

# Electronic structure of the Gd-based intermetallics GdCu<sub>2</sub>Ge<sub>2</sub> and GdCuAl<sub>3</sub>

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We present a temperature-dependent reflectivity study of single crystals of the ternary intermetallic compounds GdCu<sub>2</sub>Ge<sub>2</sub> and GdCuAl<sub>3</sub> over a broad spectral range (100–18 000 cm<sup>−1</sup>, equivalent to 12 meV–2.23 eV) down to 13 K. Below 2000 cm<sup>−1</sup>, the optical spectra are dominated by the response of itinerant charge carriers exhibiting two distinct scattering rates. While the response of the highly damped charge carriers shows negligible temperature dependence, the weakly damped carriers closely follow the dc resistivity and are significantly suppressed in GdCuAl<sub>3</sub>, consistent with its higher resistivity. Supported by density-functional theory calculations, we further show that elemental substitution from GdCu<sub>2</sub>Ge<sub>2</sub> to GdCuAl<sub>3</sub> tunes the X-site *p*-orbital character (X = Ge, Al) of the low-energy electronic structure, while preserving key features such as linearly dispersing bands and saddle points despite the accompanying structural modifications. The reduced experimental plasma frequency of both compounds signals enhanced electronic correlations, which induce moderate band-energy renormalization.

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## I. INTRODUCTION

*f*-electron-based intermetallic compounds form a remarkably rich platform in condensed matter physics, where subtle changes in chemical composition can significantly alter both the magnetic and electronic properties of the system [1–10]. Within this broad material landscape, ternary compounds derived from the layered tetragonal BaAl<sub>4</sub> structure provide a vital playground for studying the interplay between localized 4*f* magnetic moments and itinerant *s*, *p*, and *d* electrons. Based on the stacking sequence, these materials crystallize in three different space groups including the centrosymmetric *I4/mmm* (ThCr<sub>2</sub>Si<sub>2</sub> structure) and *P4/nmm* (CaBe<sub>2</sub>Ge<sub>2</sub> structure) groups and the noncentrosymmetric *I4mm* space group (BaNiSn<sub>3</sub> structure), with broken inversion symmetry [2,11]. These intermetallic ternary compounds display a wide variety of intriguing phenomena, such as unconventional superconductivity [12–17], heavy fermion physics [18–22], non-Fermi liquid behavior [1,23,24], density waves [25,26], as well as topologically nontrivial electronic states [12,27,28].

Because of their half-filled 4*f* shell, Gd-based ternary intermetallics feature strongly localized 4*f* electrons and thus provide ideal systems to study these physical phenomena in the absence of additional complications such as Kondo effects, Jahn–Teller distortion, or crystal-electric-field effects

[29–32]. Dominated by RKKY interactions, these compounds typically exhibit well-defined magnetic ordering temperatures, displaying a large variety of magnetic ground states ranging from conventional ferromagnetism to helical and modulated antiferromagnetic orders [4,29,33–36]. Recently, magnetic-field-induced emergent skyrmion lattices have been reported in the noncentrosymmetric EuNiGe<sub>3</sub> [37,38] (triangular skyrmion lattice), as well as in centrosymmetric GdRu<sub>2</sub>Si<sub>2</sub> [39,40] and Gd<sub>2</sub>PdSi<sub>3</sub> [41,42] (square skyrmion lattice), both in the absence of geometrical frustration and with negligible or even vanishing Dzyaloshinskii–Moriya interaction in the latter cases.

Here, we address the modifications in the electronic structure of the ternary intermetallic compounds GdCu<sub>2</sub>Ge<sub>2</sub> and GdCuAl<sub>3</sub> arising from elemental substitution and, in the latter case, the loss of centrosymmetry [see crystal structures in Figs. 1(a) and 1(b)]. Using Fourier-transform infrared spectroscopy as a bulk-sensitive probe together with density-functional theory (DFT) calculations, we unveil an intraband response with two distinct scattering rates and demonstrate robust band topology with tunable orbital character in ternary compounds derived from the layered tetragonal BaAl<sub>4</sub> structure.

## II. METHODS

### A. Synthesis and characterization

High-purity ingots of Gd, Cu, and Al were cut and weighed according to a 1:1:3 stoichiometry in a glove box. The elements were then transferred into a fused silica ampule, which was sealed under vacuum. The ampule was placed in a vertical tube furnace and heated from room temperature at 100 °C/h to 1190 °C, where the temperature was held for 2 h. The furnace

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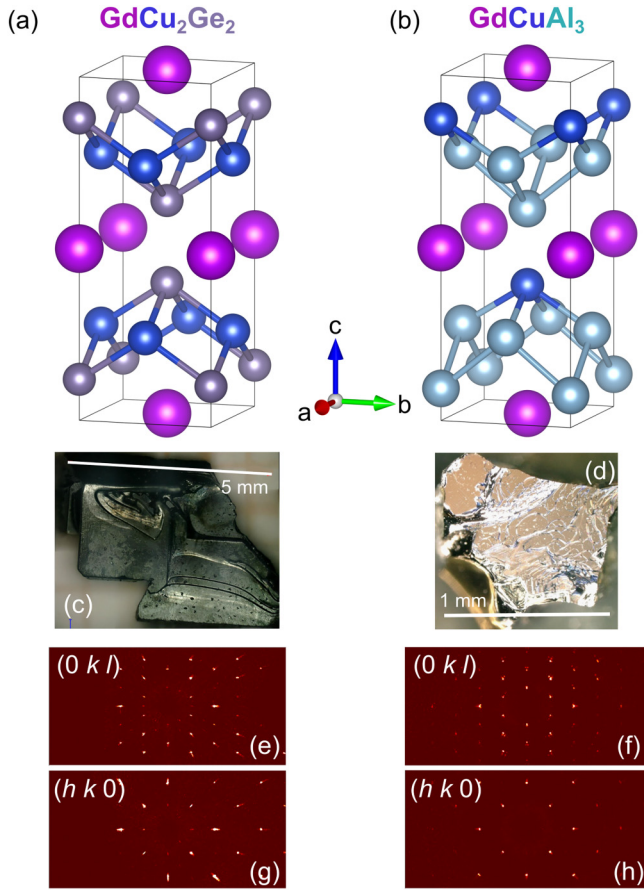


FIG. 1. (a), (b) Crystal structures of  $\text{GdCu}_2\text{Ge}_2$  and  $\text{GdCuAl}_3$ , respectively, visualized with VESTA [43]. (c), (d) Photographs of the single crystals used in the optical experiments prior to polishing. Backscattered x-ray Laue images of the same crystals are given in panels (e)–(h), confirming the tetragonal symmetry.

was then slowly cooled at 5 °C/h to 900 °C. The elongated boule was cracked and shiny crystals of typically 1 mm were extracted in air.

For  $\text{GdCu}_2\text{Ge}_2$ , a  $\text{GdCuGe}_3$  Sn-flux growth was attempted by mixing 0.22361 g Gd, 0.09036 g Cu, 0.30988 g Ge, and 3.37614 g Sn in a glove box and loading the mixture into a Canfield crucible set of two crucibles separated by a sieve. These were sealed in a fused silica ampule under vacuum.

Afterward, the ampule was heated to 1000 °C at 100 °C/h, and the temperature was held for 24 h. The furnace was slowly cooled at 1 °C/h to 550 °C, at which point the Sn flux was centrifuged through sieves. The ampules were opened in air and crystals of  $\text{Gd}_2\text{CuGe}_6$  and  $\text{GdCu}_2\text{Ge}_2$  with sizes of several millimeters were obtained.

The single crystals were characterized by single-crystal XRD. We found Gd-Cu-Ge to crystallize in the  $\text{ThCr}_2\text{Si}_2$ -type structure (space group  $I4/mmm$ ), while  $\text{GdCuAl}_3$  crystallizes in the  $\text{GdNiSn}_3$ -type structure (space group  $I4mm$ ). The corresponding Fourier maps are shown in Figs. 1(e)–1(h) and the single-crystal refinement results are summarized in Table I (see the Supplemental Material [44] for CIF files). The crystals were oriented during this process and polished for optical characterization.

Four-probe resistivity measurements were performed on the same crystals used in the optical experiments [see Figs. 1(c) and 1(d)]. The low dc resistivity values shown in Figs. 2(a) and 2(b) indicate the highly metallic nature of these samples. The kinks observed around  $T_N = 10$  K and 12 K in the resistivity curves of  $\text{GdCu}_2\text{Ge}_2$  and  $\text{GdCuAl}_3$ , respectively, correspond to the antiferromagnetic transitions, consistent with previous Mössbauer spectroscopy studies [45,46].

## B. Optical measurements

Temperature-dependent reflectivity measurements were performed on optically polished samples in the isotropic  $ab$  plane down to 13 K ( $T > T_N$ ), covering a broad frequency range 100–18 000  $\text{cm}^{-1}$  (12 meV–2.23 eV). For the high-energy range ( $\omega > 600 \text{ cm}^{-1}$ ), a Bruker Vertex 80v spectrometer coupled with a Hyperion IR microscope was used, while the low-energy range ( $\omega < 600 \text{ cm}^{-1}$ ) was measured with a Bruker IFS113v spectrometer and a custom-built cryostat. Freshly evaporated gold mirrors served as reference in these measurements. The absolute value of the reflectivity was obtained by an in situ gold-overcoating technique in the far-infrared range, as described in Ref. [47].

Below  $100 \text{ cm}^{-1}$ , we use standard Hagen–Rubens extrapolations, considering the metallic nature of our samples, while for the high-energy range we utilize x-ray scattering functions to extrapolate the data [48]. The optical conductivity is then calculated from the measured reflectivity via Kramers–Kronig analysis [49].

TABLE I. Structural parameters extracted from single-crystal refinement.  $\text{GdCuAl}_3$  crystallizes in  $I4mm$  with lattice constants  $a = b = 4.1412(3) \text{ Å}$  and  $c = 10.6232(16) \text{ Å}$ , with  $R_{\text{all}} = 0.0490$  and  $wR_{\text{gt}} = 0.1208$ .  $\text{GdCu}_2\text{Ge}_2$ , on the other hand, crystallizes in  $I4/mmm$  with lattice constants  $a = b = 4.0591(11) \text{ Å}$  and  $c = 10.233(3) \text{ Å}$ , with  $R_{\text{all}} = 0.0487$  and  $wR_{\text{gt}} = 0.0941$ . Note that  $U_{23} = U_{13} = U_{12} = 0$ .

| Atom | $x$ | $y$ | $z$         | $U_{\text{eq}}$ | $U_{11}$   | $U_{22}$   | $U_{33}$   |
|------|-----|-----|-------------|-----------------|------------|------------|------------|
| Gd1  | 1   | 1   | 0.57795(5)  | 0.0125(6)       | 0.0103(6)  | 0.0103(6)  | 0.0169(9)  |
| Cu2  | 1/2 | 1/2 | 0.4503(3)   | 0.0138(13)      | 0.0126(17) | 0.0126(17) | 0.016(3)   |
| Al3  | 1/2 | 0   | 0.3281(8)   | 0.0131(11)      | 0.015(7)   | 0.012(7)   | 0.013(2)   |
| Al4  | 1/2 | 1/2 | 0.6704(11)  | 0.015(3)        | 0.015(4)   | 0.015(4)   | 0.016(6)   |
| Gd1  | 1   | 1   | 1/2         | 0.0090(4)       | 0.0112(5)  | 0.0112(5)  | 0.0046(7)  |
| Ge2  | 1/2 | 1/2 | 0.61955(16) | 0.0101(4)       | 0.0138(5)  | 0.0138(5)  | 0.0027(8)  |
| Cu3  | 0   | 1/2 | 3/4         | 0.0120(5)       | 0.0155(6)  | 0.0155(6)  | 0.0050(10) |

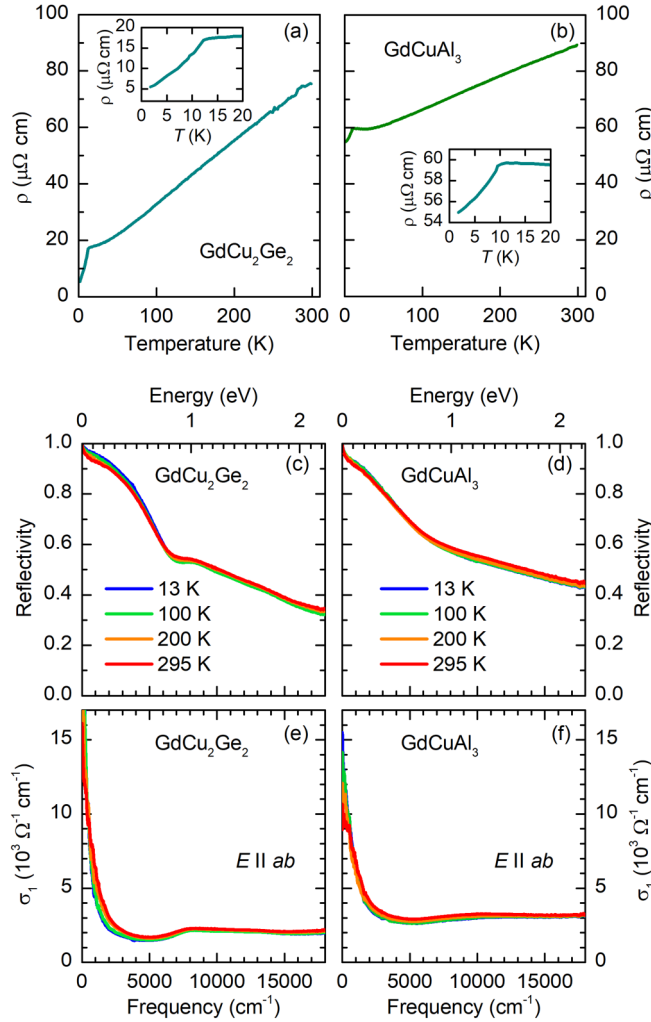


FIG. 2. DC resistivity curves of  $\text{GdCu}_2\text{Ge}_2$  (a) and  $\text{GdCuAl}_3$  (b) measured in the  $ab$  plane and normalized to the resistivity values obtained from our optical measurements via Hagen–Rubens fits. The kinks at low temperatures, highlighted in the insets, correspond to the antiferromagnetic transitions at  $T_N = 10$  K and 12 K of  $\text{GdCu}_2\text{Ge}_2$  and  $\text{GdCuAl}_3$ , respectively. Panels (c) and (d) show the in-plane reflectivity of  $\text{GdCu}_2\text{Ge}_2$  and  $\text{GdCuAl}_3$  at selected temperatures, while (e) and (f) present the corresponding calculated real part of the optical conductivity.

### C. Computational

Density-functional theory (DFT) calculations of the band structure and optical conductivity were performed using the Wien2K [50,51] code with the Perdew–Burke–Ernzerhof flavor of the exchange–correlation potential [52]. Experimentally determined structural parameters given in Table I were used in the calculations. Fully relativistic self-consistent calculations were converged on a  $12 \times 12 \times 12$   $k$  mesh. For the nonmagnetic calculations presented in the main text, the open-core approximation was employed, in which the Gd  $4f$  electrons were treated as core states. Alternatively, a Hubbard  $U = 10$  eV was added to the  $4f$  shell of Gd within the DFT +  $U$  framework, using the FLL (fully localized limit) double-counting correction to push the minority  $4f$  states well above the Fermi level. In this approach, an in-plane

antiferromagnetic order of the Gd atoms was imposed, necessary to stabilize the  $\text{Gd}^{3+}$  ground state (see the Supplemental Material [44]). The optical conductivity was calculated using the OPTIC module [53] on a denser  $k$  mesh with up to  $100 \times 100 \times 100$  points.

### III. RESULTS AND DISCUSSION

Figures 2(c) and 2(d) display the temperature-dependent reflectivity of  $\text{GdCu}_2\text{Ge}_2$  and  $\text{GdCuAl}_3$ , respectively. At low frequencies, the high reflectivity values together with the Drude-like increase in the optical conductivities, presented in Figs. 2(e) and 2(f), demonstrate the highly metallic nature of the samples. Conductivity values in the  $\omega \rightarrow 0$  limit are obtained from the Hagen–Rubens fit of the reflectivity and match well with our four-probe dc resistivity measurements performed on the same samples, as shown in Figs. 2(a) and 2(b).

To gain further insight into the different electronic contributions to the spectra, we perform a Drude–Lorentz fit by simultaneously modeling the measured reflectivity and the complex optical response obtained from the Kramers–Kronig analysis,

$$\tilde{\epsilon}(\omega) = \epsilon_\infty - \frac{\omega_{p,\text{Drude}}^2}{\omega^2 + i\omega/\tau_{\text{Drude}}} + \sum_j \frac{\Omega_j^2}{\omega_{0,j}^2 - \omega^2 - i\omega\gamma_j}. \quad (1)$$

Here,  $\epsilon_\infty$  accounts for the high-energy contributions to the real part of the dielectric permittivity [ $\tilde{\epsilon} = \epsilon_1 + i\epsilon_2$ ], and  $\omega_{p,\text{Drude}}$  and  $1/\tau_{\text{Drude}}$  are the plasma frequency and the scattering rate of the itinerant carriers, respectively.  $\omega_{0,j}$ ,  $\Omega_j$ , and  $\gamma_j$  describe the resonance frequency, width, and strength of the  $j$ th excitation, respectively. The complex optical conductivity [ $\tilde{\sigma} = \sigma_1 + i\sigma_2$ ] is then calculated as

$$\tilde{\sigma}(\omega) = -i\omega[\tilde{\epsilon} - \epsilon_\infty]/4\pi. \quad (2)$$

The decomposed experimental optical conductivities of  $\text{GdCu}_2\text{Ge}_2$  and  $\text{GdCuAl}_3$  at 13 K are shown in Figs. 3(a) and 3(b), respectively (see the decomposed optical spectra at other selected temperatures in the Supplemental Material [44]). At high frequencies ( $\omega > 2000 \text{ cm}^{-1}$ ), temperature-independent interband transitions are modeled by a total of five Lorentzians, shown in orange. At lower frequencies ( $\omega < 2000 \text{ cm}^{-1}$ ), the spectra exhibit a broad intraband contribution that, in the case of  $\text{GdCu}_2\text{Ge}_2$ , cannot be adequately described by a single Drude term. Although less apparent within the measured spectral range of  $\text{GdCuAl}_3$ , a second Drude component (shown in blue in Fig. 3) with significantly reduced spectral weight is required here as well to model the slight upturn below  $\sim 200 \text{ cm}^{-1}$ , as well as to reproduce the dc conductivity obtained from the four-contact transport measurements.

The occurrence of two Drude contributions with markedly different scattering rates is commonly observed in multiband systems, including topological materials such as Dirac and Weyl semimetals, as well as iron pnictides with the  $\text{ThCr}_2\text{Si}_2$  structure [54–60]. While the exact interpretation varies among different material classes, many of these systems host linearly dispersing bands that give rise to highly mobile carriers, whereas carriers associated with nonlinear bands tend to

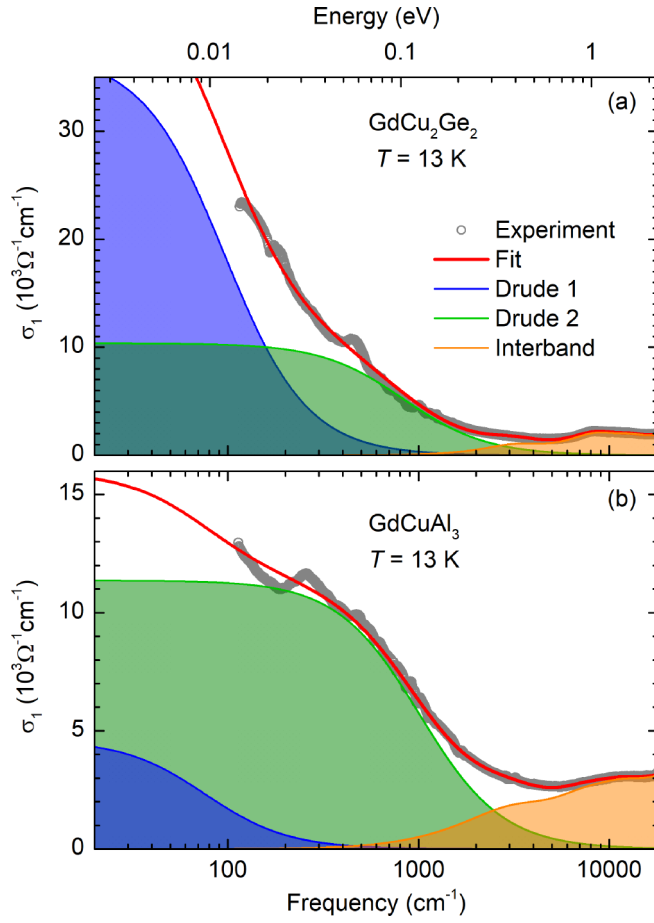


FIG. 3. Optical conductivity of  $\text{GdCu}_2\text{Ge}_2$  (a) and  $\text{GdCuAl}_3$  (b) at 13 K, modeled using two Drude contributions (blue and green) and five Lorentzians above  $2000 \text{ cm}^{-1}$ , describing the interband transitions (orange).

exhibit lower mobility. This picture is consistent with the electronic structure of the intermetallic compounds investigated here, as discussed below. At the same time, it should be noted that the two Drude components may also reflect two inherently different scattering channels rather than carriers originating from specific electronic bands [59,61–63].

As summarized in Fig. 4, the dc conductivity associated with the broad Drude term (Drude 2) remains temperature independent and exhibits similar scattering rates in both compounds within the error bars of our fits. In contrast, the dc conductivity of the narrow Drude term (Drude 1) closely follows the temperature dependence of the measured resistivity and is significantly enhanced in  $\text{GdCu}_2\text{Ge}_2$ , consistent with its lower resistivity.

The strength of the electronic correlations can be gauged by comparing the experimental and DFT-based plasma frequencies [64–66]. Here, the ratio  $\omega_{p,\text{exp}}^2/\omega_{p,\text{DFT}}^2$  is close to 1 for uncorrelated systems, while it approaches 0 for Mott insulators, indicating the full suppression of itinerant charge carriers. Given the presence of two Drude contributions in our spectra, the total experimental plasma frequency is calculated by  $\omega_{p,\text{exp}} = \sqrt{\omega_{p,\text{Drude1}}^2 + \omega_{p,\text{Drude2}}^2}$ . Using the experimental and DFT-derived plasma frequencies listed in Table II, we

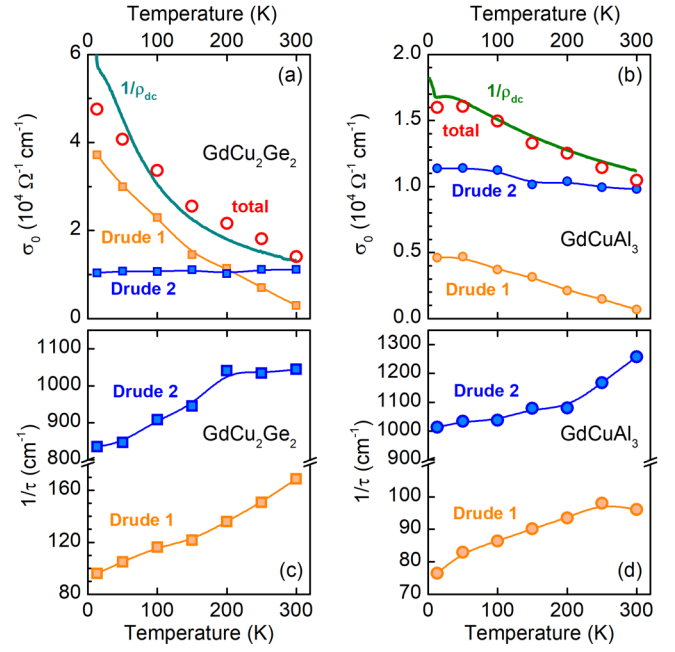


FIG. 4. (a), (b) Temperature-dependent dc conductivities of the Drude fits overlaid with the values obtained from the measured dc resistivity for  $\text{GdCu}_2\text{Ge}_2$  and  $\text{GdCuAl}_3$ , respectively. The associated scattering rates are plotted in (c) for  $\text{GdCu}_2\text{Ge}_2$  and (d) for  $\text{GdCuAl}_3$ .

calculate a ratio of 0.47 for both compounds at 13 K, indicating the presence of enhanced correlations. The determined ratios are highly robust with increasing temperature, as demonstrated by the comparable values obtained at room temperature and reported in Table II.

Further insight into the role of structural changes and differences in bonding on the low-energy electronic structure is obtained through a comparison between experimental and DFT-derived interband optical transitions summarized in Fig. 5. Panels (a) and (b) display the experimental interband optical conductivity of  $\text{GdCu}_2\text{Ge}_2$  and  $\text{GdCuAl}_3$ , respectively, obtained by subtracting the modeled Drude contributions from the data. For  $\text{GdCu}_2\text{Ge}_2$ , the interband absorption above 1 eV is well reproduced by the DFT calculations shown in gray in the same panel. At lower energies, however, the experimental

TABLE II. Drude fit parameters at 13 K and 295 K together with the calculated plasma frequencies as well as the ratio  $\omega_{p,\text{exp}}^2/\omega_{p,\text{DFT}}^2$  used as a gauge of electronic correlations.

|                                                   | $\text{GdCu}_2\text{Ge}_2$ |       | $\text{GdCuAl}_3$ |       |
|---------------------------------------------------|----------------------------|-------|-------------------|-------|
|                                                   | 13 K                       | 295 K | 13 K              | 295 K |
| $\sigma_{0,1} (\Omega^{-1}\text{cm}^{-1})$        | 37162                      | 2925  | 4619              | 679   |
| $\sigma_{0,2} (\Omega^{-1}\text{cm}^{-1})$        | 10370                      | 11121 | 11365             | 9800  |
| $\tau_1^{-1} (\text{cm}^{-1})$                    | 96                         | 169   | 76                | 96    |
| $\tau_2^{-1} (\text{cm}^{-1})$                    | 836                        | 1044  | 1013              | 1257  |
| $\omega_{p,\text{Drude1}} (\text{cm}^{-1})$       | 14633                      | 5446  | 4603              | 1979  |
| $\omega_{p,\text{Drude2}} (\text{cm}^{-1})$       | 22808                      | 26394 | 26284             | 27191 |
| $\omega_{p,\text{exp}} (\text{cm}^{-1})$          | 27098                      | 26950 | 26684             | 27263 |
| $\omega_{p,\text{DFT}} (\text{cm}^{-1})$          | 39733                      |       | 39059             |       |
| $\omega_{p,\text{exp}}^2/\omega_{p,\text{DFT}}^2$ | 0.47                       | 0.46  | 0.47              | 0.49  |

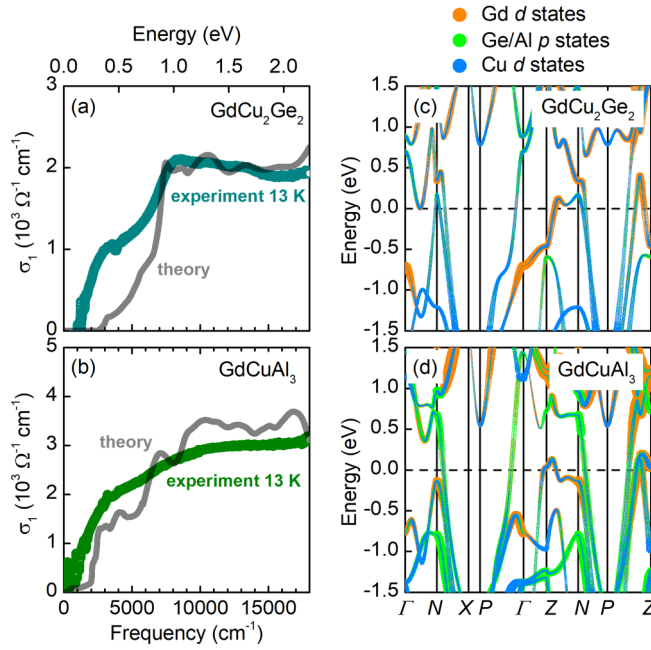


FIG. 5. (a), (b) Experimental interband optical transitions of  $\text{GdCu}_2\text{Ge}_2$  and  $\text{GdCuAl}_3$  at 13 K (colored scatter), respectively, obtained by subtracting the Drude contributions from the spectra, overlaid with the DFT-calculated optical conductivities (gray solid line). Note that a broadening of up to 0.1 eV was applied to the calculated spectra above  $5000 \text{ cm}^{-1}$  to facilitate direct comparison with the experimental data. Furthermore, the energy axis of the theoretical optical conductivity of  $\text{GdCuAl}_3$  is rescaled by a factor of 1.5. Band structures of  $\text{GdCu}_2\text{Ge}_2$  and  $\text{GdCuAl}_3$  are given in (c) and (d), respectively, with colored dots representing contributions from different atomic orbitals.

data reveal a step-like onset whose spectral weight is strongly suppressed in the DFT-derived optical conductivity. As revealed by our band-resolved calculations in Fig. S2 within the Supplemental Material [44], these low-energy interband excitations arise predominantly from transitions between linearly dispersing bands crossing the Fermi energy along  $N$ - $X$ ,  $N$ - $P$ , and  $P$ - $Z$  (see Fig. 5 for the band structures).

For  $\text{GdCuAl}_3$ , the best agreement between the experimental and DFT-derived optical spectra is obtained by rescaling the calculated energies by a factor of 1.5, as shown in Fig. 5(b). Such a rescaling is commonly observed in correlated materials and indicates a moderate renormalization of the band energies [67–69]. A similar renormalization of band energies can also be envisaged for  $\text{GdCu}_2\text{Ge}_2$ , where the possible effect would primarily concern bands “B” and “C” in close proximity to the Fermi level (see Fig. S2 within the Supplemental Material [44]) and would consequently shift the onset of interband transitions involving these two bands to lower energies.

In both compounds, the band structure is dominated by strongly hybridized  $\text{Gd } 5d$ ,  $\text{Cu } 3d$ , and  $\text{Al/Ge } p$  states, as demonstrated in Figs. 5(c) and 5(d). The overall electronic structure exhibits a multiband nature, comprising linearly dispersing bands and saddle points, and remains largely unchanged upon chemical substitution, highlighting its robustness against structural modifications. This renders these

Gd-based intermetallics promising platforms for tuning electronic properties through controlled chemical substitution or carrier doping while preserving their characteristic low-energy band features. Notably, the electronic structure of  $\text{GdCuAl}_3$  exhibits substantially enhanced contributions from  $\text{Al } 3p$  states, resulting in an upward shift of the  $\text{Al } 3p$ -derived bands. As shown in Fig. S2 within the Supplemental Material [44], these shifts do not crucially alter the shape of the interband transitions across the measured energy range. Such tunability provides an avenue for exploring filling-dependent electronic and magnetic phases, including high-temperature superconductivity and topological spin textures such as skyrmions [3,70–73].

Furthermore, this upward shift of the  $3p$  states leads to the formation of band saddle points along  $\Gamma$ - $Z$ - $N$  that lie in close proximity to the Fermi level. Together with the overall enhanced  $\text{Al } 3p$  contribution to the Fermi surface, this may account for changes in the scattering mechanism, resulting in the dampened Drude response and the overall higher resistivity observed in  $\text{GdCuAl}_3$ . Finally, the loss of centrosymmetry in  $\text{GdCuAl}_3$  gives rise to small spin-orbit-coupling-induced band splittings on the order of  $\sim 20 \text{ meV}$  along certain  $k$  directions (see Fig. S3 within the Supplemental Material [44]). These splittings do not produce additional interband transitions with sufficient spectral weight to be resolved in the optical spectra, consistent with the DFT-derived optical conductivity.

#### IV. CONCLUSIONS

We studied the broadband in-plane optical conductivity of single crystals of two Gd-based ternary intermetallic compounds: centrosymmetric  $\text{GdCu}_2\text{Ge}_2$  and noncentrosymmetric  $\text{GdCuAl}_3$ . The response of free carriers reveals two distinct scattering rates, which can be interpreted in terms of highly damped and weakly damped charge carriers, with the latter being the dominant contribution to the temperature dependence of the dc transport. The electronic structures of both compounds exhibit highly similar band dispersions and enhanced electronic correlations, evidenced by reduced experimental plasma frequencies and band-energy renormalization. In  $\text{GdCuAl}_3$ , the augmented  $\text{Al } 3p$  contribution modifies the orbital character near the Fermi surface, leading to band saddle points at the Fermi level and an overall upward shift of bands with strong  $\text{Al } 3p$  contribution. The loss of centrosymmetry in  $\text{GdCuAl}_3$ , on the other hand, has only a minor effect on the overall electronic structure, producing spin-orbit-coupling-induced splittings of several tens of meV without introducing additional interband optical transitions. These results highlight the robustness of the electronic structure in ternary intermetallics against chemical substitution, making them promising platforms for future band-filling tuning and exploration of emergent electronic and magnetic phases. The coexistence of itinerant carriers with two distinct scattering rates motivates Hall-effect measurements to further disentangle their contributions to the transport properties.

#### DATA AVAILABILITY

The data that support the findings of this article are not publicly available upon publication because it is not

technically feasible and/or the cost of preparing, depositing, and hosting the data would be prohibitive within the terms of

this research project. The data are available from the authors upon reasonable request.

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