I. INTRODUCTION

The electronic ground state of a strongly correlated system has remained a subject of great interest over decades. Heavy-fermion systems [1–4], such as the rare-earth intermetallics, are one of the most remarkable manifestations of systems with strong electronic correlations. The localized 4f states of the rare-earth element give rise to local magnetic moments that typically order magnetically at sufficiently low temperatures. In addition, the hybridization between 4f and conduction electrons leads to the Kondo effect [5, 6], which screens the local moments via the Kondo spin-singlet many-body states. The Kondo effect, thus, drives a part of the 4f spectral weight to the Kondo resonance near the Fermi energy \( \epsilon_F \), where it forms a band of lattice-coherent, heavy quasiparticles. These 4f electrons thus become itinerant, leading to the expansion of the Fermi volume, i.e., the \( k \)-space volume enclosed by the Fermi surface, to accommodate the extra number of indistinguishable 4f electrons in the Fermi sea. The ground state of many heavy-fermion compounds undergoes a second-order quantum phase transition [7–9] at the quantum critical point (QCP) [10]. While in the spin-wave or Hertz-Millis-Moriya scenario [11–13] of a magnetic quantum phase transition this heavy Fermi liquid undergoes a spin-density wave instability leaving the heavy quasiparticles intact, in some heavy-fermion compounds [8, 14–16] the Kondo screening gets disrupted near the QCP [17–20], and the ensuing Kondo breakdown leads to a partial collapse of the Fermi volume [8, 15–16].

The above-mentioned hybridization between the localized rare-earth 4f-electrons and the itinerant conduction electrons control the electronic properties, such as conductivity, in a profound way. At elevated temperatures, the electrons move in a broad conduction band, and infrared optical experiments on the low-energy electrodynamics of heavy-fermion compounds reveal Drude-like behavior in the optical conductivity, indicating light Fermi-liquid nature [21–26]. As the temperature is reduced below the lattice Kondo temperature \( T_K^* \), the Kondo spin scattering of conduction electrons from the rare-earth local moments and the subsequent mixing of nearly localized 4f electron and conduction electron states leads to the formation of a flat band crossing the Fermi level with drastically enhanced effective mass. As a consequence, deviations from the simple-metallic Drude behavior have been observed. These deviations are controlled by two types of phenomena. On the one hand, the enhanced quasiparticle effective mass leads to a more inert electronic response to external electric fields and tends to reduce the low-frequency conductivity. On the other hand, the Kondo correlation-enhanced density of states near the Fermi level tends to increase the conductivity. In addition, the enhanced spin-scattering rate at temperatures around \( T_K^* \), reflecting the crossover from the light to the heavy Fermi liquid, is expected to broaden the Drude peak in the optical conductivity as a function of frequency. The interplay of these latter many-body effects with the former effective mass, i.e., band-structure effect can lead to complex temperature dependence of the optical conductivity [21–24]. Up to now, it has been difficult to disentangle the correlation-induced many-body dynamics from single-particle bandstructure effect in physical observations. This is partly because the Kondo resonance and the Drude behavior in the low-frequency conductiv-
Recently, time-resolved terahertz (THz) spectroscopy has proven to be a powerful tool to coherently probe collective excitations in solids, in particular the correlation dynamics in heavy-fermion systems. We have shown that upon excitation with low-energy THz radiation, part of the correlated Kondo state can be extinguished and resurges back with a distinct temporal separation from the instantaneous conduction-electron response [28]. The delayed response bears distinct information on the Kondo correlation dynamics. This entails a rather direct measure of the quasiparticle spectral weight and the Kondo lattice temperature \(T_K^*\) of the heavy-fermion system within a single experiment [28, 29]. In short, the THz electric field creates (i) the intraband excitations, which leave the heavy quasiparticles intact and lead to an instantaneous response, and (ii) the resonant interband transitions, which break the Kondo singlets and lead to a time-delayed response. Taking advantage of this temporal separation, we can now discern the optical conductivity obtained from the instantaneous and the delayed, correlated responses. Moreover, as our technique is phase-sensitive, one can avoid invoking the Kramers-Kronig analysis while extracting the real and imaginary parts of the optical conductivity. This leads to two questions: First, does the optical conductivity obtained in our current approach from the instantaneous response, reconcile with the earlier observations from infrared experiments? Second, what is the behavior of the dynamical conductivity obtained from the delayed, correlated response?

In this Report, we address these questions by unravelling the temperature dependence of the dynamical conductivity obtained from the instantaneous response and the correlated delayed response at THz frequencies. We use the canonical \(\text{CeCu}_{6-x}\text{Au}_x\) system [30, 31], with \(x = 0\) as the heavy-fermion compound, \(x = 0.1\) as the quantum-critical compound and \(x = 1\) as the antiferromagnetic compound. It is important to note that \(\text{CeCu}_5\text{Au}\) is a stoichiometric compound where exactly one of the five inequivalent Cu sites is completely occupied by Au [32]. A Pt mirror is used as the reference sample for all our measurements. We show that the dynamical conductivity obtained from the instantaneous response of \(\text{CeCu}_6\) agrees well with previous observations [24, 25], i.e., it shows a metallic Drude-like low-frequency behavior at high temperatures that develops into a heavy-Fermi-liquid Drude response at \(T \leq T_K^*\). In contrast, the dynamical conductivity obtained from the delayed response does not show any Drude-like free-electron behavior. Such a non-trivial deviation corroborates that the delayed response is a unique signature of the correlated electronic states.

### III. RESULTS AND DISCUSSION

We first identify the trivial THz reflexes generated by the optical components in order not to confuse them with the THz signal generated from the \(\text{CeCu}_{6-x}\text{Au}_x\) samples. A measurement of all these THz reflexes is provided elsewhere [28]. The earliest of these artefacts appear at a delay time of \(10\) ps, outside the range used in our analysis for the optical conductivity. As an example, we show the time trace of the THz electric field reflected from the \(\text{CeCu}_{5.9}\text{Au}_{0.1}\) sample at \(20\) K in Fig. 1(a). The complete signal can be divided into two parts: (i) the instantaneous response, shown in Fig. 1(c)), from \(t = -4\) ps to \(t = 3.5\) ps (blue-shaded region in Fig. 1(a)), and (ii) the delayed response, shown in Fig. 1(d) from \(t = 3.5\) ps to \(t = 8.5\) ps (red-shaded region in Fig. 1(a)). As mentioned previously, these two temporally separated responses stem from two excitation processes induced by the THz radiation, the intraband excitations and the interband transitions. The schematic in Fig. 1(b) shows the hybridized band, elucidating the two processes. The instantaneous response originates from the stimulated single-particle response of quasiparticles within the conduction band, in other words, intraband excitations.
These intraband scattering processes leave the heavy quasiparticles intact and are expected to show a Fermi-liquid Drude response in the optical conductivity. The delayed response, on the other hand, originates from the interband transitions between the hybridized heavy and light parts of the conduction band that restores the Kondo state after the stimulated breaking (by the THz pulse) of the Kondo singlets. In CeCu$_{6-x}$Au$_x$, the delayed response appears around 6 ps and agrees well with the Kondo quasiparticle lifetime $\tau_K$ [25]. Since this response comes solely from the Kondo correlation effect, we would not expect to observe a Drude response, but rather a low-frequency Kondo resonance with a Lorentzian lineshape. The CEF resonances play a special role in heavy-fermion compounds since at high temperatures the CEF-induced high-energy scale governs the spectral weight near the Fermi level [29]. At low temperature, they appear as a set of resonances with narrow spectral width [33, 34] and, hence, long coherence time. Their signature is therefore primarily found in the delayed pulse. With increasing temperature, the resonances merge into a single peak of increased spectral width and, hence, short coherence time so that their weight shifts from the delayed pulse into the wiggles of the instantaneous pulse [29].

To obtain the dynamical conductivity from the two response windows, the THz time traces are first Fourier-transformed (FT) to obtain the complex THz spectra. Figures 2(a) and 2(b) show the FT spectral amplitude of the instantaneous and the delayed responses for CeCu$_6$ at two different temperatures (81 K, red; and 2 K, blue), respectively. The complex reflectivities are obtained by dividing the respective spectra by the Pt mirror reference spectrum (black curve in Fig. 2(a)). The corresponding reflectivities are shown in Figs. 2(c-d). The reflectivity of CeCu$_6$ at 2 K clearly shows strong deviation from the one at 81 K, particularly at low frequencies ($\nu \leq 0.75$ THz) and at higher frequencies ($1.2$ THz $\leq \nu \leq 2.2$ THz). Figures 2(e) and 2(f) show the real part of the THz conductivity evaluated from the complex reflectivity at 81 K and 2 K, for the instantaneous and delayed responses, respectively. The green-dashed curves in Fig. 2(e) show the low-frequency Drude fit according to the relation $\sigma_R(\nu) = \sigma_{DC}/(1 + \omega^2 \tau_{tr}^2)$, where $\sigma_R$ is the real part of the conductivity, $\sigma_{DC}$ is the DC conductivity, $\omega = 2\pi \nu$ is the angular frequency and $1/\tau_{tr}$ is the transport relaxation rate. The Drude fit is carried out only for the range from 0.2 to 0.75 THz, with $\sigma_{DC}$ and $\tau_{tr}$ being the only fitting parameters. The fit was pinned to the data at the highest frequency, ignoring all spectral features between 1 to 1.8 THz, to illustrate the physically meaningful situation that develops at low frequencies, i.e., $\nu \leq 0.75$ THz. The DC value of the optical conductivity is obtained from the extrapolation of the finite frequency results to zero frequency [21, 24, 37]. Because of the large overall systematic error in the absolute value of the DC conductivity and the scattering rate, we revert to relative values for their temperature dependence by taking the ratio with respect to the high temperature values (here, with $T = 145$ K for CeCu$_6$) in the ensuing discussions.

In Fig. 2(e), we find that the instantaneous low-frequency
Figure 2. (a,b) The spectra obtained by Fourier transforming the instantaneous and delayed THz responses of the heavy-fermion compound (CeCu$_6$) at sample temperatures of 81 K and 2 K. The reference spectrum is the signal from the Pt mirror. (c,d) The reflectivity curves at 81 K and 2 K, corresponding to the instantaneous and delayed THz responses. (e) The real part of the conductivity obtained from the instantaneous response shows a heavy-Fermi-liquid behavior (green dashed curve) in the low frequency region at 2 K and a peak corresponding to the CEF resonance at higher frequencies. (f) The real part of the conductivity obtained from the correlated delayed response shows the low-frequency Kondo resonance and the high-frequency CEF resonance. A smooth transfer of the correlated spectral weight from the CEF resonance at high temperatures to the Kondo resonance at low temperatures is highlighted by the arrows in the yellow-shaded regions.

Conductivity at 81 K qualitatively follows the metallic Drude response and, as the temperature is reduced to 2 K, the conductivity deviates from the simple-metallic Drude nature to a heavy-Fermi-liquid Drude behavior, which is characterized by a decreased electronic relaxation rate (see Fig. 3(d)) and an enhanced effective mass and density of states near the Fermi level. The gradual appearance of the peak around 1.5 THz in the optical conductivity below 40 K (see Fig. 3(a)) indicates the formation of a narrow quasiparticle band in the vicinity of the Fermi level, in good agreement with de Haas-van Alphen measurements [28]. Thus the changes observed in the low-temperature data result from the interaction of the delocalized $4f$ electrons, i.e., the formation of heavy quasiparticles. The conductivity peak at 1 THz is the free-carrier response that corresponds to the 1 THz intensity peak of the incident THz wave (see the reference spectrum in Fig. 2(a)). Besides, the spectral response from the Pt mirror is not exactly similar to the spectral response of the incident THz beam. This can lead to such spectral feature, when the spectrum of Pt mirror is used as the reference spectrum in our analysis. Note that this feature at 1 THz is rather temperature-independent and is present in all samples measured and hence we ignore it in the rest of our discussion.

In contrast, the optical conductivity obtained from the delayed pulse around 6 ps shows a distinct response arising from the break-up and restoration of the correlated Kondo state, see Fig. 2(f). Within the spectral range, two distinct features signaling the heavy-fermion state are observed. First, we have a quasi-Lorentzian peak at low frequencies, $\nu \leq 0.75$ THz, that is fundamentally different from the Drude response (green-dashed curve in Fig. 2(e)). The spectral weight of this low-frequency resonance peak increases as the temperature is reduced due to the increasing Kondo weight. The width of this resonance peak resembles the Kondo energy scale, which remains constant though the spectral weight reduces towards low temperatures, in agreement with our previous findings [28, 29]. The second feature is the broad peak centered at 1.75 THz that corresponds to the first CEF-excited state in the CeCu$_6$ system [39-41]. The spectral weight of this peak reduces as the temperature is reduced. Note that the occupied spectral weight of the CEF resonance at higher temperatures is smoothly transferred to the Kondo resonance at low temperatures (indicated schematically by the yellow-shaded regions in Fig. 2(f)) as the CEF occupation is frozen out. This observation agrees with our previous results, drawn from the time-domain, on the smooth crossover from a high-temperature to a low-temperature Kondo scale [29]. Also note that the low-temperature transport relaxation rate extracted from the Drude fit in Fig. 2(e) as $1/\tau_{tr} \approx 0.25$ THz (corresponding to 2 K) is significantly larger than the Kondo scale $\tau_K \approx 8$ K and of the width of the low-frequency peak in the delayed response of Fig. 2(f). This indicates that the low-frequency instantaneous response is governed by intraband transport scattering processes and concatenated Drude behavior (Fig. 2(e)), while the zero-frequency peak in the delayed response (Fig. 2(f)) is a signature of the Kondo resonance alone.

A complete temperature evolution of the THz conductivity obtained from the instantaneous signal of the heavy-fermion compound, CeCu$_6$, is shown in Fig. 3(a-b). From the low-frequency Drude fitting, mentioned above, we could retrieve the temperature dependence of the DC conductivity and hence the resistivity as well as the scattering rate, shown in Figs. 3(c) and 3(d), respectively. We see that at very high temperatures, the real part of the THz conductivity is dominated by the Drude-like free-electron response, as mentioned earlier. This behavior, however, develops slowly into a heavy-Fermi-liquid Drude response as the temperature is lowered below 10 K, characterized by the reduced scattering rate and increased effective mass, with a $T^2$ dependence of resistivity featuring the Fermi-liquid intraband quasiparticle-quasiparticle scattering (see red-dashed line in Fig. 3(a)). The gray-shaded region in Fig. 3(a) shows the evolution of the first CEF resonance. Similar low-temperature behavior was observed earlier for CeCu$_6$ from infrared measurements [21]. This certifies a strong
The real part of the optical conductivity of CeCu$_6$ obtained from the instantaneous response as a function of sample temperature. The green-shaded region indicates the low-frequency region where the Drude-like free-electron response at high temperatures deviates to a heavy-Fermi-liquid behavior at low temperatures. The grey-shaded region indicates the region where the first CEF resonance starts appearing as the temperature is reduced below 10 K. The green-dashed lines are the Drude fitting curves. (b) The low-frequency region in (a) for a few selected temperatures. (c,d) The change in DC resistivity and scattering rate, obtained from low-frequency Drude fit, normalized to the value at 145 K. (e) The real part of the optical conductivity of CeCu$_6$ obtained from the delayed correlated response as a function of sample temperature. Being a pure correlated response, a single peak Lorentz function (blue-dashed curves) with a fixed linewidth of 0.12 THz is used to fit the low-frequency region (green-shaded region). The CEF resonance (in grey-shaded region) shows the characteristic temperature dependence where the spectral weight from the CEF resonance is transferred to the Kondo resonance on reducing the temperature. (f) The temperature-dependent Lorentz functions used to reproduce the low-frequency conductivity response (all curves are colored as in (e)). (g) The temperature-dependent amplitude of the single peak Lorentz functions. Note that these reproduce the heavy-fermion Kondo-spectral-weight behavior of Ref. [28].

The $f$-character of the CEF bands at the Fermi level stems from the hybridization between the conduction electrons and the 4$f$ electrons. The temperature evolution of the conductivity obtained from the delayed response (Fig. 3(e)) is striking. The low-frequency behavior cannot be reproduced using the Drude-like response of width $1/T_{\tau_r}$ at any temperature. However, we can fit a single peak Lorentz function of smaller width $\Gamma$ (blue-dashed lines in Fig. 3(e)). The function is defined as $A(\nu) = A(T)\Gamma/[(\nu - \nu_0)^2 + (\Gamma/2)^2]$, where $A(T)$ is the temperature-dependent spectral weight, $\nu_0$ represents the low-frequency Kondo resonance and $\Gamma$ should correspond to the Kondo energy scale, see discussion above. The temperature dependence of the fitted Lorentz functions is separately plotted in Fig. 3(f). The temperature dependence of $A$, plotted in Fig. 3(g), reproduces qualitatively the temperature dependence of the Kondo weight in CeCu$_6$, obtained by integrating the spectral weight of the delayed response. [28] [29]. At high temperatures there is only one peak observed at around 1.75 THz which corresponds to the first CEF resonance. As the temperature is reduced below 300 K the spectral weight of this peak first increases down to 40 K, revealing the high-temperature Kondo scale [29]. On decreasing the temperature further, the spectral weight of the peak reduces while the spectral weight of the low-frequency Kondo resonance increases (see Fig. 3(f) for clarity). This indicates that there is a smooth transfer of the spectral weight from the high-temperature CEF resonance to the low-temperature Kondo resonance, as mentioned before. Further, we observe that roughly $A(T \rightarrow 0) \approx 0.7 \times A(T = 35 K)$, in very good agreement with Ref. [28].

In the case of the quantum-critical sample, CeCu$_{5.9}$Au$_{0.1}$, the optical conductivity obtained from the instantaneous response (see Fig. 4(a) and 4(b)) shows a very similar high-temperature Drude behavior as observed in the case of the heavy-fermion compound, CeCu$_6$. However, for $T < 10 K$ the low-frequency part of the conductivity shows deviation from the Drude behavior that presumably reflects the onset of the non-Fermi-liquid behavior [31] [42] near the quantum-critical point (see the yellow-shaded region in Fig. 4(b)). From the low-frequency Drude fitting, the temperature dependence of the DC conductivity as well as the scattering rate are obtained as explained above and are shown in Figs. 4(c) and 4(d),...
respectively. The resistivity behavior is distinctly different from CuCu₆ for $T < 10 \text{ K}$. While CeCu₆ has a $T^2$ resistivity dependence (see Fig. 3(a)), CeCu₅.₉Au₀.₁ shows a resistivity compatible with linear-in-$T$ dependence for $T < 10 \text{ K}$ (see Fig. 3(b)), in line with the linear DC resistivity found below 1 K [31]. This difference can be associated with the fact that, while CeCu₆ is a heavy-Fermi-liquid compound at these temperatures, CeCu₅.₉Au₀.₁ is quantum-critical. The temperature dependence of the conductivity from the delayed response (see Fig. 3(e)), is once again striking. A single peak Lorentz function is used, as before, to model the low-frequency conductivity behavior, see Fig. 3(f). We find that the spectral weight of the low-frequency Kondo resonance increases as we lower the temperature, reaches a maximum and then smoothly reduces as we lower the temperature below $T^*_K$. This corroborates that the sample is entering into the quantum-critical regime where the Kondo quasiparticle weight almost completely dissappears, again in agreement with Ref. [28], see Fig. 4(g). Further, the high-frequency CEF resonance also reduces its spectral weight as we lower the temperature, quite similar to what is observed in CeCu₆. These results reproduce our previous work [28, 29], where our analysis was carried out purely from the responses in the time domain. Thus, we consistently agree with the physics of the Kondo-correlated part from the delayed, echo-like response in both time and the frequency domain. The spectral weight in the time domain corresponds to the optical conductivity in the frequency domain. Yet in addition to the earlier results, a proper identification of the purely correlated response in the time-domain enables us to discern (i) the Kondo resonance from the Drude behavior and (ii) the effect of CEF states on the high-energy Kondo scale.

Figure 6(a) shows the temperature evolution of the optical conductivity for the antiferromagnetic compound, CeCu₅.₉Au₀.₁, obtained from the instantaneous response. Remarkably, the antiferromagnetic compound does not show a delayed correlated response at any temperatures [28]. Apart from the conductivity peak at 1 THz that, as mentioned before, is present in all samples, there seems to be an underlying broad low-frequency feature that blue-shifts as we lower the temperature (see Fig. 6(b) for clarity). Such an underlying feature makes it difficult to perform a conclusive Drude-fit of the experimental data. We thus
speculate about the origin of our observations as follows: It is known from previous temperature-dependent nuclear magnetic resonance studies that the nuclear spin-lattice relaxation mechanism in CeCu$_5$Au is rather anisotropic. For crystallographic orientations orthogonal to c-axis (as in our current configuration), nuclear spin-spin coupling prevails, mediated by conduction electrons [43]. This implies that temperature-dependent fluctuations in the spin relaxation time can significantly modify the electronic scattering times, thus leading to the observed temperature-dependent complex low-frequency conductivity response. Taking a closer look at the high-frequency response of the conductivity at 2 THz, it can be argued that a resonance structure appears as the temperature is lowered, possibly indicating the emergence of CEF correlated states. This feature is caused by incoherent Kondo signatures present at all temperatures in our earlier time-domain analysis (see supplement of Ref. [28]). Such incoherent Kondo signatures have also been observed in previous transport and thermodynamic measurements [44], despite the dominance of RKKY interaction over the Kondo interaction in the antiferromagnetic compound [20] that entirely suppresses the coherent Kondo state [28]. Further, it can also be seen in Fig. 6(b) that the low-frequency-averaged THz conductivity of the antiferromagnetic sample increases as the temperature is reduced. This implies that the resistivity decreases as the sample enters the antiferromagnetically-ordered phase, shown in Fig. 6(c), which agrees with the electrical transport measurements [32, 34, 35]. Note that the difference in the absolute value of the resistivity measured in earlier transport measurements [32] compared to our measurements stems from two main reasons. Firstly, it is due to the difference in the probing techniques and secondly, because in the earlier transport studies, the DC values of the resistivity were measured, while in our study, we consider the frequency-averaged resistivity values at THz frequencies.

IV. CONCLUSION

We have explored the temperature dependence of the dynamical conductivity at THz frequencies for the canonical CeCu$_{6-x}$Au$_x$ system. Terahertz excitation creates both intraband excitations as well as resonant interband transitions between the hybridized heavy and light parts of the conduction band. In the heavy-Fermi-liquid phase, CeCu$_6$, the intraband excitations lead to Drude response embedded within the instantaneous response. The interband transitions create a time-delayed response, featuring Kondo correlations. We have systematically separated the conductivity responses of the instantaneous signal from the delayed correlated signal. The instantaneous response of the heavy-fermion sample and the quantum-critical sample have a simple metallic Drude low-frequency behavior at high temperatures that develops into heavy-Fermi-liquid Drude and non-Fermi-liquid behavior, respectively, below the Kondo lattice temperature. In contrast, the optical conductivities obtained from the correlated, delayed response do not show Drude-like behavior at any temperatures, neither for CeCu$_6$ nor CeCu$_{5.9}$Au$_{0.1}$. We have observed a smooth transfer of the conductivity spectral weight from the CEF resonance at high temperature to the Kondo resonance at low temperature. By contrast, in CeCu$_{5.9}$Au$_{0.1}$ the spectral weight
of the Kondo resonance continuously vanishes below $T^*_K$, implying that the sample undergoes a Kondo breakdown as we approach the QCP, reconciling with our previous analysis in the time-domain [28, 29]. On the other hand, the antiferromagnetic compound, CeCu$_6$Au$_1$, shows a complete absence of coherent Kondo states. Our time-resolved and phase-sensitive measurements open up the possibility to determine the optical conductivity without resorting to Kramers-Kronig analysis, and, thus, to temporally separate the purely correlated response in the time domain. This allows to distinctly identify different contributions to the dynamic conductivity. We not only reconcile the previous results obtained from infrared optical measurements, but also show a very general way to uniquely discern the dynamic processes in Kondo systems.

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