

Insulating Out-of-Plane Transport in Anisotropic Metals: The Case of Sr_2RuO_4 and Related Layered Oxides

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We reveal a mechanism that may explain the nonmetallic out-of-plane resistivity in layered metals. By carefully examining how the Drude-Boltzmann expression for the c -axis conductivity emerges out of the Kubo formula, we find, besides the standard metallic term proportional to the carrier lifetime τ , a non-Drude contribution proportional to $1/\tau$. The Drude behavior breaks down when $1/\tau > 2\eta^*$, the crossover value η^* being small (and hence observable) when the c -axis velocities vary rapidly with the distance from the Fermi surface. We consider the Hund metal Sr_2RuO_4 as a test case, which we study within a realistic dynamical mean-field theory approach. The non-Drude behavior observed experimentally in c -axis transport is reproduced and explained by our considerations, showing that earlier invoked extrinsic mechanisms that involve either impurities or phonons are unnecessary. Although the small value of η^* arises from a peculiar accidental cancellation due to destructive interference, characteristic of body-centered tetragonal lattices, we emphasize and discuss the generality of this mechanism.

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The electronic response of metals is often described by the simple Drude formula, $\sigma = ne^2\tau/m$, where n is the carrier density, m the carrier mass, and τ the momentum relaxation time. This expression embodies the intuitive idea that less frequent scattering (i.e., longer τ) enhances electrical conductivity. Although derived from a classical equation of motion, this framework is used to describe a broad range of metallic systems, including unconventional ones [1–3]. However, this description clearly does not apply in strongly anisotropic systems whenever in- and out-of-plane conductivities behave qualitatively differently. Notable examples include cobaltates [4] and layered ruthenates [5], where the in-plane resistivity remains

metallic while the out-of-plane resistivity exhibits insulatinglike behavior ($d\rho/dT < 0$) above some characteristic temperature (see Fig. 1).

These observations have been loosely associated with the “coherence-incoherence crossover,” i.e., the disappearance of quasiparticles that was reported in photoemission experiments [7,8] and argued to indicate a breakdown of

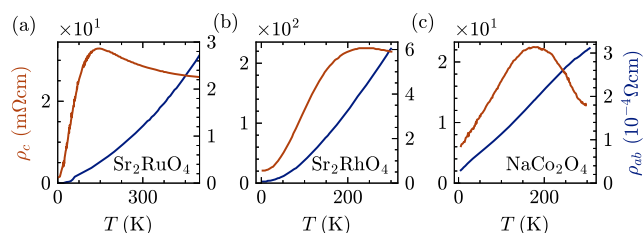


FIG. 1. Measurements of the out-of-plane (left axis, dark red) and in-plane (right axis, blue) resistivity in layered Sr_2RuO_4 (a), Sr_2RhO_4 (b), and NaCo_2O_4 (c). The data are digitized from Refs. [5–7]. The out-of-plane resistivity exhibits a maximum near the crossover temperature T_M , indicating the breakdown of Drude transport.

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Boltzmann transport. At first sight this is an appealing picture: when there are no quasiparticles, metallic transport cannot occur. However, as noted by Millis [9], it raises another question, namely why then is the in-plane transport metallic?

Subsequent work has exposed additional limitations of the coherence-incoherence crossover picture. Firstly, recent experiments on Sr_2RuO_4 [10] have shown that the quasiparticle weight actually does not vanish with increasing temperature and that quasiparticles can be resolved above the crossover temperature T_M associated with the resistivity maximum (see also Refs. [11,12]). Secondly, as first emphasized by Prange and Kadanoff [13], the existence of long-lived quasiparticles is not a strict prerequisite for the applicability of Boltzmann theory. Boltzmann theory may give accurate results at temperatures far above the Fermi liquid temperatures [12,14] or even in the strange metal regime of cuprates [15,16]. Thus, the puzzling question of the c -axis resistivity maximum remains open.

This question is especially pertinent in the case of Sr_2RuO_4 , a well-characterized, strongly correlated metal with a body-centered tetragonal structure for which the experiments are particularly controlled due to the existence of large ultraclean single crystals. Its electronic properties are determined by four electrons occupying the low-energy t_{2g} orbitals, with the d_{xz}/d_{yz} orbitals defining the out-of-plane conduction. Transport in this material is highly anisotropic, with ρ_c/ρ_{ab} exceeding 1000 at low temperatures [5]. Its Fermi surface and low-energy electronic structure have been accurately captured by density functional theory + dynamical mean-field theory (DFT + DMFT) approaches [17,18], yet an understanding of its transport properties [19], in particular the origin of the c -axis resistivity maximum, remains incomplete.

It should be noted that a substantial body of theoretical work has successfully explained and modeled the c -axis maximum through nonelectronic, extrinsic mechanisms, invoking either polaronic effects [20,21] or conduction via (resonant) interlayer impurities [22,23]. These approaches most often rely on simplified noninteracting models (an important oversimplification, as will become clear in the following) and additional assumptions, while generally not attempting to describe other observables at the same time.

Although the proposed theories are plausible and should be pursued further by carefully assessing their validity through complementary experimental studies, we argue that there is a simpler argument that can equally well explain the anisotropic behavior. It originates entirely in a peculiarity of the band structure—destructive interference of hopping amplitudes suppresses contributions to out-of-plane conduction in a way that strongly depends on in-plane momentum—that leads to a nonisotropic breakdown of the Drude picture. Namely, as we show in the following, if one starts from band theory and calculates the c -axis conductivity from the Kubo formula within a simplification

that is valid for strongly anisotropic metals, one finds that the conductivity quite generically behaves as

$$\sigma^c(\tau) = a_0\tau + a_1/\tau + \mathcal{O}(1/\tau^3). \quad (1)$$

The second term dominates when the lifetime becomes sufficiently short: counterintuitively the conductivity starts to grow with $1/\tau$. We first derive this behavior with qualitative considerations, then illustrate it for a DFT-derived band structure but taking the scattering rate $1/\tau$ as a parameter, and finally report temperature-dependent results with the scattering given by self-energies calculated using DMFT.

Our analysis is based on the following expression for the c -axis conductivity:

$$\sigma^c \propto \int d\omega \left(-\frac{\partial f}{\partial \omega} \right) \int d^2k_{\parallel} dk_{\perp} v_{\mathbf{k}}^z A_{\mathbf{k}}(\omega) v_{\mathbf{k}}^z A_{\mathbf{k}}(\omega). \quad (2)$$

Here, $v_{\mathbf{k}}^z$ is the electron velocity along the z direction and $A_{\mathbf{k}}(\omega) = (-1/\pi) \text{Im}[\omega + \mu - \epsilon_{\mathbf{k}} - \Sigma(\omega)]^{-1}$ is the single particle spectral function at energy ω with $\epsilon_{\mathbf{k}}$ the single-particle dispersion, Σ the self-energy, and μ the chemical potential. When applied to the multiorbital case as in realistic calculations reported later, these quantities become matrices in which case the trace over internal spin, orbital, and band indices is implicit.

Expression (2) can be derived from the Kubo formula. It can be viewed as an exact expression to first nontrivial order in the interlayer hopping, which is justified when the latter is smaller than the scattering rate. In that case, $\epsilon_{\mathbf{k}}$ only involves the intralayer dispersion, and it can be shown that vertex corrections vanish in this limit, which is the relevant one for most of the analysis below. Away from this limit, the Kubo formula involves vertex corrections which are neglected in Eq. (2) but are expected to be small in c -axis transport [24–26]; see the Supplemental Material [27] for a more detailed discussion.

We write the momentum integration in terms of the components parallel $k_{\parallel}$ and perpendicular $k_{\perp}$ (labeled as k in the following) with respect to the Fermi surface, and set $\hbar = 1$. The conductivity is a sum of contributions along the Fermi surface $k_{\parallel}$, i.e., $\sigma^c = \int d^2k_{\parallel} \sigma_{k_{\parallel}}^c$. Let us investigate the behavior of the integrand $\sigma_{k_{\parallel}}^c$. Changing temperature in Eq. (2) leads to a change of the energy integration window through Fermi functions, and to a change of scattering through the self energy $\Sigma(\omega)$. It is instructive to consider the two effects separately. Let us hence assume a frequency-independent scattering rate $\Sigma = -i\eta = -i/(2\tau)$ (originating, for instance, from impurity scattering), i.e., spectral functions to be given by a simple Lorentzian:

$$A_k(\omega) \approx -\frac{1}{\pi} \text{Im} \frac{1}{\omega - v_F k + i\eta}, \quad (3)$$

where the band dispersion perpendicular to the Fermi surface has been linearized, $\epsilon_k = v_F k$. At the same time,

we retain the momentum dependence in the velocities, and expand in momentum perpendicular to the Fermi surface as

$$v_{k,k_{\parallel}}^z \approx v_0 + v_1 k + \frac{v_2}{2} k^2. \quad (4)$$

We now consider the low temperature limit ($k_B T \ll \eta$), where $-\partial f / \partial \omega \rightarrow \delta(\omega)$ in Eq. (2). Evaluating the remaining integral over momentum [see the Supplemental Material [27]] we get

$$\sigma_{k_{\parallel}}^c = \frac{\alpha v_0^2}{2\pi v_F} \left(\frac{1}{\eta} + \frac{v_1^2 + v_0 v_2}{v_0^2 v_F^2} \eta \right) \quad (5)$$

with a unit-dependent constant α . There are, thus, two contributions: a standard metallic Drude term with conductivity proportional to the scattering lifetime $1/\eta$, and a higher-order term directly proportional to the scattering rate η . The standard Drude behavior dominates for small η . The corrections come from a change of the velocity away from the Fermi level that leads to an increase of conductivity (if $v_1^2 + v_0 v_2 > 0$, a situation relevant to materials discussed in this Letter). Hence, when η becomes larger than a certain limiting value

$$\eta^* = \sqrt{\frac{v_F^2 v_0^2}{v_1^2 + v_0 v_2}}, \quad (6)$$

the contribution to conductivity at that value of $k_{\parallel}$ goes from a metallic dependence ($\propto \tau$) to an insulating one ($\propto 1/\tau$).

Several comments are in order: (i) notice that in the derivation we used the peculiarities of the strongly anisotropic band structure. Retaining the momentum dependence of v^z but truncating the band energy at the linear term $v_F k$ is consistent if the c -axis dispersion is negligible compared to the in-plane one. Nonlinearities in the in-plane dispersion on the other hand affect both velocities and the spectral functions; the two effects work in opposite directions and largely compensate. This is crucial to

understand why the proposed effects apply for the case of the out-of-plane conductivity but *not* the in-plane one. (ii) Dependence on $k_{\parallel}$ can be restored simply by performing a Fermi surface average of contributions in Eq. (5). This will affect the values of prefactors in front of the $1/\eta$ and η terms, but the form of the function remains the same, leading to Eq. (1). (iii) Fine details of the c -axis dispersion determine η^* , and the emergence of non-Drude behavior depends sensitively on those details as will be shown next.

We now turn to Sr_2RuO_4 , for which the above discussion is directly relevant: the evaluated η^* is indeed found to be small enough for the non-Drude contribution to have observable effects in the experimentally accessible temperature range. We further demonstrate that, in order to properly describe these effects, it is important to retain hopping amplitudes to neighbors beyond the nearest ones in the c direction. The Fermi surface of Sr_2RuO_4 consists of three sheets: the quasi-1D α and β sheets associated with the d_{xz}/d_{yz} orbitals, and the γ sheet associated with the in-plane d_{xy} orbital—the latter contributes to c -axis transport less due to smaller out-of-plane hoppings. To systematically document this we consider, besides a full *ab initio* description of the electronic structure, two simplified models that differ only in the range of c -axis hopping included. The hopping amplitudes are determined from DFT calculations followed by construction of Wannier functions and a careful tight-binding (TB) analysis as outlined in the Supplemental Material [27]. The number of in-plane hopping amplitudes does not affect our conclusions; for consistency, we use the six largest ones (see Table 1 in the Supplemental Material [27]). For now, we neglect spin-orbit coupling (SOC) and will return to its effects later. Along the c axis, only two intraorbital hopping amplitudes are relevant: the nearest neighbor $t_1^c = -16.2$ meV (orange) and the next-nearest neighbor $t_2^c = -5.9$ meV (blue), as shown in Fig. 2. Model *M1* includes the six in-plane hopping amplitudes and t_1^c , while model *M2* also includes t_2^c . The corresponding parameters are listed in Table 2 in the Supplemental Material [27]. We note

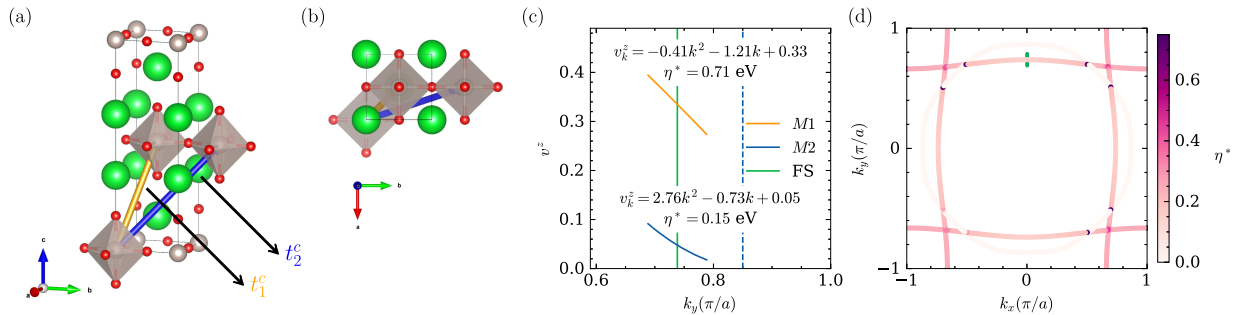


FIG. 2. (a) Side view of the body-centered tetragonal lattice of Sr_2RuO_4 indicating the dominant out-of-plane hopping amplitudes. (b) Top view. (c) The dependence of v^z along a cut perpendicular to the Fermi surface ($k_x = 0.0\pi/a$) [shown in green in panel (d)], with expansion in k (with respect to k_F) and derived η^* values listed, for both the *M1* (orange) and *M2* model (blue). The position of the Fermi surface at k_F is marked in green, and the position of the extrapolated vanishing Fermi velocity for *M2* is marked in dashed blue. (d) The dependence of η^* along the Fermi surface for *M2* (a comparison with other models is shown in the Supplemental Material [27]).

that truncating hopping amplitudes by size or distance is a common strategy to reduce model complexity, but in this case, such a simplification has drastic consequences. Remarkably, as shown in the Supplemental Material [27] the models are virtually indistinguishable if compared on the basis of band structure and Fermi surface alone. Figure 2(c) shows the behavior of v_k^z at a specific point on the Fermi surface, crossing the β sheet, comparing the models *M1* and *M2*. One observes a substantial suppression of the overall magnitude of the velocity (i.e., v_0), along with noticeable changes in the quadratic expansion coefficients. The resulting η^* is accordingly reduced by a factor of 5 from 0.71 to 0.15 eV going from model *M1* to *M2* (and is approximately 0.3 eV in the full model; see the Supplemental Material [27]). The magnitude of η^* thus becomes comparable to expected scattering rates around room temperature, implying that the effect pointed out here is experimentally relevant. To rule out that this effect is confined to a specific region of the Brillouin zone, Fig. 2(d) demonstrates that the suppression occurs along the entire Fermi surface. In the following we illustrate why including t_2^c is essential to reproduce the breakdown of the Drude model as found for the *ab initio* model.

To investigate the effect on the conductivity, we evaluate the models using the full Kubo formalism with a constant scattering rate $\Sigma_m(\omega) \rightarrow -i\eta$, independent of orbital index m . The effect from the nontrivial temperature, frequency, and orbital dependence of the scattering rates will be discussed further below. Figure 3 shows the resulting

resistivities, which show strikingly different behavior. Model *M1* exhibits standard Drude-like behavior, whereas in Model *M2* the inclusion of the next-nearest-neighbor hopping causes a pronounced change: the resistivity increases substantially and develops a maximum at η^* , with a value consistent with the discussion above. The right panel shows contour plots of the c -axis velocity on the Fermi surface, revealing a substantial suppression. This arises from an accidental cancellation due to destructive interference. Namely, in momentum space, the two dominant c -axis hopping amplitudes contribute to the xz - xz hopping Hamiltonian as follows (analogously for yz - yz):

$$\epsilon_{xz,xz}^c = [t_1^c \cos(k_x a/2) + t_2^c \cos(k_x 3a/2)] \times \cos(k_y a/2) \cos(k_z c/2). \quad (7)$$

Near the Brillouin-zone boundary, $k_x \sim \pi/a$, the contributions to the velocity from these two terms have opposite signs. Whenever $t_2^c/t_1^c > 1/3$ there exists an in-plane momentum k at which the c -axis contribution to the hopping vanishes; for the parameters listed in the Supplemental Material [27], this occurs at $k = 0.84\pi/a$ [blue dashed line in Fig. 2(c)], i.e., remarkably close to the Fermi momentum (green). As a result, interference leads to an accidental cancellation of the c -axis velocity in close proximity to the Fermi surface. The Fermi-surface value of the velocity, v_0 , is thus small, making higher-order corrections comparatively important and leading to a correspondingly small value of η^* via Eq. (6).

An important consequence of the strongly momentum-dependent velocity is a strong temperature dependence of the calculated resistivity. In Fig. 3(a), the averaging around the Fermi surface introduced by the thermal broadening of the Fermi window function, $-\partial f/\partial\omega$, has little effect. By contrast, in model *M2* [panel (b)], increasing the temperature strongly suppresses the resistivity, as momenta with larger c -axis velocity begin to contribute significantly to the c -axis response.

These effects survive in the full *ab initio* model when all hopping amplitudes and spin-orbit couplings are included [panel (c)]. Spin-orbit coupling is included as an on-site $L \cdot S$ term in the Wannier Hamiltonian using the effective value $\lambda = 0.2$ eV established previously [18,53]. The corresponding resistivity is significantly reduced compared to the *M2* model, primarily because direct xz - yz interorbital hopping amplitudes lead to an increase of the conductivity near the Brillouin-zone diagonals. This contribution softens the maximum in the total resistivity, but the qualitative physics discussed above remains the same (see the Supplemental Material [27] for further details).

Finally, we turn to a realistic analysis where scattering is temperature dependent as governed by electronic many-body effects. We calculate conductivities using orbital-, frequency-, and temperature-dependent DMFT self-energies in the spectral functions, following Refs. [17,18]. The

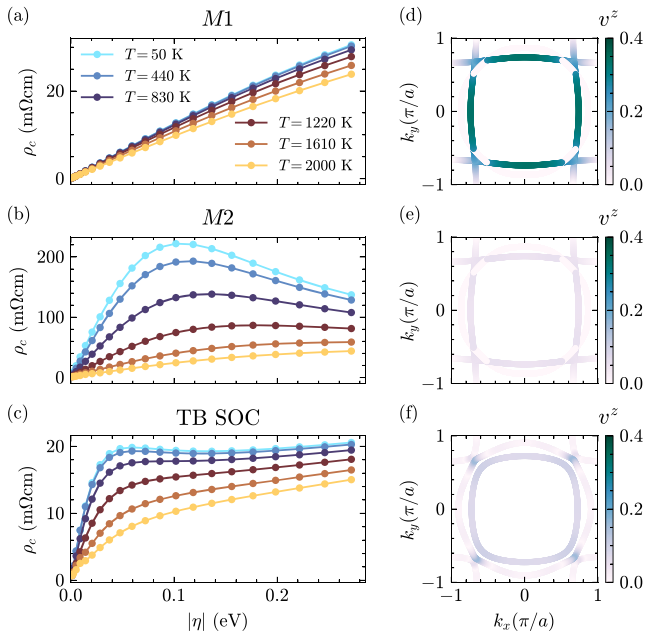


FIG. 3. (a)–(c) Resistivity ρ^c as a function of scattering η for the *M1* (top), *M2* (middle), and full *ab initio* model including SOC (bottom). (d)–(f) Corresponding contour maps of v^z at $k_z = 0.5\pi/c$.

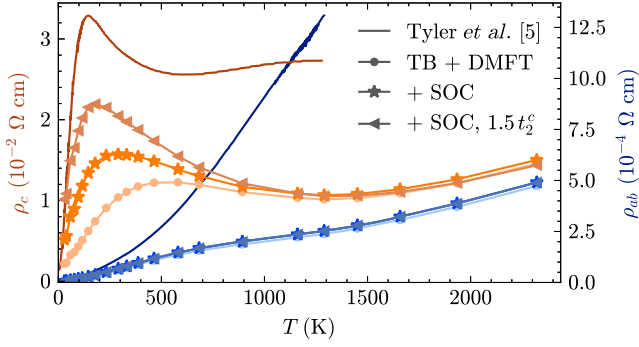


FIG. 4. Comparison of out-of-plane resistivity (left axis, orange) and in-plane resistivity (right axis, blue). Legend symbols refer to both axes; “+SOC” and “+SOC, $1.5t_2^c$ ” denote TB + SOC + DMFT and TB ($1.5t_2^c$) + SOC + DMFT. The c -axis resistivity shows a significant dependence on model details, while the in-plane resistivity remains unaffected.

resulting data, shown in Fig. 4 alongside the digitized experimental results of Ref. [5], reveal strongly anisotropic behavior in the full TB + DMFT model: the in-plane resistivity (blue) increases monotonically, while the out-of-plane resistivity (orange) shows an intermediate maximum. The occurrence of the maximum can be traced back to the inclusion of t_2^c that leads to the accidental cancellation, as discussed above and further outlined in the Supplemental Material [27]. When spin-orbit coupling is included (TB + SOC + DMFT), the maximum shifts to lower temperature, and its peak height increases by about 10% (consistent with its effect for constant η ; see the Supplemental Material [27]). We note that orbital differentiation has little influence on the present results, as assigning the same DMFT self-energy to all t_{2g} orbitals yields nearly identical c -axis resistivities (see the Supplemental Material [27]).

As can be seen in Fig. 2(c), the cancellation is not maximal since v_0 is still finite at the Fermi surface (i.e., the green and the orange dashed lines marking the location of the Fermi surface and the extrapolated vanishing Fermi velocity, respectively, do not coincide). Tuning the region of maximal interference in the Brillouin zone to fall on the Fermi surface by increasing t_2^c by a factor of 1.5 further increases this trend dramatically, producing a sharp peak as is present in the experimental data. These results demonstrate the extreme sensitivity of the c -axis resistivity to very small changes of subdominant hopping matrix elements of the order of only a few meV.

Our theoretical calculations capture some of the salient qualitative aspects of the experiments, making us confident that the proposed mechanism may indeed provide an explanation to the long-standing puzzle of the c -axis resistivity maximum in Sr_2RuO_4 . For instance, it is consistent with the experimental observation that hydrostatic pressure shifts the maximum in ρ_c to higher temperatures while at the same time decreasing the scattering rate [54]. Chemical doping, inflating the Fermi surface toward the point where the accidental cancellation occurs (and

increasing the scattering rate), has the opposite effect [55]. Uniaxial c -axis pressure of 1.5 GPa reduces resistivity at low temperatures by 25% [56], whereas hydrostatic pressure of the same magnitude has little effect [57]. Our proposed mechanism, which relies on a delicate balance between velocities and scattering rate, naturally explains such a sensitivity to external constraints. Preliminary calculations are consistent with these experimental trends, suggesting that moderate c -axis compression affects the crossover more strongly than comparable hydrostatic compression, motivating future experimental and theoretical studies under pressure and strain.

There are, however, significant quantitative differences in both the absolute resistivity values and the position of the maximum, that we now discuss. Both the in-plane and out-of-plane resistivities are underestimated by theory. For the in-plane resistivity, this issue was noted earlier in Ref. [19] where it was also shown that a perturbative treatment of the electron-phonon coupling cannot explain the missing scattering. It is therefore not surprising that the c -axis resistivity is likewise underestimated. Within our framework, the crossover is primarily controlled by the scattering rate. Therefore, an increase of the scattering rate at a given temperature simultaneously leads to higher resistivities and shifts the crossover to lower temperatures, and could hence improve the agreement with experiment further. Missing scattering processes, such as phonons, impurities, or non-local correlations, are therefore expected to affect both discrepancies in a correlated manner. One possible mechanism for this enhanced scattering is an enhancement of the effective electron-phonon coupling by the strong electronic correlations present in this system [58]. We also note that available experimental datasets exhibit noticeable variation in the magnitude and position of the c -axis maximum, while the crossover itself remains robust [5,59].

Does the proposed mechanism apply to other layered materials? As shown in the Supplemental Material [27], data on several other body-centered tetragonal compounds demonstrate that the mechanism indeed applies more broadly. In particular, our data provide a plausible explanation for the similarly anomalous c -axis resistivity maximum in Sr_2RhO_4 [6,60] shown in Fig. 1 and predict plateauing behavior for the c -axis resistivity for Sr_2FeO_4 under pressure but that conversely, in Sr_2MoO_4 the Drude behavior is obeyed with in-plane and out-of-plane resistivity behaving alike. These predictions should be experimentally tested. The mechanism likely extends to other lattices, as long as the c -axis velocity depends strongly on in-plane momentum, which requires sideways vertical hopping amplitudes comparable to nearest-neighbor direct ones. Whereas we did not perform specific calculations in that case, we do notice that in layered cobaltates, a strong dependence of the out-of-plane velocity on the in-plane momentum is indeed seen from the results of Ref. [61].

In summary, our results explain why Drude behavior can break down in a direction-selective way in anisotropic

metals. We show that the c -axis resistivity becomes non-metallic when the scattering exceeds a characteristic value η^* , which is small when the electronic velocities vary strongly with momentum perpendicular to the Fermi surface. The novel mechanism is realized in layered ruthenates and rhodates, where η^* is small due to a cancellation of out-of-plane velocities resulting from opposite contributions of out-of-plane nearest- and next-nearest-neighbor tunneling paths. The mechanism is general and our results call for a reexamination of c -axis resistivity in other layered metals, such as cobaltates. The existence of a maximum in the c -axis resistivity hence does not by itself imply that the transport across layers is incoherent, as often argued. How the newly revealed mechanism for the insulating transport in the c direction combines with other earlier discussed mechanisms, such as interplane defects, remains an important question for future work.

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Data availability—The computed data underlying the findings in the main text are openly available [62].

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