Anomalous frequency and temperature dependent scattering and Hund’s coupling in the almost quantum critical heavy fermion system CeFe$_2$Ge$_2$

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We present THz range optical conductivity data of a thin film of the near quantum critical heavy fermion compound CeFe$_2$Ge$_2$. Our complex conductivity measurements find a deviation from conventional Drude-like transport in a temperature range previously reported to exhibit unconventional behavior. We calculate the frequency dependent effective mass and scattering rate using an extended Drude model analysis. We find the inelastic scattering rate can be described by a temperature dependent power-law $\omega^{n(T)}$ where $n(T)$ approaches $1.0 \pm 0.2$ at 1.5 K. This is compared to the $\rho \sim T^{1.5}$ behavior observed in dc resistivity data and the $\rho \sim T^2$ expected from Fermi-liquid theory. In addition to a large low temperature mass renormalization, we find an anomalously large mass renormalization that persists as high as room temperature. We attribute this to a Hund’s coupling in the Fe states in a manner similar to that recently proposed in the ferro-pnictides. CeFe$_2$Ge$_2$ appears to be a very interesting system where one may study the interplay between the usual 4$f$ lattice Kondo effect and this Hund’s enhanced Kondo effect in the 3$d$ states.

Many heavy fermion (HF) systems can be described within the framework of Fermi liquid (FL) theory, with quasiparticles exhibiting renormalized masses that are sometimes up to three orders of magnitude larger than bare electrons. However, it is not universal that these systems always obey FL predictions; many HF systems deviate from FL response for electrical resistivity and specific heat. One explanation of such non-FL properties involves the proximity to magnetic order and a quantum critical point. Quantum criticality is seemingly ubiquitous across the diverse landscape of correlated matter and at the forefront of current research in novel phenomena of the cuprates, ruthenates and iron pnictides. Probing the emergent state of matter near a quantum critical point can, then, lead to further understanding of unsolved challenges, such as in high temperature superconductivity or hidden order phases.

There is a burgeoning list of HF materials that can be tuned toward quantum criticality. CeFe$_2$Ge$_2$ (CFG) is a HF compound with a non-magnetic ground state and a moderately enhanced heat capacity of 210 mJ/mol-K$^2$. It exhibits a metamagnetic anomaly at 300 kOe and is also believed to be close to a quantum critical point. This behavior is reminiscent of other 4$f$-electron systems: CeRu$_2$Si$_2$, CeNi$_2$Ge$_2$ and CeCu$_6$. In addition to metamagnetic anomalies, deviations from ordinary FL behavior occur under certain circumstances in these materials. CFG has been reported to show deviations from FL predictions; a $T^{1.5}$ dependence of resistivity in the temperature range $\sim$2-15 K is observed instead of the FL $T^2$ dependence. A crossover to a typical FL ground state is observed below a temperature $T_{FL} \approx$ 2 K. By examining the Ce(Ru$_{1-x}$Fe$_x$)$_2$Ge$_2$ series, a phase diagram was constructed which shows that CFG is in proximity to a quantum critical point (QCP). In this series, ferromagnetic CeRu$_2$Ge$_2$ transitions to antiferromagnetic order around $x=0.3$ and then goes through a QCP at $x=0.9$ into a paramagnetic state. The existence of a QCP near the CFG end of the series could explain the non-FL behavior previously mentioned.

In this paper we use time domain terahertz spectroscopy (TDTS) to study the low-frequency complex conductivity of thin films of CFG and calculate the renormalized frequency dependent scattering rate and mass using an extended Drude model analysis. Previously, the deviation from FL behavior in HF materials has been revealed primarily in the temperature dependence of the resistivity. How this non-FL behavior may be seen in an ac technique is not as clear, but keeping within a quasi-Boltzmann transport point of view, a natural analog to the temperature dependence of resistivity may be the frequency dependence of the scattering rate. Therefore, deviations from FL $\omega^2$ dependence can be analyzed by fitting the frequency dependent scattering rate using a power law constrained by the dc scattering rate.

In TDTS an infrared femtosecond laser pulse is split between two paths and sequentially excites a pair of “Auston” switch photoconductive antennas. The first switch generates the THz pulse, which then travels through the sample. The second antenna receives the THz pulse and measures both the phase and amplitude of its electric field. By dividing the Fourier transform of transmission through the sample by the Fourier transform of transmission through a reference substrate, the full complex transmission $T(\omega)$ as function of frequency can be obtained over a frequency range as broad as 100 GHz to 3.5 THz. Details can be found elsewhere.
where [11]. The complex transmission is used to calculate the complex conductivity \(\sigma(\omega)\) without the need for Kramers-Kronig transformation using the expression
\[
T(\omega) = \frac{1}{1+n+\rho(\omega)dZ_0} \cdot e^{\frac{i\omega\Delta L(n-1)}{\epsilon}}.
\]
In this expression \(n\) is the index of refraction of the substrate, \(\Delta L\) is a correction factor that accounts for thickness differences between the reference substrate and the sample substrate, \(d\) is the film thickness, and \(Z_0\) is the impedance of free space (377 \(\Omega\)). The films studied in this work were grown by molecular beam epitaxy to a thickness of 524 \(\AA\) using flux-matched codeposition of the constituent atoms on MgO substrates. Further details of the growth have been reported elsewhere [17].

In Fig. 1 we show dc resistivity which decreases monotonically as temperature is lowered. Around 15K there is a subtle crossover that can be better seen in the inset of Fig. 1, where the zero temperature residual resistivity \(\rho(0)\) has been subtracted off. We find \(\rho(T) - \rho(0) \propto T^{1.4}\) for temperatures between \(\sim 2-15K\), which is consistent with previous measurements [17]. A \(T^{1.5}\) dependence of the resistivity was previously explained as an anomalous property around a magnetic instability, similar to other HF systems [18].

In Fig. 2 we show the real and imaginary parts of the complex conductivity \(\sigma(\omega)\) for a few temperatures (see Ref. [19] for all data). At high temperatures both parts of the complex conductivity are flat and featureless. As the temperature is lowered a shift in spectral weight toward low frequencies is seen in the real part of the conductivity \(\sigma_1\). A narrow Drude-like peak grows as the temperature is decreased to 1.55 K. Using the dc data, the low-temperature terahertz (THz) conductivity can be fit to a Drude model using two zero-frequency oscillators and a high-frequency dielectric constant \(\epsilon_\infty\), which accounts for high-frequency contributions above the measured spectral range [20]. This two Drude component fit should be interpreted as a simple Kramers-Kronig consistent parameterization of the data. It does not necessarily correspond to two distinct charge carrier species (see Ref. [19] for details). Fits to the complex conductivity are shown as dashed lines in Fig. 2. These fits are highly constrained by the use of both THz and dc data. Implicit in this fitting is the reasonable assumption that there are no spectral peaks in \(\sigma_1\) below the lower end of our measured range [11, 21, 22].

The fact that we cannot fit our data with a single term Drude model demonstrates that the transport cannot be described by a single energy-independent relaxation time. An alternative to fitting the data with multiple Drude-like terms is the extended Drude model [23]. In this formalism, a frequency-dependent mass \(m^*(\omega)/m_b\) and scattering rate \(1/\tau(\omega)\) are extracted from the measured optical constants by the relations
\[
\frac{m^*(\omega)}{m_b} = -\frac{\omega_p^2}{4\pi^2} \text{Im} \left[ \frac{1}{\sigma(\omega)} \right] \tag{1}
\]
\[
\frac{1}{\tau(\omega)} = \frac{\omega_p^2}{4\pi} \text{Re} \left[ \frac{1}{\sigma(\omega)} \right] \tag{2}
\]
where \(\omega_p\) is the plasma frequency that is a measure of the total Drude spectral weight (proportional to \(\omega_p^2\)) of all free charge carriers and \(m_b\) is the band mass. To determine the total spectral weight of all the free charge carriers one must measure to much higher frequencies than the THz regime. In this regard, we also performed Fourier
transform infrared spectroscopy transmission and reflection measurements from 100 to 8000 cm\(^{-1}\) from room temperature down to 5 K. With the TDTS results, we fit the full measured spectral range (from 0 to 8000 cm\(^{-1}\)) to a generalized Kramers-Kronig consistent Drude/Drude-Lorentz model (see Ref. [19] for details). A partitioning of spectral weight was seen such that we could extract the total effective spectral weight of all free Drude carriers as \(\omega_p = 2\pi \times (12150 \pm 1000)\) cm\(^{-1}\) (see Ref. [19] for details). There was no noticeable change between 295 K and 5 K in the infrared transmission data. The uncertainty on the absolute numerical value of \(\omega_p\) arises from uncertainties in assigning spectral weight to either free carrier Drude or to finite frequency Drude-Lorentz oscillators. While this uncertainty sets the overall scale of the renormalized optical quantities, it does not affect their frequency or temperature dependence.

In Fig. 3 (a) we show the frequency dependence of the scattering rate for a few temperatures between 1.55-100 K calculated using Eq. [10] The dc points are calculated using the plasma frequency obtained above and resistivity data. Fits to the data, shown as dashed lines, were created by using the 2 Drude component model, which previously parameterized the conductivity, in extended Drude model equations. These fits serve as a Kramers-Kronig compatible way to connect the THz data to the dc data. The detailed functional dependence should not be viewed as exact, but the general dependence should be valid. For temperatures below 15K, on top of a large constant offset, a frequency dependence develops to the scattering rate. We fit the scattering rate data to a power law \(\frac{1}{\tau(\omega,T)} = \frac{1}{\tau(0,T)} + A\omega^n\) where \(\frac{1}{\tau(0,T)}\) is the dc scattering rate and \(n\) is a temperature dependent exponent.

The scattering rate minus the dc value is plotted versus frequency with a corresponding power law fit for a few temperatures below 12 K in Fig. 3 (b). These fits are constrained to fit data from 0.2-1.5 THz and pass through zero at zero frequency. In Fig. 3 (c) we show the power law exponent of the fits shown in Fig. 3 (b).

In the temperature range previously known to exhibit non-FL behavior, we observe patterns that suggest a deviation from conventional FL dependence: for temperatures below ~3 K the power law exponents are on average 0.94 ± 0.02. From 3-10 K the power law exponent averages 1.30 ± 0.27. Above 10 K the exponent is close to 2. Errors in the power law fits mostly originate in determining over which frequencies to fit and, unfortunately, make it difficult to distinguish whether there is an unequivocal difference between the frequency dependence found here and the \(T^{1.5}\) behavior reported for the dc resistivity [7] at temperatures between 1.5 and 10 K. The presence of different exponents appearing for the temperature and frequency dependencies of the scattering rate is anomalous and does not find ready explanation in the Hertz-Millis-Moriya (HMM) model of quantum criticality above the critical dimension [5, 24-27]. Despite the seeming conflict, we note that this difference in the functional dependence of the optical self-energy is similar to the differences in the functional dependence expected within the HMM formalism for the susceptibility itself [28, 29]. In view of this, a possible connection between these quantities is left open for future theoretical models. An alternative explanation is that the reported \(T^{1.5}\) dependence in the resistivity, in fact, reflects a crossover from a larger exponent to the approximately 1 we find at 1.55K. In this case, we would find the same approx-
approximately linear dependence for $T$ and $\omega$ in both low energy limits. This may be consistent with some models for HMM criticality in clean systems.\cite{27} Whichever the case, our data provides further evidence that there is a deviation from canonical FL theory which predicts the scattering rate’s dependence on frequency and temperature to differ only by a factor of $4\pi^2$ via the relation $\tau_{\omega,T} = A[(\omega)^2 + (2\pi k_B T)^2]$, where $A$ is a constant.\cite{30} While our data could be described by a somewhat modified version of this equation, in which the coefficient of the temperature term differs from $4\pi^2$, with quadratic dependencies it is only possible to fit the data in a very narrow frequency range, reinforcing the idea that FL theory is not a good description at this energy scale.

Continuing with our extended Drude model analysis, we show the renormalized mass as a function of frequency and the $\omega \rightarrow 0$ limit of the masses as a function of temperature in Fig. 4. The renormalized mass does not exhibit strong dependence in the measured frequency range; the renormalization is largest below 0.3 THz, which is just above the cut-off of our data. However, because we know the dc scattering rate and the renormalized mass is related to this via Kramers-Kronig, we can determine how the mass interpolates to zero frequency using an extended Drude model fitting technique which simultaneously fits the scattering rate and mass (further details are discussed in \cite{19}).

We would like to emphasize that one powerful feature of the TDTS technique is its ability to measure the temperature dependence of the mass renormalization. Other techniques (e.g. heat capacity and quantum oscillations) that have contributed to the study of HFs typically only extract the mass renormalization at the lowest temperatures through the temperature dependence itself once the coherent state has developed. In contrast, TDTS allows us to see the formation of the heavy state; with the extended Drude model analysis we can measure the mass renormalization from high temperatures all the way into the coherent state. In this regard, one particularly notable feature of this data is the large mass renormalization that persists as high as $m^*/m_b \sim 6$ at room temperature, as seen in Fig. 4(b). The overall scale of the mass is set by the measured plasma frequency and although it is possible that we have parameterized it inaccurately, the present value of the low temperature renormalized mass is consistent with heat capacity measurements and materials with similar specific heat coefficients.\cite{6, 17}.

We believe that this large room temperature mass enhancement follows from a “Hund’s coupling enhanced” Kondo effect as proposed for ferropnictide compounds with related crystal structures and composition.\cite{31-34} In the case of KFe$_2$As$_2$, local density approximation + dynamical mean field theory calculations suggest that the mass enhancement in iron pnictides occurs because the electrons, which are somewhat localized at high temperature due to Hund’s rule coupling, form coherent quasi-particle bands with the underlying Fermi surface.\cite{35} Further Gutzwiller corrected electronic structure calculations for this material support the idea of an “orbital selective” Mott transition, in which localization due to strong Coulomb repulsion occurs for only some of the orbitals.\cite{35}. Qualitative aspects of these calculations should apply to the present case, in which Fe$^{2+}$ is found in the same 122 crystal structure. The interplay of the localized and delocalized bands can then give rise to behavior very similar to that of 4f-based heavy fermion systems, even though all states formed in KFe$_2$As$_2$ are from Fe 3d orbitals.\cite{35}. With that in mind, it is likely that a strong Hund’s interaction in the 3d Fe atoms creates a mass enhancement at temperatures higher than naively expected from the usual treatment of the 4f Ce moments hybridizing with the conduction band. Bolstering this interpretation is the observation that both the 4f moment free compounds of LaFe$_2$Ge$_2$ and LaFe$_2$Si$_2$ show anomalously large low temperature specific heat coefficients of 37 mJ/mol-K$^2$ and 22.7 mJ/mol-K$^2$ respectively.\cite{6, 36}. These can be compared to La 122 compounds based on wider d-band materials with Ru and Cu that have more conventional magnitudes of 5-7 mJ/mol-K$^2$ respectively.\cite{37, 38}. CFG (with a heat capacity of 210 mJ/mol-K$^2$) then appears to be a very appealing system to study the interplay between the usual 4f Kondo effect and this Hund’s enhanced Kondo effect in the 3d states.

In summary, we have applied time domain THz spectroscopy to the heavy fermion compound CeFe$_2$Ge$_2$ and investigated the non-FL behavior of its optical properties using an extended Drude model analysis. Evidence was found in the frequency dependent scattering rate to support previous suggestions that CFG shows non-FL behavior in the temperature range 2-15K. We find a crossover in the inelastic scattering rates that can be

![FIG. 4. (Color online) (a) Renormalized masses as functions of frequency. (b) The $\omega \rightarrow 0$ limit of the frequency dependent renormalized masses as functions of temperature extracted from extended Drude model fits. Error bars reflect uncertainty in the extended Drude model fits, while the grey shadow reflects the uncertainty from the range of possible values of $\omega_p$.](image-url)
described by a power-law $\omega^n$ where $n$ becomes approximately unity in the same temperature range where the dc resistivity is reported to show $T^{1.5}$ dependence. Counter to the usual Kondo scheme for Ce based compounds, we find that the mass enhancement persists as high as room temperature, an effect that we believe originates in a “Hund’s coupling enhanced” Kondo effect. CeFe$_2$Ge$_2$ seems to be an interesting system where one may investigate the interplay between the standard 4$f$ lattice Kondo effect and this Hund’s enhanced Kondo effect in the 3d states. In further experiments it would be interesting to compare LaFe$_2$Ge$_2$ with LaCu$_2$Ge$_2$ and LaRu$_2$Ge$_2$ to isolate the effect of Hund’s coupling.

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Conductivity measurements and fitting analysis

Conductivity measurements were taken at temperatures between 1.55 K and room temperature in a continuous-flow liquid He-4 cryostat. Because of the large number of temperatures at which data was taken, displaying the complex conductivity as a function of frequency for all temperatures at once masks the details of the individual curves and was not shown in the primary text. However, the systematic development of a narrow Drude peak in the real part of the conductivity as temperature is lowered is seen very nicely in this fashion. The real and imaginary parts of the complex conductivity are shown for the full temperature spectrum in Fig. 5. The data is cut to a frequency range of 200 GHz-1.5 THz. The dc conductivity points, calculated from the dc resistivity data taken using a 4-probe technique, are also shown as solid markers. Drude-Lorentz model fits, shown as dashed lines, were performed assuming there are no sharp features below 200 GHz and the dc value.

The Drude-Lorentz model fits were performed using the software RefFit [1]. This program is capable of fitting multiple complex valued datasets of different experimental types simultaneously with multiple models, which may depend on the same parameters. We fit the real and imaginary parts of our measured conductivity with multiple complex valued datasets of different experiments to the well known sum rule for optical conductivity

\[ 8 \int_0^\infty \sigma_1(\omega) d\omega = \frac{4\pi Ne^2}{m_e} \]

where \( N \) is the total number of charges and \( m_e \) is their mass. However, the integration extending to infinity presents a considerable experimental complication and the sum rule is usually restricted to a partial sum rule

\[ 8 \int_0^W \sigma_1(\omega) d\omega = \frac{4\pi Ne^2}{m_b} \]

where \( W \) is the unrenormalized electronic bandwidth, \( N \) is the number of mobile charge carriers and \( m_b \) is the band mass. From this, we can define the plasma frequency \( \omega_p^2 = \frac{4\pi Ne^2}{m_b} \). If \( W \) is sufficiently high enough we recover the full sum rule. For strongly interacting systems we can further limit the sum rule and perform the integration over an energy \( E \) only several times the Drude width to determine the renormalized mass \( m^* \) due to interactions

\[ 8 \int_0^E \sigma_1(\omega) d\omega = \frac{4\pi Ne^2}{m^*} \]

In this case we can define a renormalized plasma frequency \( \omega_{p^*}^2 = \frac{4\pi Ne^2}{m^*} \), which only describes the spectral weight of a very narrow interaction derived band, not the full contribution described in Eq. 6. The ratio of these two plasma frequencies is often cited in the context of heavy fermion systems \( \frac{\omega_{p^*}^2}{\omega_p^2} = \frac{m^*}{m_b} \) to describe the large renormalization of the charge carrier mass due to hybridization of the band of conduction electrons with the localized band of magnetic moments.

The question remains: which plasma frequency should be used in the extended Drude model Eqs. 3-4? Recall that the origin of the extended Drude model is to account for the various inelastic channels that contribute to the scattering rate with different frequency dependencies. While the Drude-Lorentz model treats the scattering rate

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

\[ \frac{1}{\tau(\omega)} = \frac{\omega_p^2}{4\pi} Re \left[ \frac{1}{\sigma(\omega)} \right] \]
The first step of the fitting analysis is to model the composite system. By doing this, we are able to isolate the contribution from each layer and outputs the dielectric function for each layer. We can then use the models that parameterize the dielectric function to create a “skeleton model” which will characterize the CFG layer. For this we separate the oscillators into low-frequency Drude and Drude-Lorentz terms, which characterize the intraband transitions, and higher frequency Drude-Lorentz oscillators, which characterize interband transitions. The plasma frequencies associated with the intraband terms are then added in quadrature to determine \( \omega_p \) that we will use in the extended Drude model calculations. Finally, we create another model, which is specially designed for multilayer systems (special code -33). The normal parameters of any generic model are reassigned in this model: each row in the model represents a layer of the system. There are actually two of these models—one for transmission and another for reflection fitting. We can now fit these models to the CFG + MgO multilayer FTIR transmission data, the CFG+MgO+gold FTIR reflection data and the THz conductivity data. The only parameters free to fit are in the CFG layer model.

The main source of error in determining \( \omega_p \) within this fitting technique arises from distinguishing at what frequency the influence of interband transitions begins to contribute to the low-frequency physics that is most interesting in. To quantify this error we took our best model and split a “mid-frequency” oscillator (1500 cm\(^{-1}\)), which had a very broad scattering rate, into two oscillators: one at a lower frequency with a sharper scattering rate and the other at a higher frequency with a broad scattering rate. By doing this, the distribution of the spectral weight of the original oscillator is now more easily identified with either intra or interband transitions while the quality of the fit is maintained.

Deciding how to separate low(intraband) from high(interband) frequency processes is motivated from the spectral weight of the narrow Drude peak associated with the heavy charge carriers and the spectral weight of the high frequency processes. Once this is known, these models are fixed and are not be adjusted during further fitting steps. Next, we consider the role of the interband contribution as being characterized by a single frequency independent relaxation time, the extended Drude model captures these frequency dependencies by explicitly making the scattering rate and mass frequency dependent. Therefore, \( \omega_p \) must come from the spectral weight of the full intraband contribution. In the context of sum rules,

\[
8 \int_0^\infty \sigma_1^{\text{intra}}(\omega) d\omega = \omega_p^2 \tag{8}
\]

where \( \sigma_1^{\text{intra}} \) represents the conductivity from all intraband contributions. Again, the integration to infinity is usually replaced by a high energy cut-off that is large enough to capture all intraband transitions while minimizing the contaminating contribution of interband transitions. In our case, the plasma frequency that enters into the extended Drude model equations reflects both the spectral weight of the narrow Drude peak associated with the heavy charge carriers and the spectral weight of other higher frequency scattering processes.

To experimentally determine \( \omega_p \) of CFG we performed Fourier transform infrared (FTIR) spectroscopy in transmission and reflection geometries from 100-8000 cm\(^{-1}\) and with the THz data, we used the same modeling software, RefFit, to parameterize the spectra\[1]. In the reflection geometry, the sample and substrate were mounted on top of gold mirrors and referenced to identical mirrors. A special function in RefFit that models a multilayer sample was chosen to fit the CFG thin film on an MgO substrate (special code -33). This function takes as input the models that parameterize the dielectric function of each layer and outputs the dielectric function of the composite system. By doing this, we are able to isolate the optical response of the CFG film.

An example of the fitting technique is seen in Fig. 6. The first step of the fitting analysis is to model the dielectric function of the MgO substrate and the gold mirror. Once this is known, these models are fixed and are not be adjusted during further fitting steps. Next, we create a “skeleton model” which will characterize the CFG layer. For this we separate the oscillators into low-frequency Drude and Drude-Lorentz terms, which characterize the intraband transitions, and higher frequency Drude-Lorentz oscillators, which characterize interband transitions. The plasma frequencies associated with the intraband terms are then added in quadrature to determine \( \omega_p \) that we will use in the extended Drude model calculations. Finally, we create another model, which is specially designed for multilayer systems (special code -33). The normal parameters of any generic model are reassigned in this model: each row in the model represents a layer of the system. There are actually two of these models—one for transmission and another for reflection fitting. We can now fit these models to the CFG + MgO multilayer FTIR transmission data, the CFG+MgO+gold FTIR reflection data and the THz conductivity data. The only parameters free to fit are in the CFG layer model.

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FIG. 6. A screenshot of the fitting program, RefFit, used to determine $\omega_p$ of CFG. A special multilayer model is necessary to fit the FTIR reflection and transmission data.

previous work done on similar compounds \cite{4,5}. Two methods are commonly used: 1) the frequency at which there is a minimum in the real part of the conductivity and 2) the frequency at which the imaginary part of the conductivity crosses from positive to negative. At these frequencies the influence of interband processes dominates the Drude response. Unfortunately, instrument and sample limitations do not allow us to measure the conductivity beyond the THz regime, but the model that we generate using Reffit shows these features and confirms our belief that the cut-off energy scale in CFG is $\sim 3000 \text{ cm}^{-1}$. This is similar to other Fe based materials with the same crystal structure \cite{4,5}. Table I shows the original model with all oscillators and the final model after splitting the oscillator at 1500 cm$^{-1}$. Table II gives more detail about this splitting process. It is seen in this table that an acceptable range of plasma frequencies to characterize the lower intraband oscillator is $\sim 7000$-10000 cm$^{-1}$ since their scattering rates are sufficiently narrow that most of their spectral weight still lies below the high energy cut-off of $\sim 3000 \text{ cm}^{-1}$. Using this range and adding the spectral weight of the oscillators centered below 3000 cm$^{-1}$ in quadrature we determine $\omega_p = 12150 \pm 1000 \text{ cm}^{-1}$.

### Extended Drude model analysis

Fits to the renormalized scattering rate and mass were also performed using RefFit software\cite{1}. A built-in function (special code -6) specifically designed for the extended Drude model takes as input a dielectric function $\epsilon(\omega)$ given by the dielectric model calculated previously in our conductivity fitting and converts it to the frequency-dependent scattering rate and effective mass.

| type       | $\omega_o$ | $\omega_p$ | $\Gamma$ |
|------------|------------|------------|-----------|
| intraband  | 0          | 3900       | 67        |
| intraband  | 40         | 1200       | 65        |
| intraband  | 60         | 1755       | 50        |
| intraband  | 250        | 7500       | 400       |
| ?          | 1500       | 25500      | 8000      |
| interband  | 6000       | 55000      | 25000     |

**Table I.** The original model that fit the data very well, but had an oscillator centered at 1500 cm$^{-1}$ that was not well defined as originating from intraband or interband transitions. To provide a better assignment of spectral weight we split this ambiguous oscillator into two (see Table II for details). All units are cm$^{-1}$. 

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splitting of 1500 cm$^{-1}$ osc.

| $\omega_0$ | $\omega_p$ | $\Gamma$ |
|------------|------------|---------|
| 1500       | 25500      | 8000   |
| 1500       | 18000      | 8000   |
| 1500       | 18000      | 8000   |
| 1000       | 14000      | 6500   |
| 2000       | 24000      | 12000  |
| 500        | 10000      | 3500   |
| 2500       | 29000      | 14000  |
| 500        | 8000       | 1300   |
| 4000       | 3400       | 18000  |
| 500        | 7000       | 1000   |
| 6000       | 54000      | 34000  |
| 500        | 7000       | 1000   |
| 8000       | 70000      | 55000  |
| 500        | 7000       | 900    |
| 10000      | 90000      | 90000  |

TABLE II. Details of how the broad oscillator, originally centered at 1500 cm$^{-1}$, was split and its spectral weight was redistributed. All units are cm$^{-1}$.

according to the formulas

$$\frac{m^*(\omega)}{m_b} = -\frac{\omega_p^2}{\omega^2} Re\left[\frac{1}{\epsilon(\omega) - \epsilon_\infty}\right]$$  \hspace{1cm} (9)

$$\frac{1}{\tau(\omega)} = -\frac{\omega_p}{\omega} Im\left[\frac{1}{\epsilon(\omega) - \epsilon_\infty}\right]$$  \hspace{1cm} (10)

where $\omega_p$ is the full plasma frequency obtained using Fourier transform infrared spectroscopy. This parameter describes the high frequency spectral weight that represents the spectral weight from all free carriers and is not to be confused with the narrowest low frequency Drude plasma frequency, which is commonly associated with the heavy charge carriers. Further discussion of this parameter is described elsewhere in the Supplemental Material. $\epsilon_\infty$, as previously discussed, represents the contributions of all oscillators at very high frequencies. It should be noted that $\omega_p$ and $\epsilon_\infty$ used in this model are not the same as the parameters of the model used to calculate $\epsilon(\omega)$. An example of this fitting process is seen in Fig. 7.

The extended Drude model fits are essential to determining the zero frequency limit of the renormalized mass.

From the THz data alone one would not conclude that a large mass renormalization takes place. However, with the combination of the THz data AND the dc resistivity data along with an extended Drude model fit, it is possible to infer how the mass renormalization changes as one connects the THz data to the dc data. The extended Drude model applied to the two Drude oscillator fits allows us to make this connection within a model that only depends on the Kramers-Kronig transform. Of course the exact shape of the frequency dependence in the range where there is no data should not be believed, but the general dependence of going from the high mass value at low $\omega$ to the low mass at high $\omega$ will be correct as long as the Kramers-Kronig relations hold.

### Scattering rate power law fits

A full view of the scattering rate as a function of frequency for all temperatures is shown in Fig. 8. It is clear that the scattering rate is systematically suppressed as the temperature is lowered. It is difficult, when looking at all the spectra at once, to notice frequency dependence to any individual curve. However, a frequency dependence does begin to form for temperatures below $\sim 15$ K. To quantify this dependence we chose to fit the scattering rates to a simple power law $\frac{1}{\tau(\omega,T)} = \frac{1}{\tau(0,T)} + A\omega^n$. The frequency range we fit over had a noticeable influence on the exponent calculated. To account for this, we fit over a few different frequency ranges (i.e 0.2-1.25 THz, 0.2-1.5 THz, 0.2-1.75 THz) and average the calculated exponents. Table III shows the value of the exponents and error bars seen in the manuscript Fig. 3 are the average values and one standard deviation, respectively.

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FIG. 7. A screenshot of the fitting program, RefFit, used to perform the extended Drude model fits on the previously calculated optical conductivity models.

FIG. 8. Scattering rate as a function of frequency for all temperatures 1.55-250 K.
TABLE III. The exponents from fitting the scattering rate as a function of frequency using a simple power law over a few frequency ranges.

| Temp (K) | Exponent (fit range: 0.2-1.25 THz) | Exponent (fit range: 0.2-1.5 THz) | Exponent (fit range: 0.2-1.75 THz) | Average Exponent | Standard Deviation |
|----------|-----------------------------------|-----------------------------------|-----------------------------------|------------------|--------------------|
| 1.55     | 0.660                             | 1.21                              | 0.955                             | 0.227            |
| 1.75     | 0.835                             | 0.965                             | 0.916                             | 0.057            |
| 3        | 0.695                             | 0.905                             | 0.240                             | 0.111            |
| 3.75     | 1.11                              | 1.34                              | 1.30                              | 0.142            |
| 4        | 1.10                              | 1.37                              | 1.23                              | 0.109            |
| 5        | 1.00                              | 1.24                              | 1.09                              | 0.150            |
| 6        | 1.26                              | 1.30                              | 1.38                              | 0.147            |
| 8        | 1.65                              | 1.38                              | 1.72                              | 0.056            |
| 10       | 1.40                              | 1.63                              | 1.56                              | 0.113            |
| 10.5     | 1.74                              | 1.71                              | 1.77                              | 0.068            |
| 11       | 1.89                              | 1.83                              | 1.88                              | 0.038            |
| 11.5     | 1.87                              | 1.85                              | 1.87                              | 0.015            |
| 12       | 2.19                              | 1.97                              | 2.07                              | 0.092            |