Non-local microwave electrodynamics in ultra-pure PdCoO\textsubscript{2}

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(Dated: May 2, 2022)

The motion of electrons in the vast majority of conductors is diffusive, obeying Ohm’s law. However, the recent discovery and growth of high-purity materials with extremely long electronic mean free paths has sparked interest in non-ohmic alternatives, including viscous and ballistic flow. Although non-ohmic transport regimes have been discovered across a range of materials, including two-dimensional electron gases, graphene, topological semimetals, and the delafossite metals, determining their nature has proved to be challenging. Here, we report on a new approach to the problem, employing broadband microwave spectroscopy of the delafossite metal PdCoO\textsubscript{2} in three distinct sample geometries that would be identical for diffusive transport. The observed differences, which go as far as differing power laws, take advantage of the hexagonal symmetry of PdCoO\textsubscript{2}. This permits a particularly elegant symmetry-based diagnostic for non-local electrodynamics, with the result favouring ballistic over strictly hydrodynamic flow. Furthermore, it uncovers a new effect for ballistic electron flow, owing to the highly faceted shape of the hexagonal Fermi surface. We combine our extensive dataset with an analysis of the Boltzmann equation to characterize the non-local regime in PdCoO\textsubscript{2}. More broadly, our results highlight the potential of broadband microwave spectroscopy to play a central role in investigating exotic transport regimes in the new generation of ultra-high conductivity materials.

There has been significant recent interest in unconventional electronic transport regimes in conductors in which a local relationship between electric current and electric field, as described by Ohm’s law, breaks down. In the absence of frequent momentum-relaxing scattering, non-local effects can develop: when momentum-conserving scattering is sufficiently frequent, electrons flow collectively as a viscous fluid; when neither form of scattering is frequent, electrons propagate ballistically. While viscous and ballistic effects are conceptually distinct, they can lead to similar experimental signatures, creating the possibility of misinterpreting experiments by comparing to theory rooted in one origin or the other. Additional impetus for comparing to theory incorporating both effects comes from the expectation that the hierarchy of scattering rates in most ultra-pure materials places them near the ballistic-to-viscous crossover, rather than at an extreme. Adding to the complexity, while early experiments and theories were based on materials with isotropic Fermi surfaces [1, 2], recent experimental work on the delafossite metals has demonstrated that faceted Fermi surfaces lead to novel ballistic effects dependent on the geometry and orientation of the Fermi surface [3–5].

Differentiating between non-local effects requires transport measurements on length scales comparable to the momentum-relaxing and momentum-conserving mean free paths, $\lambda_{\text{mt}}$ and $\lambda_{\text{mc}}$. To date, this has been approached by studying how DC transport properties vary with the dimensions of micro-structured samples. Here we take a new approach by measuring the AC properties of bulk samples, using the skin effect to impose a tunable length scale, the skin depth. In any metal, AC electromagnetic fields decay as they propagate. The result is that the electromagnetic fields—and resulting current density—are confined to a “skin layer” at the surface. A key advantage to AC measurements, in principle, is that this skin depth is frequency dependent: the DC approach necessitates additional fabrication each time sample dimensions are varied, but AC measurements offer the possibility of continuously varying the skin depth. Although conceptually simple, this requires broadband measurement over the microwave range of the spectrum. To date, high-sensitivity microwave measurements of ultra-high-conductivity materials have typically been limited to fixed-frequency measurements via resonant cavities. Here, we report the use of a bespoke bolometric tech-
FIG. 1. Summary of skin effect regimes. The black lines and accompanying equations give the approximate locations of potential crossovers in the skin effect, as deduced by comparing the relevant length and frequency scales. In blue are the names that have historically been associated with these regimes [6, 7].

The skin effect takes several forms depending on the nature of the electron dynamics within the skin layer, each characterized by unique frequency dependence and symmetry constraints of the AC surface resistance. Mathematically, electromagnetic propagation can be conveniently described via a propagator $A$ which depends on wavevector $q$ and frequency $\omega$:

$$A_{ij}(q, \omega) = \frac{\mu_0}{i \mu_0 \omega \sigma_{ij} + \omega^2/c^2 - q^2}.$$  \hspace{1cm} (1)

The poles of $A$ are solutions to Maxwell’s equations and give the dispersion relations $q(\omega)$ governing the propagation of electromagnetic modes within the metal. The effect of a metal’s conductivity $\sigma$ is to induce a complex wavevector; the skin depth is given by $\delta = 1/\text{Im}(q)$. In most metals, the relationship between the AC electric field and the induced current density is local—the electric current at a given point in space depends only on the electric field at that same point. This is expressed by Ohm’s law:

$$J_i(r, \omega) = \sigma_{ij}(q, \omega) E_j(r, \omega).$$  \hspace{1cm} (2)

The reason Ohm’s law is valid is that frequent scattering randomizes an electron’s momentum on a scale much shorter than the variation of the decaying electric field. This gives rise to the classical skin effect (CSE), for which surface resistance is directly related to the local conductivity:

$$R_i(\omega) = \text{Re} \sqrt{\frac{i \mu_0 \omega}{\sigma_{ii}(\omega)}}.$$  \hspace{1cm} (3)

At low frequency, $R \sim \omega^{1/2}$. The symmetry of $R$ is that of the local conductivity tensor $\sigma_{ij}$, which is set by the crystal system. However, Ohm’s law cannot always be valid: in the absence of momentum-relaxing collisions, an electron’s momentum will depend on the entire history of the varying electric field along its trajectory. This can be resolved via a generalized, non-local version of Ohm’s law using a wavevector-dependent conductivity:

$$J_i(q, \omega) = \sigma_{ij}(q, \omega) E_j(q, \omega).$$  \hspace{1cm} (4)

In this case, $R$ is a wavevector-integrated function of the non-local conductivity, gaining additional anisotropy via the direction of the wavevector $q$:

$$R_i(q, \omega) = \text{Re} \int dq A_{ii}(q, \omega).$$  \hspace{1cm} (5)

An established instance of non-local electrodynamics is the anomalous skin effect (ASE), in which electrons propagate ballistically within the skin layer. It is expected to occur when $\delta \ll \{\lambda_{mr}, \lambda_{mc}\}$ and has historically been associated with $\omega^{3/4}$ behavior. The ASE has been observed experimentally at low temperature in a select number of high-purity elemental metals [8–11]. Another instance of non-local electrodynamics—the viscous skin effect (VSE)—is predicted to occur in an intermediate regime $\sqrt{\lambda_{mr}\lambda_{mc}} \ll \delta \ll \lambda_{mc}$ and is characterized by $\omega^{2/3}$ behavior [7]. To our knowledge, it has not yet been observed experimentally.

In PdCoO$_2$, whose reported low-temperature $\lambda_{mr}$ of 20 $\mu$m is the longest among the delafossites [13], evidence for both viscous [14] and directional ballistic effects [3–5] has been reported in DC transport properties at low temperature. This paper reports on spectroscopic measurements of PdCoO$_2$ at microwave frequencies, where its skin depth is estimated to be comparable to the reported values of $\lambda_{mr}$ and $\lambda_{mc}$. Our measurements were performed at 2 K so as to match previous studies [3–5, 14], using a unique, home-built spectrometer [15]. In Figure 1, we show how the boundaries for potential crossovers in the skin effect vary as a function of $\lambda_{mc}$, assuming the published 2 K value of $\lambda_{mr} = 20 \mu$m. The crystal structure of PdCoO$_2$, with a triangular in-plane lattice, is shown in Figure 2a. Its Fermi surface, shown in Figure 2b, consists of a single sheet which is nearly cylindrical and has a nearly hexagonal cross section. This immediately suggests three extremal wavevector directions for which to perform measurements: $q \parallel (110)$ (“0”), $q \parallel (110)$ (“30”), and $q \parallel (001)$ (“e”). Samples of PdCoO$_2$ grow as platelets with in-plane dimensions around 1 mm and typical thicknesses of 10s of $\mu$m. To reflect the underlying symmetry of the crystal structure, we cut samples to have hexagonal cross section, with lateral dimensions of about 0.5 mm (Figure 2c). We applied a spatially-uniform, microwave-frequency magnetic field parallel to the $c$ axis, inducing eddy currents which flow in loops perpendicular to the magnetic field. This establishes two skin regions: near the edges of the two large hexagonal faces, with wavevector along the $c$ axis, and on the six small rectangular faces, with wavevector...
perpendicular to the $c$ axis. By measuring three different sample geometries—two orientations relative to the crystal structure and two thicknesses—we obtained the necessary data to determine the three components we sought. The fact that the three raw measurements differ (Figure 2d) provides immediate evidence of non-local electrodynamics. In all cases, current flows in the plane perpendicular to the $c$ axis; for local electrodynamics, the triangular in-plane lattice dictates that the conductivity tensor has no in-plane anisotropy and so all three
FIG. 3. Measurements with in-plane wavevector. (a) Data obtained by subtracting the $q \parallel c$ component from the raw data. The different power-law behavior originates from predominantly ballistic propagation within the skin layer coupled with a strongly-facetted Fermi surface, as illustrated in (b) and (c). (b) Calculated surface resistance based on the experimentally-determined Fermi surface, capturing the different power-law behavior of the two orientations. The calculation is in the ballistic limit, i.e. $\gamma_{mc} = \gamma_{mr}$, and uses published values with no free parameters [12]. (c) Illustration of ballistic propagation within the skin layer. Top: There are two main directions of electron propagation, both at an angle to the surface. As frequency increases, the skin depth becomes shallower. The electrons spend an increasingly smaller fraction of a mean free path inside the skin layer, leading to an increasing surface resistance—the anomalous skin effect. Bottom: There are three main directions of electron propagation. Electrons propagating parallel to the sample’s surface spend the entirety of a mean free path within the skin layer, regardless of how shallow the skin depth becomes. Often, this is a negligible fraction of the Fermi surface; in PdCoO$_2$, approximately a third of the Fermi surface propagates parallel to the sample’s surface. The anomalous skin effect is largely suppressed even when the mean free path is much larger than the skin depth.

measurements would be identical. To proceed further, we used electromagnetic simulations to isolate the three individual components.

The measured surface resistance for the two in-plane wavevectors is shown in Figure 3a. Surprisingly, the two orientations exhibit distinct power-law behaviors. A useful property of the viscosity tensor in a plane with six-fold rotational symmetry provides an elegant avenue for differentiating ballistic and viscous effects: in this setting, as is the case in PdCoO$_2$, the in-plane viscosity tensor is isotropic [16]. This implies the qualitative insight that the anisotropy in the surface resistance at 2 K for the two

\[ R / \mu_0 \omega \]

\[ \omega / 2\pi \text{ [GHz]} \]

\[ \hat{j} 0 \rightarrow \hat{q} 30 \]

\[ \vec{u}_k \cdot \vec{q} \]

\[ \hat{j} 0 \rightarrow \hat{q} 30 \]

\[ \hat{j} 30 \rightarrow \hat{q} 0 \]
orientations cannot be due to purely viscous effects.

With this in mind, we turn to the possible ballistic origin of this effect. The standard theory of the anomalous skin effect (i.e., ballistic propagation within the skin layer)—Pippard theory—predicts that any orientation should exhibit $R \sim \omega^{2/3}$, with only the pre-factor being orientation-dependent. Our data is at odds with Pippard theory: While one orientation exhibits behavior close to $\omega^{2/3}$, the other exhibits only a weak deviation from classical behavior. This breakdown of Pippard theory is all the more surprising because—aside from its ubiquity—Pippard theory has previously demonstrated success in describing the behavior of anisotropic Fermi surfaces. Famously, Pippard performed the first ever experimental determination of a Fermi surface by applying his eponymous theory to measurements of the ASE in Cu, revealing deviation from a spherical Fermi surface [17]. Nonetheless, Pippard theory treats Fermi surface geometry phenomenologically, and was originally justified by agreement with more rigorous treatments based on solving the Boltzmann equation for spherical [18] and spheroidal [19] Fermi surfaces.

To model our results, we solved the Boltzmann equation using a realistic parameterization of the Fermi surface of PdCoO$_2$ based on ARPES and quantum oscillation measurements [20]. As seen in Figure 3b, our calculations reproduce the difference in power-law behavior between the two orientations. An intuitive explanation for the difference in power laws comes from applying Pippard’s “ineffectiveness concept” [8] to the Fermi surface of PdCoO$_2$ (Figure 3c): Only those electrons that spend an entirety of a mean free path within the skin layer are effective at screening electromagnetic fields. As the ratio of mean free path to skin depth increases, electrons spend an increasingly small fraction of a mean free path within the skin layer, so the surface resistance becomes increasingly larger than the classical value. Mathematically, this can be described as an effective mean free path for each state $k$, which represents that state’s contribution to the overall conductivity:

$$\lambda_k^\text{eff} = \frac{(\hat{v}_k \cdot \hat{E})^2 \lambda_0}{1 + i(\hat{v}_k \cdot \hat{q}) q \lambda_0}$$

where $\lambda_0$ is the bare mean free path and $\hat{v}_k$ is the unit velocity vector. (Because the present discussion is focused on purely ballistic effects, here we have taken $\lambda_{mr} = \lambda_{mc} = \lambda_0$). In PdCoO$_2$, a third of electrons propagate nearly parallel to the Fermi surface, such that $\hat{v}_k \cdot \hat{q} = 0$. These electrons remain effective at screening regardless of the ratio of mean free path to skin depth, largely suppressing the increase in surface resistance. Indeed, there have been several theoretical works predicting extreme Fermi surface geometries for which Pippard theory would break down [21–23]. To our knowledge, the present results represent the first experimental confirmation of these ideas.

The measured surface resistance for wavevector along the $c$ axis, shown in Figure 4a, exhibits a clear frequency-
dependent crossover from local to non-local electrodynamics. The observation of non-local electrodynamics in this orientation is surprising: as per the ineffectiveness concept, this means that electrons must be able to propagate in and out of the skin layer within a single mean free path. However, the nearly cylindrical geometry of the Fermi surface means that in this orientation, electrons propagate at a shallow angle relative to the skin layer (Figure 4c). PdCoO$_2$ is often described as electronically two-dimensional or quasi-two-dimensional, as supported by its low-temperature resistivity anisotropy of $\rho_c/\rho_a \approx 1000$ [20]. However, in a perfectly two-dimensional material, the ASE would be completely suppressed. Its presence here is a result of the subtle warping of the Fermi surface along $k_z$, as was resolved by quantum oscillations, and highlights the limitations of a purely two-dimensional description of transport properties in PdCoO$_2$. This observation has implications for DC transport measurements. To date, studies have focused on how resistivity varies when restricting in-plane dimensions; these results imply that size effects will also be present while varying thickness along the c axis. The crossover to non-local electrodynamics occurs at an estimated skin depth on the order of 100 nm. This implies that size effects are likely to be especially important to thin films, which have been the subject of recent growth efforts [24–26].

Having identified a ballistic origin for the main features of the three measured surface resistances, we now turn to placing a quantitative bound on the rate of momentum-conserving scattering by comparing to calculations. In Figure 5 we show the calculated power-law behavior of the surface resistance as a function of the relative size of the momentum conserving and momentum relaxing scattering rates $\gamma_{mc}/\gamma_{mr}$. The colour scale displays the evolution of the frequency dependence in this cross-over regime, with the frequency range of our measurement indicated by vertical green lines. Too high a rate of momentum-conserving scattering ($\gamma_{mc}/\gamma_{mr} > 10$) leads to too little anisotropy between $R_{a,q||0}$ and $R_{a,q||30}$. Below this the model captures the higher power law seen for $R_{a,q||0}$. However, for $\gamma_{mc}/\gamma_{mr} < 3$, the calculated anisotropy overestimates that seen in the data. We thus indicate lower and upper bounds in Fig. 5, corresponding to $3 < \gamma_{mc}/\gamma_{mr} < 10$ for measurements at 2K. This raises the question of the source of MC scattering at this temperature. Given that $T \ll T_F$, it is expected that direct electron-electron scattering is negligible [27]. Recent theoretical work has explored electron-phonon scattering as a source of MC scattering [28, 29], though we estimate that it is insufficient to give rise to our experimentally-determined value of $\gamma_{mc}$ [12]. Another recent work has proposed a phonon-mediated electron-electron interaction as a potential source of sufficient MC scattering [30], but such a mechanism in PdCoO$_2$ would be predominantly momentum-relaxing due to electron-electron Umklapp processes. The low temperature of our measurements is suggestive of impurity scattering as the source of the observed $\gamma_{mc}$ [31, 32]. It was noted by Ref. [27] that the evidence for viscous effects in the width-dependent resistivity measurements from Ref. [14] only sets in at low temperature, further corroborating the impurity scattering scenario.

Our experiments demonstrate the utility of broadband microwave spectroscopy in the investigation of non-local electrodynamics. With the foundational measurements of the ASE having been performed at fixed frequencies [8–10, 17], to our knowledge the data in Figure 4a presents the first frequency-resolved measurement of the crossover from local to non-local electrodynamics. In the present work, continuous-frequency measurements were critical to the interpretation of our results: in particular, in identifying a ballistic- rather than viscous-dominated regime, and in revealing the predicted breakdown of Pippard theory as a result of a strongly-faceted Fermi surface. These effects are also technologically relevant, as future applications of ultra-high-conductivity materials are likely
to operate at GHz frequencies. The ASE is known to limit the conductance of interconnects in integrated circuits when operated at these high frequencies [33]. The present results demonstrate that conductance can be improved by aligning interconnects along a direction for which the ASE is suppressed. Finally, to our knowledge, our findings represent the first experimental observation of the ASE outside of elemental metals, suggesting experimental opportunities among new-generation ultra-high-conductivity materials. The interplay of frequency, scattering rates, carrier density, and Fermi surface geometry gives rise to a rich phenomenological landscape for non-local electrodynamics, particularly in the microwave and THz range—which, to date, remains largely unexplored [6, 34–38].

ACKNOWLEDGMENTS

We are grateful to Alex Levchenko for helpful discussions. G.B., T.W.B., J.D., M.H., and D.A.B. acknowledge support from the Max Planck-UBC-UTokyo Center for Quantum Materials and the Canada First Research Excellence Fund, Quantum Materials and Future Technologies Program, as well as the Natural Sciences and Engineering Research Council of Canada (RGPIN-2018-04280). D. V. acknowledges partial support by the Swiss National Science Foundation (SNSF) through the SNSF Early Postdoc Mobility Grant P2GEP2_18145. D.V. and J.S. acknowledge support by the European Commission’s Horizon 2020 RISE program Hydrotronics (Grant No. 873028). P.S. acknowledges the support of the Narendowe Centrum Nauki (NCN) Sonata Bis grant 2019/34/E/ST3/00405 and the Nederlandse Organisatie voor Wetenschappelijk Onderzoek (NWO) Klein grant via NWA route 2. The work in Dresden was in part supported by the Deutsche Forschungsgemeinschaft (DFG) through the Würzburg-Dresden Cluster of Excellence on Complexity and Topology in Quantum Matter – ct.qmat (EXC 2147, project ID 39085490) and the Leibniz Prize programme.

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S1. SAMPLE PREPARATION

A mixture of PdCl$_2$ and CoO was used to grow single crystals of PdCoO$_2$ by the following methathetical reaction in an evacuated quartz ampule: PdCl$_2$ + 2CoO → 2PdCoO$_2$ + CoCl$_2$. The ampule was heated to 1000°C for 12 h and to between 700 and 750°C for 5 d. The product was washed and distilled with water and ethanol to remove CoCl$_2$.

Platelet samples were cut into hexagons using a high-precision wire saw with a 50 µm tungsten wire and 50 nm Al$_2$O$_3$ abrasive suspended in glycerin. The orientation of the crystals was determined via their growth edges, which are oriented perpendicular to the crystallographic axes. A goniometer mounted to the wire saw was used to rotate the samples in between cuts. Samples 1 and 2 were cut from the same original crystal. For Sample 3, a smaller thickness was desired and so it was cut from a separate original crystal from the same batch. The sample dimensions listed in table S1 were determined via optical microscopy and were used for the data analysis in section S2.

| Sample | Hexagonal face area [mm$^2$] | Hexagonal face perimeter [mm] | Thickness [µm] | Cut orientation |
|--------|-----------------------------|-----------------------------|----------------|----------------|
| 1      | 0.187                       | 1.72                        | 88             | $a \parallel x$ |
| 2      | 0.169                       | 1.62                        | 79             | $a \parallel y$ |
| 3      | 0.191                       | 1.76                        | 49             | $a \parallel y$ |

TABLE S1. Sample information. The cut orientations refer to the directions defined in Figure 2 of the main text.

S2. DETERMINATION OF SURFACE RESISTANCES FROM RAW DATA

A. Effective surface resistance

To quantitatively interpret our measurements, it is necessary to account for the extrinsic geometric effects which lead to a difference between the externally applied, spatially-uniform magnetic field $H_0$ and the magnetic field $H(r)$ at the sample’s surface. Consider a sample characterized by two dimensions: $c$, parallel to $H_0$, and $a$, perpendicular to $H_0$. In a conductor for which the skin depth $\delta \ll \{c, a\}$, eddy currents will act such that at the sample’s surface, $H(r)$ is nearly parallel to the surface. For a sample with $c \gg a$, $H(r) \approx H_0$ at the sample’s surface. However, for $c < a$, as in the present work, $H(r)$ will have a non-trivial variation over the sample’s surface. This effect is well-known in the context of magnetization measurements, where it is common practice to account for extrinsic geometric effects using demagnetization factors [1]. In this work, we measured the power dissipated by the sample. In analogy with demagnetization factors, here we examine “power factors” to account for extrinsic geometric effects in our measurements.
The time-averaged power dissipated in a sample is equal to the mean electromagnetic energy entering the sample per unit time:

\[ P = \int \bar{\mathbf{S}} \cdot d\mathbf{A} = \frac{1}{2} \text{Re} \left[ \int d\mathbf{A} \mathbf{E}_t \times \mathbf{H}_t^* \right] = \frac{1}{2} R_{\text{eff}} \int d\mathbf{A} H_t^2 \] (1)

where \( \bar{\mathbf{S}} \) is the time-averaged Poynting vector, \( E_t \) and \( H_t \) are the magnitudes of the fields tangential to the sample’s surface, and \( R_{\text{eff}} \) is the sample’s effective surface resistance. When \( \delta \ll \{c, a\} \), the variation of \( H_t^2 \) over the sample’s surface is that of a perfect conductor of the same shape and is independent of absolute sample size [2]. Therefore, we define the power factor for a given sample shape

\[ \alpha \equiv \frac{1}{AH_0^2} \int H_t^2 dA \] (2)

where \( H_t \) is the tangential magnetic field strength experienced by a perfect conductor of that shape. The surface resistance reported in Figure 1 of the main text was found from measurements of power dissipation via

\[ R_{\text{eff}} = \frac{2P}{\alpha AH_0^2}. \] (3)

\( H_0 \) is the magnitude of the applied magnetic field, as determined via an in-situ reference sample. \( A \) is the sample’s surface area. To confirm the validity of taking \( \delta \ll \{c, a\} \), we have estimated the skin depth across the range of measured frequencies using the classical expression, as shown in fig. S1. We see that the classical skin depth is always < 1 \( \mu \)m, and thus always more than a magnitude less than the smallest sample dimension.

![FIG. S1. Classical skin depth across the range of measured frequencies.](image)

We studied the power factor \( \alpha \) as a function of sample aspect ratio \( c/a \) for several different shapes, as shown in fig. S2. For spheroids, it was possible to treat the problem analytically. For cylinders and hexagonal prisms, finite-element simulations were performed [3]. Because the axial symmetry of cylinders allows for faster convergence of simulation results, this shape was studied over a wide range of aspect ratios. Since the simulation of hexagonal prisms was more resource-intensive, we only focused on directly relevant aspect ratios. Within the range of aspect ratios relevant to the samples studied here, all three shapes yielded similar results.

### B. Separating components

Because \( \delta \ll \{c, a\} \), \( R_{\text{eff}} \) can be viewed as a sum of two independent components: \( R_\perp \) comes from the faces perpendicular to \( \mathbf{H}_0 \) (the two large hexagonal faces) and \( R_\parallel \) comes from the faces parallel to \( \mathbf{H}_0 \) (the six small rectangular faces). The relative contribution of the two components is set by a weight \( w_\perp \):

\[ R_{\text{eff}} = w_\perp R_\perp + (1 - w_\perp)R_\parallel \] (4)
The weight $w_\perp$ can be found via the variation of the tangential magnetic field strength over the surface of a perfect conductor of the same shape:

$$w_\perp = \frac{\int A_\perp H_t^2 dA}{\int A_\perp + A_\parallel H_t^2 dA}$$

where $A_\perp$ ($A_\parallel$) is the area of the perpendicular (parallel) faces. The weights determined by our finite-element simulations are shown in fig. S3.

\[ R_1 = w_1 R_{a,c} + (1 - w_1) R_{30,0} \]

\[ R_2 = w_2 R_{a,c} + (1 - w_2) R_{0,30} \]

\[ R_3 = w_3 R_{a,c} + (1 - w_3) R_{0,30} \]
where on the left side of the equations, \( R_i \) refers to \( R_{\text{eff}} \) for Sample \( i \), and on the right side of the equations, the two subscripts refer to the directions \( \hat{J}, \hat{q} \). Using eqs. (7) and (8), \( R_{a,c} \) was found from the measurements of Samples 2 and 3:

\[
R_{a,c} = \frac{w_2R_3 - w_3R_2}{w_2(1 - w_3) - (1 - w_2)w_3}. \tag{9}
\]

\( R_{a,c} \) was then used to determine \( R_{30,0} \) and \( R_{0,30} \) from eqs. (6) and (7):

\[
R_{30,0} = \frac{R_1 - w_1}{1 - w_1} \tag{10}
\]

\[
R_{0,30} = \frac{R_2 - w_2R_{a,c}}{1 - w_2}. \tag{11}
\]

The values of \( \alpha \) and \( w_\perp \) that were used in eqs. (3) and (9) to (11) to arrive at the data in the main text are summarized in table S2. For Samples 1 and 2, we used the values of \( \alpha \) and \( w_\perp \) from our simulations. For Sample 3, some adjustment relative to the simulation values was necessary in order to obtain physically plausible results for \( R_{a,c}, R_{30,0}, \) and \( R_{0,30} \). We found that the value of \( R_3 \) determined using the simulation value \( \alpha^\text{sim}_3 \) was larger than expected. Specifically, the low-frequency asymptote of \( R_3 \) was a factor of \( \approx 1.6 \) greater than that of \( R_2 \). There are a few potential explanations for this:

1. We failed to accurately account for geometric effects in our simulations, with \( \alpha^\text{sim}_3 \) being smaller than the true value of \( \alpha_3 \). This difference in magnitude would result if the power ratio \( \alpha \) increases more rapidly with decreasing aspect ratio \( c/a \) than captured by our simulations.

2. Sample 3, being cut from a different original sample, has a higher residual scattering rate than Sample 2. In the classical case, \( R \propto \sqrt{1/\tau} \), so a factor of 1.8 difference in the low-frequency asymptotes corresponds to a factor of \( \approx 2.6 (\approx 1.6^2) \) difference in residual scattering rates. This is consistent with the variation in residual resistivities reported by Nandi et al. [4].

3. There is an intrinsic difference in frequency dependence that persists to the lower end of our frequency range. However, this scenario is unlikely because it would require that \( R_{a,c} \) have an unrealistically large magnitude.

We found that in order to get reasonable results, it was necessary to scale \( R_3 \) so that it was comparable to \( R_2 \) at the lowest frequencies. Otherwise, we got negative values for \( R_{0,30} \) (unless we used extreme values of \( w_2 \) and \( w_3 \)), which is unphysical. We found that values of \( \alpha_3 \) close to 4 led to reasonable results. We also found that if we used too small a value for \( w_3 \), the resultant \( R_{0,30}(\omega) \) contained a kink above which it displayed power law behaviour with exponent \( < 1/2 \), which is unphysical behaviour. We found that values of \( w_3 \) close to 0.7 produce a smooth result for \( R_{0,30} \), whereas simulations found that \( w_3 \approx 0.45 \). As with \( \alpha \), the difference between empirical and simulated results may indicate that simulations are underestimating the magnitude of extrinsic geometric effects for small aspect ratios. Despite that it was necessary to adjust \( \alpha_3 \) and \( w_3 \) relative to the simulation values to get physically reasonable results, in both cases there is a range of values which lead to qualitatively similar results.

| Sample | \( \alpha \) | \( w_\perp \) |
|--------|--------------|--------------|
| 1      | 2            | 0.4          |
| 2      | 2            | 0.4          |
| 3      | 4            | 0.7          |

| TABLE S2. Data calibration parameters. |

S3. SURFACE RESISTANCE CALCULATIONS

A. Distribution function

We start with the Boltzmann equation for the evolution of the electronic distribution function \( f_k(r,t) \) in the presence of a spatially- and time-varying electric field \( \vec{E}(r,t) \):

\[
\left[ \frac{\partial}{\partial t} + \vec{r} \cdot \nabla \vec{r} + \vec{k} \cdot \nabla \vec{k} \right] f_k(r,t) = -C_k [f_k(r,t)] \tag{12}
\]
with semi-classical equations of motion

\[ \dot{r} = v_k = \frac{1}{\hbar} \nabla_k E_k \]

and

\[ \dot{k} = -\frac{e}{\hbar} E(r, t) \]

and where \( C_k[f_k] \) is the collision integral. We expand the distribution function around its equilibrium value \( f_0 \):

\[ f_k = f_0 + w_k \psi_k \]

with

\[ w_k = k_B T \left( -\frac{\partial f_0}{E_k} \right) = f_0 (1 - f_0) \]

We take the Fourier transform of eq. (12) and keep terms to linear order in \( \psi_k \) to obtain

\[ [-i\omega + iv_k \cdot q] \psi_k(q, \omega) + \frac{e}{k_B T} E(q, \omega) \cdot v_k = -\hat{C}_k \psi_k(q, \omega) \]

where \( \hat{C} \) is the linearized collision operator

\[ \hat{C} \psi_k = \frac{1}{w_k} \int_{k'} \delta C_k \psi_{k'} \psi_{k'} \]

Let \( \psi_k \) be an element of a function space with inner product

\[ \langle \phi | \psi \rangle = \int_k w_k \phi_k^* \psi_k \]

We use as a basis the complete and orthonormal set of eigenfunctions \( \chi_{k,m} \) of the collision operator:

\[ \hat{C} \chi_{k,m} = \gamma_{k,m} \chi_{k,m} \]

with

\[ \sum_m \langle \chi_{k,m} | \chi_{k,m} \rangle = 1 \]

and

\[ \langle \chi_{k,m} | \chi_{k,m'} \rangle = \delta_{m,m'} \]

We assume that the eigenfunctions \( \chi_{k,m} \) include

\[ \chi_{k,0} = c_0 \]

and

\[ \chi_{k,i} = c_i c_0 \hat{v}_{k,i} \]

where \( i \in \{x, y, z\} \) and that the eigenvalue spectrum is given by

\[ \gamma_{k,m} = \begin{cases} 0 & m = 0 \\ \gamma_{k,i} & m = i \\ \gamma_{mc} & \text{otherwise.} \end{cases} \]

This describes a scenario in which collisions conserve charge, relax momentum in the \( i \) direction at a rate \( \gamma_{mc} \), and relax all other modes at a rate \( \gamma_{mc}^{\text{mc}} \). Using eq. (21), we may write the collision operator as

\[ \hat{C} = \gamma_{mc}^{\text{mc}} (1 - |\chi_{k,0} \rangle \langle \chi_{k,0}|) - \sum_i \delta \gamma_{k,i} |\chi_{k,i} \rangle \langle \chi_{k,i}| \]
where $\delta \gamma_{k,i} \equiv \gamma^m_k - \gamma^{mr}_{k,i}$.

Throughout, we will use that

$$\int k \cdots \equiv \frac{2}{(2\pi)^d} \int d k \cdots = \frac{2}{(2\pi)^d \hbar} \int_0^\infty d \epsilon \int_{S(\epsilon)} \frac{d S}{v_k} \cdots. \quad (27)$$

Furthermore, we will assume that $T \ll T_F$ such that

$$\int -\partial f_k \cdots = \frac{2}{(2\pi)^d \hbar} \int S_F \frac{d S}{v_k} \cdots \quad (28)$$

where $S_F$ is the Fermi surface $S(E_k = E_F)$. Finally, for simplicity, we will assume that on the Fermi surface the magnitude of the velocity is isotropic: $v_k = v_F \hat{v}_k$ for $k$ on $S_F$.

The constants $c_0$ and $c_i$ are determined by eq. (22). We find

$$\frac{1}{c_0^2} = \frac{2k_B T}{(2\pi)^d \hbar v_F} S_F \quad (29)$$

and

$$\frac{1}{c_i^2} = \int_{S_F} d S \hat{v}_{ki}^2 \quad (30)$$

where

$$S_F \equiv \int_{S_F} d S. \quad (31)$$

We can now rewrite the collision integral as

$$\hat{C} \psi_k = \gamma^m_k \psi_k - \gamma^m_k n_0 - \sum_i c_i^2 \delta \gamma_{k,i} \hat{v}_{ki} p_i \quad (32)$$

where

$$n_0 \equiv \int_{S_F} d S \frac{S}{S_F} \psi_k \quad (33)$$

and

$$p_i \equiv \int_{S_F} d S \frac{S}{S_F} \hat{v}_{ki} \psi_k \quad (34)$$

The solution to the Boltzmann equation is then

$$\psi_k(q, \omega) = \frac{-\frac{e}{k_B T} E(q, \omega) \cdot v_k + \gamma^m_k n_0(q, \omega) + \sum_i c_i^2 \delta \gamma_{k,i} \hat{v}_{ki} p_i(q, \omega)}{\gamma^m_k - i\omega + iv_F \hat{v}_k \cdot q} \quad (35)$$

Because we are ultimately interested in finding the transverse conductivity, so we take $E \perp q$ with $E \parallel \hat{\alpha}$ and $q \parallel \hat{\beta}$. We define

$$\langle A \rangle \equiv \int_{S_F} d S \frac{S}{S_F} \frac{A}{\gamma^m_k - i\omega + iv_F \hat{v}_k \cdot q} \quad (36)$$

We find that

$$\left( \begin{array}{cccc} \gamma^m_k (1) - 1 & c_i^2 \delta \gamma_{k,\beta} \hat{v}_{k\beta} & c_i^2 \delta \gamma_{k,\alpha} \hat{v}_{k\alpha} & c_i^2 \delta \gamma_{k,\gamma} \hat{v}_{k\gamma} \\ c_i^2 \delta \gamma_{k,\beta} \hat{v}_{k\beta} & c_i^2 \delta \gamma_{k,\alpha} \hat{v}_{k\alpha} & c_i^2 \delta \gamma_{k,\gamma} \hat{v}_{k\gamma} & c_i^2 \delta \gamma_{k,\gamma} \hat{v}_{k\gamma} \\ c_i^2 \delta \gamma_{k,\alpha} \hat{v}_{k\alpha} & c_i^2 \delta \gamma_{k,\gamma} \hat{v}_{k\gamma} & c_i^2 \delta \gamma_{k,\gamma} \hat{v}_{k\gamma} & c_i^2 \delta \gamma_{k,\alpha} \hat{v}_{k\alpha} \\ c_i^2 \delta \gamma_{k,\gamma} \hat{v}_{k\gamma} & c_i^2 \delta \gamma_{k,\gamma} \hat{v}_{k\gamma} & c_i^2 \delta \gamma_{k,\alpha} \hat{v}_{k\alpha} & c_i^2 \delta \gamma_{k,\gamma} \hat{v}_{k\gamma} \end{array} \right) \cdot \begin{pmatrix} n_0 \\ p\beta \\ p\alpha \\ p\gamma \end{pmatrix} = \frac{e}{k_B T} E_y v_F \begin{pmatrix} \hat{v}_{k\alpha} \\ \hat{v}_{k\beta} \hat{v}_{k\alpha} \\ \hat{v}_{k\gamma} \hat{v}_{k\alpha} \\ \hat{v}_{k\gamma} \hat{v}_{k\alpha} \end{pmatrix} \quad (37)$$
and assuming three mirror planes, we get

\[
\begin{pmatrix}
\gamma_{mc}(1) - 1 & c_\beta^2 \delta \gamma_{k,\beta} \langle \hat{v}_{k\beta} \rangle & 0 & 0 \\
\gamma_{mc} \langle \hat{v}_{k\beta} \rangle & c_\beta^2 \delta \gamma_{k,\beta} \langle \hat{v}_{k\beta}^2 \rangle - 1 & 0 & 0 \\
0 & 0 & c_\alpha^2 \delta \gamma_{k,\alpha} \langle \hat{v}_{k\alpha}^2 \rangle - 1 & 0 \\
0 & 0 & 0 & c_\gamma^2 \delta \gamma_{k,\gamma} \langle \hat{v}_{k\gamma}^2 \rangle - 1
\end{pmatrix}
\begin{pmatrix}
\langle n_0 \rangle \\
\langle p_0 \rangle \\
\langle p_\beta \rangle \\
\langle p_\alpha \rangle \\
\langle p_\gamma \rangle
\end{pmatrix} = \frac{e}{k_B T} E_y v_F \begin{pmatrix}
0 \\
0 \\
0 \\
0 \\
0
\end{pmatrix}
\] (38)

To ensure that \( \psi_k = 0 \) when \( E = 0 \), we must also have \( n_0 = p_\beta = 0 \). For \( p_\alpha \), we have

\[
\psi_k = \frac{-eE v_F}{k_B T} \frac{1}{\gamma_{mc} - i\omega + iqv_F \hat{v}_{k\beta}} \frac{\hat{v}_{k\alpha}}{1 - c_\alpha^2 \delta \gamma_{k,\alpha} \langle \hat{v}_{k\alpha}^2 \rangle}.
\] (40)

**B. Non-local transverse conductivity**

Electrical current is given by

\[
J = -e \int_k \mathbf{v}_k \mathbf{j}_k.
\] (41)

Via

\[
J_i(q, \omega) = \sigma_{ij}(q, \omega) E_j(q, \omega)
\] (42)

and assuming \( \delta \gamma_{k,\alpha} \) to be \( k \)-independent, we find the non-local transverse conductivity for \( E \parallel \hat{\alpha} \) and \( q \parallel \hat{\beta} \) as

\[
\sigma(q, \omega) = \epsilon_0 \Omega_p^2 \frac{G_0(q, \omega)}{1 - c_\alpha^2 \delta \gamma_{0,\alpha} G_0(q, \omega)}
\] (43)

where

\[
G_0(q, \omega) \equiv \langle \hat{v}_{k\alpha}^2 \rangle = \int_{S_F} dS \frac{\hat{v}_{k\alpha}^2}{\gamma_{mc} - i\omega + iqv_F \hat{v}_{k\beta}}.
\] (44)

and

\[
\Omega_p^2 = \sum_i \omega_{p,ii}^2
\] (45)

where plasma frequency is given by

\[
\epsilon_0 \omega_{p,ii}^2 = \frac{\epsilon_0^2 v_F^2}{\epsilon_0^2 k_B T} \int_{S_F} dS \frac{\hat{v}_{ki}^2}{S_F}.
\] (46)

**C. Parameterization of Fermi surface**

For a Fermi surface \( S_F \) parametrized by the Fermi vector \( k_F(g, h) \) with \( g \in \{g_1, g_2\} \) and \( h \in \{h_1, h_2\} \), we define the vector \( \mathbf{n} \) as

\[
\mathbf{n}(g, h) = \frac{\partial k_F(g, h)}{\partial g} \times \frac{\partial k_F(g, h)}{\partial h}.
\] (47)

Then the Fermi surface integral is given by

\[
\int_{S_F} dS \cdots = \int_{g_1}^{g_2} dg \int_{h_1}^{h_2} dh \mathbf{n}(g, h) \cdots
\] (48)

with \( n \equiv |\mathbf{n}| \). The unit vector normal to the Fermi surface (parallel to the Fermi velocity) is given by

\[
\hat{\mathbf{n}}(g, h) = \frac{\mathbf{n}(g, h)}{\mathbf{n}(g, h)}
\] (49)
D. Surface resistance

For specular scattering of electrons at the sample’s surface, surface impedance is given by [5]

\[ Z = i\mu_0 \omega \frac{2}{\pi} \int_0^\infty dq \left[ i\mu_0 \omega \sigma(q, \omega) + \frac{\omega^2}{c^2 q^2} - q^2 \right]^{-1} \]  

while for diffuse scattering, it is given by [6]

\[ Z = i\mu_0 \omega \pi \left( \int_0^\infty dq \ln \left[ i\mu_0 \omega \sigma(q, \omega) \right] \frac{\omega^2}{c^2 q^2} - q^2 \right) \left( \frac{1}{\omega^2 c^2 q^2} - 1 \right)^{-1}. \]

Finally, surface resistance is given by

\[ R = \text{Re}(Z). \]  

E. Calculations for PdCoO$_2$

We used the Fermi surface parameterization from Hicks et al. [7]:

\[ k_F(\phi, \phi_0, k_z) = \rho(\phi - \phi_0, k_z)[\cos \phi \hat{i} + \sin \phi \hat{j}] + k_3 \hat{k} \]

where

\[ \rho(\phi - \phi_0, k_z) = \sum_{\mu, \nu} k_{\mu,\nu} \cos[\mu(\phi - \phi_0)] \begin{cases} \sin[\nu dk_z] & k_{31} \\ \cos[\nu dk_z] & \text{otherwise} \end{cases} \]  

with \( d = c/3 \) where \( c = 17.743 \) Å and with the Fermi surface harmonics listed in table S3. The angle \( \phi_0 \) sets the in-plane rotation of the Fermi surface relative to the coordinate system. To take advantage of the simplifications arising from three mirror planes, we set \( k_{31} = 0 \). We assumed diffuse surface scattering when calculating the surface resistance. We used the reported experimental parameters as given in table S4. We took \( \gamma_{mr}^{\text{mr}} = \gamma_{kx}^{\text{mr}} = \gamma_{kx}^{\text{mc}} \) using the value from table S4, and took \( \gamma_{k}^{\text{mc}} = \gamma_{mc}^{\text{mc}} \) with \( \gamma_{mc}^{\text{mc}} \) as a free parameter. Calculations for varying \( \gamma_{mc}^{\text{mc}} \) are shown in fig. S4.

| \( \mu \) | \( \nu \) | \( k_{\mu,\nu} \) |
|---|---|---|
| 0 | 0 | 0.9538 |
| 6 | 0 | 0.040 |
| 12 | 0 | 0.007 |
| 0 | 1 | 0.0107 |
| 0 | 2 | -0.009 |
| 3 | 1 | 0.0010 |

TABLE S3. Harmonics for parameterization of Fermi surface of PdCoO$_2$ from Hicks et al. [7].

| Parameter | Value |
|---|---|
| \( v_F \) | \( 7.5 \times 10^5 \) m/s |
| \( \omega_{p,ab} \) | \( 7.2 \times 10^{15} \) Hz |
| \( \rho_{ab}(T = 2 \text{ K}) \) | \( 7.5 \) nΩ cm |
| \( \gamma^{\text{mr}} (T = 2 \text{ K}) \) | \( 34 \) GHz |

TABLE S4. Parameters for PdCoO$_2$. All parameters are directly from Hicks et al. [7], except for \( \gamma^{\text{mr}} \) which was found as \( \gamma^{\text{mr}} = \epsilon_0 \omega_{p,ab}^2 \rho_{ab} \).
S4. ESTIMATE OF MOMENTUM-CONSERVING ELECTRON-PHONON SCATTERING

Here we estimate the rate of momentum-conserving electron-phonon scattering in PdCoO$_2$. First, we obtained an experimental MR scattering rate as $\gamma_{\text{exp}}^1 = \rho_{xx}/\epsilon_0 \omega_{p,xx}^2$ using $\rho_{xx}$ for a 155 $\mu$m channel from Nandi et al. [4] and using $\omega_{p,xx} = 7.2 \times 10^{15}$ Hz from Hicks et al. [7]. Next, we performed fits to $\gamma_{\text{exp}}^1$. The results are shown in fig. S5 and the fit parameters given in table S5. To find the MR electron-impurity scattering rate $\gamma_{\text{imp}}^1$, we fit $\gamma_{\text{exp}}^1$ to a constant over the range $2 \text{ K} < T < 10 \text{ K}$. Next we considered electron-phonon scattering. It has previously been noted that at high temperature $\gamma_{\text{exp}}^1 \propto T^\alpha$ with $\alpha > 1$, in contrast with expectation that $\alpha = 1$ within the Bloch-Grüneisen treatment of electron-acoustic phonon scattering. This discrepancy has been attributed to electron-optical phonon scattering [7, 8]. Therefore, we fit $\gamma_{\text{exp}}^1 - \gamma_{\text{imp}}^1$ to a sum of Einstein and Debye contributions using

$$h\gamma_1^{\text{Ein}} = \frac{\pi}{2} \lambda_E k_B T_E \frac{T_E/T}{\sinh^2(T_E/2T)}$$

and

$$h\gamma_l^{\text{Deb}} = 4\pi \lambda_D k_B T \left(\frac{T}{T_D}\right)^2 \int_0^{T_D/T} dx \frac{x^3}{\cosh(x) - 1} \left[1 - P_l \left(1 - (T/T_D)^2 x^2\right)\right]$$

with $l = 1$ (cf. eq. A.45 from Levchenko and Schmalian [9]). The fit returned Debye and Einstein temperatures $T_D$ and $T_E$ and transport electron-phonon couplings $\lambda_D$ and $\lambda_E$. Following Hicks et al. [7], the fit was performed over the range $60 \text{ K} < T < 300 \text{ K}$. Having determined $T_D$ and $\lambda_D$, we then used eq. (56) to determine $\gamma_l^{\text{Deb}}$ for all $l$, as shown in fig. S6. We see that at 2 K, all $\gamma_l$ for electron-Debye phonon scattering are less than the experimentally determined MR scattering rate. Therefore, electron-phonon scattering is unlikely to be responsible for the MC scattering inferred from our measurements.

| Parameter | Value |
|-----------|-------|
| $\gamma_{\text{imp}}^1$ | 39 GHz |
| $\gamma_1^{\text{imp}}$ | 1.5 |
| $\gamma_1^{\text{Deb}}$ | 0.030 |
| $T_D$ | 331 K |
| $T_E$ | 1120 K |
| $\lambda_D$ | 0.049 |
| $\lambda_E$ | 0.030 |

TABLE S5. Fit parameters.

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FIG. S5. Momentum-relaxing scattering rates determined by fitting to bulk resistivity measurements.

FIG. S6. Spectrum of scattering rates for electron-Debye phonon scattering compared with 2 K momentum-relaxing scattering rate.

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