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Evidence for Correlation Effects in Noncentrosymmetric Type II Weyl Semimetals

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Topological fermions have mainly been addressed in the limit of weakly correlated systems, while the realisation of Kondo-like physics arising from Dirac/Weyl fermions and implying strong correlations is rare. The noncentrosymmetric Weyl fermion $R\text{AlGe}$ compounds (with $R = \text{La}$ and Ce) provide a promising playground for revealing the impact of electronic correlations driven by f -electron states on Weyl fermions. Here, we tackle their charge dynamics as a function of temperature. Besides spotting typical optical signatures of a type II Weyl semimetal in both materials, we discover that electronic correlations at low temperatures renormalise the non-trivial bands hosting the Weyl nodes and lead to a reduction of the Fermi velocity as well as an enhancement of the charge carriers effective mass in CeAlGe with respect to LaAlGe , both being a fingerprint of a so-called Weyl-Kondo system.

The interplay of topology with electronic correlations is acquiring increasing interest in the on-going solid state physics research and leads to concepts of paramount importance for the understanding of novel functionalities in quantum materials^{1–3}. Charge, spin and orbital degrees of freedom of electrons in solids are intimately related to and shaped by the topological nature of the host material. For instance, a topological state of relevance in condensed matter derives from the split of the doubly degenerate, massless Dirac states into a pair of Weyl nodes, as consequence of broken time-reversal or space-inversion symmetry^{4,5}. On the other hand, electronic correlations are a basic asset for materials featuring a large variety of astonishing properties⁶. The link between topology and many-body ground states has been explored only recently, with proposals advancing topological Weyl superconductors⁷ and charge-density-wave states in Weyl semimetals^{8–10} as well as Weyl-Mott insulators¹¹.

Another possible consequence of electronic correlations is exemplary epitomised by the Kondo materials, for which the coupling between f -electrons and conduction-band states notably leads to strong renormalisation of the electronic bands in the vicinity of the Fermi energy (E_F), then possessing f -character and displaying the formation of heavy electrons at low temperatures (T)¹². Several topological phases are emerging, when the single-particle band structure is modified by Kondo-like electron-electron correlations, within concepts ranging from hybridisation-driven heavy Weyl-Kondo semimetals^{13,14} to Kondo effects of magnetic impurities in Dirac and Weyl systems^{15,16}.

While CeRu_4Sn_6 was proposed as a promising heavy Weyl fermion candidate¹⁷, $\text{Ce}_3\text{Bi}_4\text{Pd}_3$ ^{13,18,19} and YbPtBi ²⁰ are to date among the rare experimental realisations of a Weyl-Kondo compound. This motivates the investigation of condensed matter systems provid-

ing the tunability of the Kondo coupling in an environment of Weyl states, upon switching-on the presence of f -electrons.

In this respect, the family of the noncentrosymmetric $R\text{AlGe}$ ($R = \text{rare earth}$) compounds²¹ is a suitable arena in order to advance our knowledge. First of all, $R\text{AlGe}$ span all varieties of Weyl semimetals depending on the choice of R ; namely, the standard type I with point-like Fermi surface and the initially overlooked type II arising from the tilting of the Weyl cones with the Fermi level along the line boundaries of electron and hole pockets^{21–23}. Second, LaAlGe ($4f^0$ orbitals) is nonmagnetic, while first-principles calculations prefigure CeAlGe (with $\text{Ce } 4f^1$) and PrAlGe (with $\text{Pr } 4f^2$) to order ferromagnetically²¹. Even though the magnetism in CeAlGe is matter of debate^{24–28}, it turns out that the prediction of Weyl nodes in $R\text{AlGe}$ as well as their band structure at low energies seem to be robust enough and less dependent on the details of the magnetism²¹. We note in passing that the presence of both time-reversal and space-inversion broken symmetry should favour spin current without concomitant charge current²⁹, an aspect of both fundamental and technical interest. Third, it has been postulated^{13,14,20} that the Kondo coupling between f -electrons and bands hosting the Weyl nodes close to E_F could lead to a greatly reduced Fermi velocity (v_F) and to the enhancement of the itinerant quasiparticles effective mass (m^*/m_e) compared to the bare band values at low T .

The signatures of Weyl fermions as well as m^*/m_e and v_F are accessible from the excitation spectrum. In this letter, we exploit the T dependence of the real part ($\sigma_1(\omega)$) of the optical conductivity of LaAlGe and CeAlGe , obtained over a very broad energy interval from the far-infrared (FIR) up to the ultra-violet (UV) spectral range³⁰. While a variety of representative Dirac and Weyl semimetals were recently addressed from the

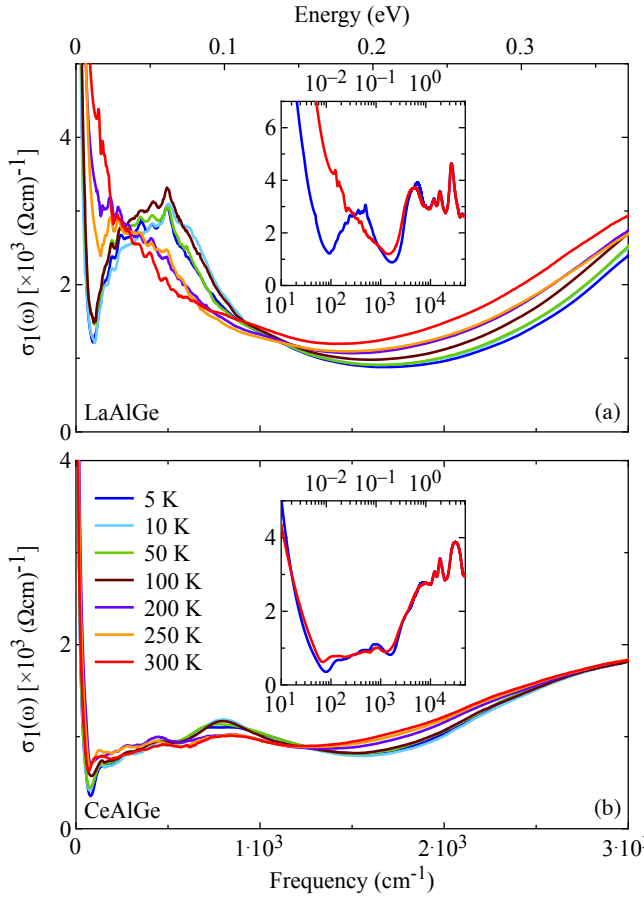


Figure 1. (color online) T dependence of the real part $\sigma_1(\omega)$ of the optical conductivity of (a) LaAlGe and (b) CeAlGe from the far-infrared up to the mid-infrared frequency range ($1 \text{ eV} = 8.06548 \times 10^3 \text{ cm}^{-1}$). The insets show $\sigma_1(\omega)$ in the whole measured energy interval at 300 and 5 K (please note the logarithmic energy scale).

perspective of their optical response (Ref. 31 for a recent review), our results display the characteristic interband feature in $\sigma_1(\omega)$ of the less scrutinised type II Weyl semimetals as well as a depleted v_F and a remarkably enhanced quasiparticles m^*/m_e at low T in CeAlGe than in LaAlGe. Our inferences are thus of guiding relevance when establishing Kondo-like physics in these noncentrosymmetric (type II) Weyl semimetals.

We start off with a survey of the relevant data in Fig. 1, which displays the T dependence of $\sigma_1(\omega)$ below 3000 cm^{-1} for LaAlGe and CeAlGe, while its insets compare their spectra at 5 and 300 K over the whole measured spectral range. We refer to the Supplemental Material for more details on the original data³². Besides the T -independent absorptions at UV energy scales, which are almost identical in both compounds, we encounter a relevant T dependence in $\sigma_1(\omega)$ from FIR up to the visible spectral range (i.e., $\omega < 10^4 \text{ cm}^{-1}$). In agreement with the dc transport results, LaAlGe and CeAlGe are metallic, as pointed out by their Drude-like zero energy mode

which narrows with decreasing T . The T dependence at FIR and mid-infrared (MIR) energies is rather distinct between the two investigated materials. In LaAlGe (Fig. 1(a)) there is a depletion of $\sigma_1(\omega)$ below 300 cm^{-1} , signalling a spectral weight (SW) reshuffling⁴⁵ from FIR towards high frequencies, mainly up to 1000 cm^{-1} . A SW removal is also observed in the MIR energy interval (i.e., $1500\text{--}4000 \text{ cm}^{-1}$) with accompanying accumulation at even higher energies, up to the near-infrared (NIR) spectral range (i.e., $4000\text{--}8000 \text{ cm}^{-1}$). We note a qualitative similarity with the SW redistribution in the optical response of the noncentrosymmetric, transition-metal monpnictide TaAs, for which type I Weyl states as consequence of broken space-inversion symmetry⁴⁶ as well as some reminders of nodal semimetals³¹ were actually envisaged. For CeAlGe (Fig. 1(b)), the SW reshuffling at FIR-MIR energy scales is less pronounced, while something equivalent to LaAlGe happens at MIR-NIR energies. Otherwise, the MIR optical conductivity is quite featureless, just displaying a moderate increase with frequency in the interval $100\text{--}1000 \text{ cm}^{-1}$, thus above the shallow minimum in $\sigma_1(\omega)$ around 100 cm^{-1} , right at the onset of the Drude resonance. The trend in the charge dynamics from LaAlGe to CeAlGe is however different from what has been encountered in $\text{Nd}_2(\text{Ir}_{1-x}\text{Rh}_x)_2\text{O}_7$, for which the ground state undergoes a putative crossover from a narrow-gap Mott insulator to a Weyl semimetal and then to a correlated metal upon increasing x ^{47–49}.

As shown in SM³², $\sigma_1(\omega)$ can be well decomposed within the phenomenological Drude-Lorentz approach³⁰, of which we specifically highlight the reproduction of the metallic contribution ($\sigma_1^{\text{Drude}}(\omega)$, insets of Fig. 2). The zero-energy mode in $\sigma_1(\omega)$ is well accounted for by two Drude terms (Fig. S5 in SM³²). By subtracting those calculated Drude intraband components from the total $\sigma_1(\omega)$ at each T , we can better emphasise its interband contribution (i.e., $\Delta\sigma_1(\omega) = \sigma_1(\omega) - \sigma_1^{\text{Drude}}(\omega)$), shown in Fig. 2 at 5 K. In both cases, $\Delta\sigma_1(\omega)$ presents two distinct intervals (colored shaded areas in Fig. 2), characterised by (quasi) linear frequency dependences, yet with different slopes. Even though the intervals for the linearities in $\Delta\sigma_1(\omega)$ are rather small, the encountered behaviour is definitely different from the typical optical response of type I Weyl semimetals³¹ and corresponds to the expected characteristic features describing the interband contribution in $\sigma_1(\omega)$ for type II. Indeed, it has been theoretically shown that the tilting of the Dirac cone within a single band framework considerably affects the overall shape of the excitation spectrum⁵⁰. In the case of type II upon increasing the frequency, there is first a (quasilinear) sharp, steep onset of $\Delta\sigma_1(\omega)$, occurring at $\omega > \Omega_1$ (turquoise area in Fig. 2), then followed by a linear frequency dependence of $\Delta\sigma_1(\omega)$ at $\omega > \Omega_2$ (green area in Fig. 2). The latter has a smaller slope, such that it induces a kink in $\Delta\sigma_1(\omega)$ and it does not extrapolate to the origin in the limit $\omega \rightarrow 0$ (red dashed line in Fig. 2), as it is instead predicted in the optical conductivity for type I Weyl semimetals⁵⁰. The tilting of

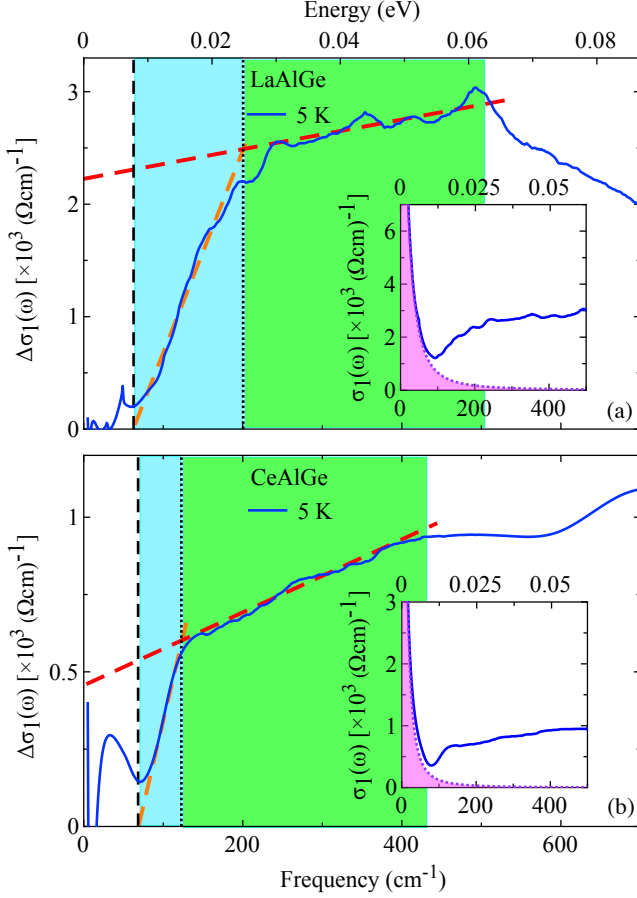


Figure 2. (color online) Interband contribution $\Delta\sigma_1(\omega) = \sigma_1(\omega) - \sigma_1^{Drude}(\omega)$ at 5 K for (a) LaAlGe and (b) CeAlGe, obtained by subtracting the calculated Drude intraband ($\sigma_1^{Drude}(\omega)$) component from the total $\sigma_1(\omega)$ (see insets). The thick dashed lines emphasise the linear frequency dependences of $\Delta\sigma_1(\omega)$, encountered in selected energy intervals (colored shaded areas). The vertical dashed and dotted lines mark the characteristic frequencies Ω_1 and Ω_2 , which define the onset of linearities in $\Delta\sigma_1(\omega)$ ^{32,50}. The insets highlight $\sigma_1(\omega)$ at FIR energies together with the contribution $\sigma_1^{Drude}(\omega)$ obtained within the two-Drude approach (i.e., sum of the narrow (D1) and broad (D2) Drude terms, insets of Fig. S5 in SM³²). A similar procedure was applied at all T .

the Weyl cones can be quantified by the tilting parameter $\hat{w}$ (Fig. 3), which is significantly larger than one in both materials³², thus alluding to an overtilted regime⁵⁰. The optical response devoid of its metallic term hence allows the most direct evidence of type II Weyl semimetals and is pertinent when confirming the predictions of de-voted band structure calculations, signalling the presence of so-called tilted (W_2) Weyl cones located at E_F in both materials²¹. This is in contrast to previous attempts on WTe₂^{51–53}, whose Weyl states are however well above E_F and $\sigma_1(\omega)$ is dominated by the (Drude) intraband component. Furthermore, since here the trivial bands are quite negligible at E_F (besides some small pockets in LaAlGe) and interband transitions involving them prosper above

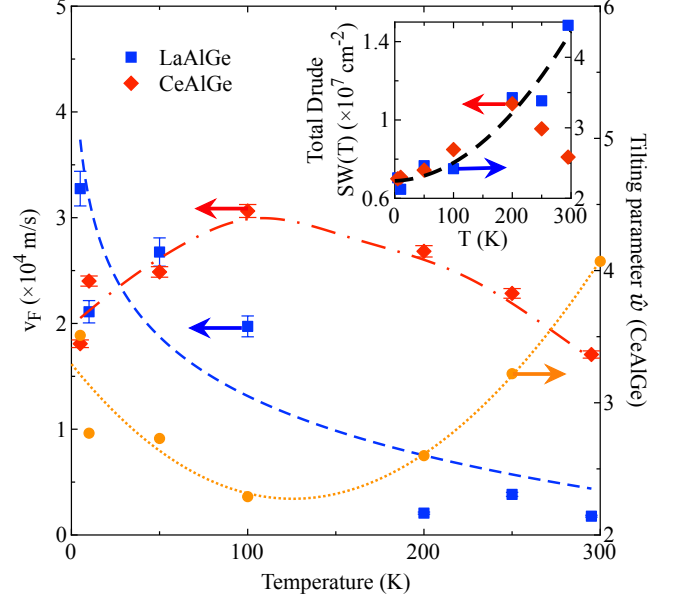


Figure 3. (color online) T dependence of the Fermi velocity v_F , extracted from $\Delta\sigma_1(\omega)$ (Fig. 2). The dash-dot line for CeAlGe is a guide to the eyes, while the dashed one for LaAlGe corresponds to the functional $v_F \sim A + B \ln(1/T)$ (see text)⁵⁵. The tilting parameter (i.e., $\hat{w}$ in Eq. S3 and in Table I in SM³²) is shown for CeAlGe, as well⁵⁰. Inset: T dependence of the total Drude SW (i.e., $\omega_p^2 = \omega_{p,D1}^2 + \omega_{p,D2}^2$, $\omega_{p,Di}$ being the plasma frequencies of D1 and D2 Drude terms³²), which follows a T^2 behaviour (thick dashed line) below 200 K for both compounds^{56,57}.

1000 cm⁻¹²¹, our data even overrule possible ambiguities in the interpretation of the kink in $\Delta\sigma_1(\omega)$, driven by the tilting of the Weyl cones, which could arise when several trivial bands close to E_F interplay with the non-trivial ones, like in Te⁵⁴.

The slope of $\sigma_1(\omega)$ at $\omega > \Omega_2$ turns out to be inversely proportional to v_F . We use the formalism of Ref. 50, in order to infer v_F (for details we refer to SM³²), which is shown in Fig. 3 as a function of T . We obtain an overall increase of v_F with decreasing T in LaAlGe, while for CeAlGe v_F displays a broad maximum upon approaching 100 K, below which it evidently decreases. The low T values of v_F in LaAlGe, despite lacking momentum resolution, are very much compatible on the average with the estimation from the angle-resolved-photoemission-spectroscopy (ARPES) spectra⁵⁸ and are typical for weakly interacting Weyl semimetals^{59,60}. The T dependence of v_F in LaAlGe quite agrees with the expectations for a two and three dimensional Dirac material within the Hartree-Fock approximation for which a $\ln(1/T)$ behaviour has been foreseen (dashed line in Fig. 3)⁵⁵. Such a logarithmic divergence is supposed to vanish if correlation effects are taken into account⁶¹. We propose that the decreasing v_F below ~ 100 K in CeAlGe (Fig. 3) hints at the progressive onset of correlation effects, similar to observations in YbPtBi²⁰ as well as in nodal-

line Dirac semimetals⁶². Therefore, the broad maximum²⁷⁰ in $v_F(T)$ of CeAlGe at 100 K (Fig. 3), coincident with²⁷¹ the prominent change of slope in the dc transport prop-²⁷² erties (Fig. S3 in SM³²) at the same T , may define the²⁷³ T scale for the onset of correlation effects^{6,12,63,64}. A²⁷⁴ similar and possibly correlations-related non-monotonic²⁷⁵ T dependence is encountered in $\hat{w}$ of CeAlGe (Fig. 3), as²⁷⁶ well³². Although the role of correlation effects is not yet²⁷⁷ fully disentangled in the Weyl semimetal YbMnBi₂^{65,66}²⁷⁸ and the linear frequency dependences of its optical con-²⁷⁹ ductivity look different than in the title compounds, it²⁸⁰ is worth pointing out that its v_F is also suppressed with²⁸¹ decreasing T ⁶⁵.²⁸²

So far, we centred our discussion around the interband²⁸³ contribution to the optical response (Figs. 2 and 3). In²⁸⁴ order to further elaborate on the impact of correlation²⁸⁵ effects, we now approach the intraband part of $\sigma_1(\omega)$.²⁸⁶ It is a generic property of correlated materials to dis-²⁸⁷ play a strongly reduced Drude SW with respect to the²⁸⁸ (non-interacting) band theory calculations^{67,68}. More-²⁸⁹ over, theoretical calculations suggest that in Dirac and²⁹⁰ Weyl semimetals there is a SW transfer from the Drude²⁹¹ to the interband component of $\sigma_1(\omega)$ upon lowering T .²⁹² and the Drude SW itself undergoes a T^2 dependence^{56,57}.²⁹³ Such a Drude SW suppression at low T fairly applies to²⁹⁴ our findings below 200 K in both compounds (inset of Fig.²⁹⁵ 3 and SM³²) and is also in agreement with experimental²⁹⁶ observations in other topological materials^{46,48,62,69,70}.²⁹⁷ Furthermore, the coefficient of the T^2 dependence of the²⁹⁸ Drude SW is inversely proportional to v_F ^{56,57}, thus al-²⁹⁹ lowing its average estimation from the intraband contri-³⁰⁰ bution to $\sigma_1(\omega)$ and a comparison with the v_F interband³⁰¹ values (Fig. 3). For CeAlGe, the resulting intraband³⁰² $v_F \sim 8.1 \times 10^4$ m/s is considerably larger than the in-³⁰³ terband one at any T (Fig. 3). A discrepancy in the³⁰⁴ estimation of v_F from both intra- and interband contri-³⁰⁵ bution to the optical conductivity was likewise encoun-³⁰⁶ tered in the correlated Nd₂Ir₂O₇ semimetal⁴⁸. This may³⁰⁷ indicate that correlation effects, related to heavy Bloch³⁰⁸ states at low T (see below), are not properly considered³⁰⁹ in the approach of Refs. 56 and 57 for the T dependence³¹⁰ of the Drude SW and caution is needed when extracting³¹¹ sensitive quantities from it. On the contrary, the intra-³¹² band $v_F \sim 5 \times 10^4$ m/s for LaAlGe rather agrees with its³¹³ estimation at low T from $\Delta\sigma_1(\omega)$ (Fig. 3), since correla-³¹⁴ tion effects are indeed not supposed to be relevant. For³¹⁵ the sake of completeness, it is worth mentioning that re-³¹⁶ cent measurements of the T - and angular-dependent de-³¹⁷ Haas-van Alphen oscillations in LaAlGe⁷¹ nevertheless³¹⁸ imply very small cyclotron masses, very high v_F (up to³¹⁹ two orders of magnitude than the low T values in Fig.³²⁰ 3) and consequently strongly reduced electronic specific³²¹ heat, the latter still not compatible with the measured³²² quantity^{71,72}. This however signals that not all portions³²³ of the Fermi surface detected by quantum oscillations do³²⁴ contribute to the thermodynamic properties, which we³²⁵ address next in connection with the optical data.

The specific heat ($C(T)$) measurements as a function³²⁶

of T and magnetic field in CeAlGe are indeed very in-
strumental towards elucidating the presence and role of
electronic correlations, mediated by f -electrons. The
 $C(T)$ data at zero and low fields display a peak at
the magnetic transition at about 5 K, which is progres-
sively washed out and then transformed into a broad
(Schottky-type) feature upon increasing the magnetic
field^{26,72}. Such a behaviour is reminiscent of the expec-
tations for the screened Kondo model including antifer-
romagnetic interactions⁷³ as well as for the Kondo reso-
nance model⁷⁴. Here, we emphasise though the analogy
with similar data in YbPtBi²⁰ in the spirit of a Weyl-
Kondo scenario¹³. In fact, for a Weyl-Kondo semimetal
the linear dispersion relation at the Weyl nodes dic-
tates the low T dependence of $C(T) \sim k_B \left(\frac{k_B T}{\hbar v_F} \right)^3¹³. In
CeAlGe, v_F from $C(T)$ at all fields^{26,72} are significantly
reduced (i.e., $v_F \sim 6 - 9 \times 10^3$ m/s), compared to values
for LaAlGe, in a quite similar manner as in YbPtBi with
respect to its reference compound LuPtBi²⁰. Further-
more, v_F in CeAlGe from $C(T)$ at zero field is roughly a
factor of three smaller than v_F at low T inferred from
 $\Delta\sigma_1(\omega)$ (Fig. 3), thus establishing a fair agreement
among different experiments.$

We now turn our attention to the estimation of m^*/m_e ,
which may be achieved from the charge dynamics in con-
junction with the additional constraint given by the Som-
merfeld γ value of the electronic component in $C(T)$.
From its definition together with the Drude plasma fre-
quency ω_p it follows that $\frac{m^*}{m_e} = \left(\frac{\gamma}{V_m} \right)^{3/4} \frac{1}{\sqrt{\omega_p}}$ ^{12,32,75}, V_m
being the molar volume⁷⁶. We consider the total Drude
 SW from our phenomenological fit (inset of Fig. 3), in
order to define the effective, total plasma frequency ω_p ³².
With $\gamma \simeq 1.61$ and 50 mJ/(mol K²)^{71,72} we achieve $\frac{m^*}{m_e} \simeq$
2.7 and 49 at 5 K for LaAlGe and CeAlGe, respectively.
Even though the values of v_F from $\Delta\sigma_1(\omega)$ (Fig. 3) in-
dicate a renormalisation factor of about two, the result-
ing enhancement factor from m^*/m_e is approximately
twenty at low T between CeAlGe and LaAlGe, the latter
supplying the bare band values. It is worth underscor-
ing that the enhancement of the optical m^*/m_e pairs
with the narrowing of the metallic component in $\sigma_1(\omega)$
of CeAlGe (i.e., reduction of the Drude scattering rates,
Fig. S6 in SM³²). This bears testimony to a substan-
tial band renormalisation driven by Kondo-like electronic
correlations^{6,12}.

In a broader perspective, we may then claim that the
 f -electrons play here different roles, since they are un-
doubtedly involved in shaping the complex magnetism
in CeAlGe^{21,24-28} and equally govern the transport and
thermodynamic properties, thus the physics at low en-
ergy scales close to E_F . Our bulk sensitive optical results
complement and in this specific case even outreach the
(surface sensitive) ARPES experiments, which did fail so
far resolving the heavy Weyl fermion states.

In conclusion, upon comparing the charge dynam-
ics of the f -electron CeAlGe with that of its refer-
ence, empty f -states LaAlGe material we provide evi-

dence that correlation-driven Kondo physics (i.e., band-hybridisation with f -electrons) affects the dispersion in momentum space near the type II Weyl points in CeAlGe upon decreasing T below 100 K. The resulting band renormalisation greatly reduces v_F and enhances the charge carriers m^*/m_e in CeAlGe with respect to LaAlGe, both revealing a heavy Weyl-Kondo state¹³. How precisely the present results and drawn conclusions might be influenced by the controversial magnetic transition^{24–28} in CeAlGe at low T remains to be determined by future studies. Furthermore, Berry curvature fields associated with the Weyl nodes at E_F generate an intrinsic (topological) anomalous Hall effect, which should survive and get enhanced at low T into the correlated state, as evinced in YbPtBi²⁰ and Ce₃Bi₄Pd₃¹⁹. In this context, the ferromagnetic PrAlGe Weyl semimetal²¹, characterised by a large anomalous Hall effect^{77,78}, sparks our interest in chasing the related spectroscopic signatures⁶⁴; the ongoing investigations of its charge dynamics will be of relevance as a proof of concept.

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by publisher] for additional information on the samples
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