraband plasma frequency. The exact value of the plasma frequency only serves to scale the renormalized mass and scattering rate and has no impact on their temperature and frequency scaling. The intraband plasma frequency was determined by fitting the complex THz conductivity and the MIR transmission and summing the spectral weight of each of the fitted low-frequency Drude oscillators. In YbAl$_3$, since the high frequency Drude-Lorentz oscillator was centered at 2800 cm$^{-1}$, our parameterization gave clear separation between the intraband and interband oscillators. Hence, the intraband plasma frequency was determined to be $\omega_p = 2\pi \times (95 \pm 10)$ THz.

In Fig. 5 we present the extended Drude scattering rate as both a function of frequency and temperature. At low temperature frequency dependence of the scattering rate does not exhibit a large frequency depen-

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**Fig. 5** Extended Drude scattering rate $\Gamma = 1/2\pi\tau$ of YbAl$_3$ as a function of frequency (a) and temperature (b) taken along frequency cuts indicated by vertical dashed lines in the upper figure.
Extended Drude scattering rate of the YbAl$_3$ sample at 1 THz plotted against $T^2$ to compare to FL predictions, with an inset to focus on how the data begins to deviate for $T \sim 100$ K. This minor deviation of the experimental data from the $\Gamma \propto T^2$ linear fit for $T < 40$ K up to 100 K is suggestive of a slow crossover from the FL state to the normal state above $T^\ast$.

Extended Drude effective mass of the YbAl$_3$ sample as a function of frequency (a) and temperature (b) taken as a frequency cut at $\omega = 1.0$ THz normalized to the high temperature value at 300 K. We observe a nearly fivefold increase from the normal state at high temperatures to 5 K in the FL state.
In, ...) from transport measurements. Based on results from a more recent photoemission study on the same class of YbXCu$_4$ compounds, the phenomena of slow crossover between the low temperature and the high temperature normal state was found consistent with residual low energy spectral weight that persists into intermediate temperatures [40]. This residual spectral weight phenomena is consistent with what we observe in YbAl$_3$ as seen by the enhanced effective mass above the FL coherence temperature $T^*$. This suggests that in YbAl$_3$ hybridization effects are still present above $T > 40K$ and demonstrate the slow crossover in YbAl$_3$ as driven by protracted screening processes predicted by the PAM.

5 Conclusion

The field of heavy-fermion physics is relevant to answering questions regarding exotic phenomena that emerge from strongly correlated electronic systems. The recent development of high-quality MBE thin films have allowed the first TDTS experiments to be conducted on YbAl$_3$ and LuAl$_3$. The low energy scales of the heavy quasiparticles makes TDTS an ideal probe to study quasiparticle dynamics and enhance our understanding of these complex systems. From our results, we find that the LuAl$_3$ film is well described by conventional Drude transport as expected for a normal metal. The extended Drude formulism was used to study YbAl$_3$ and the quasiparticle scattering rate demonstrates $T^2$ scaling up to the FL coherence temperature $T^*$ consistent with 4-probe measurements and previous studies on single crystals. Due to the small spectral range of our TDTS measurement, we do not see clear signatures of $\omega^2$ scaling for the scattering rate in the scattering rate. There was also a moderate quasiparticle mass enhancement observed at the lowest temperatures, albeit less than the values suggested by quantum oscillations, specific heat measurements or optical spectroscopy on bulk samples. The reason for the smaller mass enhancement is unclear, but may be attributed to strong disorder scattering in these relatively low RRR films. However, from the $T^2$ temperature scaling of the quasiparticle scattering rate and effective mass, we find that above the coherence temperature $T > T^*$ and up to intermediate temperatures $\sim 100 K$ there are signatures of FL-type behavior. This is evidence for a slow-crossover driven by extended screening processes from the FL state to the normal state observed in YbAl$_3$ and other mixed-valent Yb compounds.

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Author contributions

DB performed the THz experiments and analyzed the data. SC grew the thin films. DB and NPA prepared the first draft, and all authors contributed to writing the manuscript.

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