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Extremely Large Magnetoresistance in the Nonmagnetic Metal \(\text{PdCoO}_2\)

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Extremely large magnetoresistance is realized in the nonmagnetic layered metal \(\text{PdCoO}_2\). In spite of a highly conducting metallic behavior with a simple quasi-two-dimensional hexagonal Fermi surface, the interlayer resistance reaches up to 35 000% for the field along the [110] direction. Furthermore, the temperature dependence of the resistance becomes nonmetallic for this field direction, while it remains metallic for fields along the [110] direction. Such severe and anisotropic destruction of the interlayer coherence by a magnetic field on a simple Fermi surface is ascribable to orbital motion of carriers on the Fermi surface driven by the Lorentz force, but seems to have been largely overlooked until now.

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Finding new systems exhibiting a large resistance change by a magnetic field has driven crucial progress in both condensed matter physics and device application. The most well-known examples are the giant magnetoresistance in magnetic multilayers [1,2] and colossal magnetoresistance in manganites [3,4], both of which rely on coupling between spin configuration and charge transport. Even among nonmagnetic materials, the magnetoresistance (MR) may become large in systems such as semimetals with Fermi surface (FS) compensation [5–7]. Here we report extremely large MR and metal-nonmetal crossover in a highly conducting nonmagnetic metal \(\text{PdCoO}_2\). Realized with a simple quasi-two-dimensional (quasi-2D) FS, this huge MR simply originates from the orbital motion of the highly conducting electrons.

The delafossite compound \(\text{PdCoO}_2\) has a layered hexagonal structure with the space group \(\text{R}3\text{m}\) consisting of alternating stacking of Pd triangular layers and CoO\(_2\) triangular slabs [8–17]. The metallicity is predominantly attributed to the Pd 4d electrons [14,18,19]; the densities of states of Co and O, as well as Pd 5s, are very low at the Fermi level [17]. The band calculations indicate that the FS consists of a single rounded hexagonal prism [11,13,15–17]. The electronic specific-heat coefficient \(\gamma\approx 1\text{mJ/molK}^2\), indicating that electron correlation in \(\text{PdCoO}_2\) is not strong. The carrier density estimated from the Hall coefficient is \(n_{\text{obs}}\approx 1.6 \times 10^{22}\text{ cm}^{-3}\), consistent with one electron carrier per formula unit [12,21]. This agreement indicates that this oxide is a high-carrier-density, free-electron-like system. Reflecting the 3\(d^9\) low-spin state of Co\(^{3+}\) with zero total spin, \(\text{PdCoO}_2\) is nonmagnetic in the whole temperature and field ranges investigated [10,12,22,23].

In this Letter, we report that the electrical resistivity along the \(c\) axis \(\rho_c\) is surprisingly enhanced by the application of the in-plane magnetic field along the [110] direction, reaching 35 000% of the zero-field resistance at 2 K and 14 T, and continues to increase linearly with field. We also found that the temperature dependence of \(\rho_c\) for this field direction exhibits a metal-nonmetal crossover at around 120 K. This behavior is sensitively suppressed by a small tilt or rotation of the magnetic field. Semiclassical calculations of the MR based on the tight-binding band structure of \(\text{PdCoO}_2\) qualitatively reproduce the observed field-angle dependence of the MR. Thus, this extremely large MR is a generic but overlooked phenomenon in a highly conducting system with a simple quasi-2D FS; it is due to the destruction of interlayer coherence by an orbital motion of the carriers on the FS driven by the Lorentz force.

Single crystals of \(\text{PdCoO}_2\) used in this study are grown by a stoichiometric self-flux method. They were characterized by the powder x-ray diffraction and the energy dispersive x-ray analysis [12]. The out-of-plane resistivity \(\rho_c\) was measured on single-domain crystals with the hexagonal plate shape having an area of about 5 mm\(^2\) and a thickness of about 0.03 mm. A conventional four-probe method was employed from 2 to 300 K in a field up to 14 T with a commercial apparatus (Quantum Design, model PPMS). We attached two gold wires (\(\phi = 25\mu\text{m}\)) for current and voltage (\(I_+\) and \(V_+\)) to one \(ab\) surface of the crystal and another two wires (\(I_-\) and \(V_-\)) to the other side. The wires were attached with silver paste (Dupont, 6838). The silver paste for the current electrodes was made into a ring shape to cover as wide an area of the crystal surfaces as possible, while the voltage leads...
were placed at the center of the ring of the current
were placed at the center of the ring of the current
electrodes, as shown in the inset of Fig. 1(b). This wire
electrodes, as shown in the inset of Fig. 1(b). This wire
arrangement is justified for anisotropic metals, and is
arrangement is justified for anisotropic metals, and is
indeed often used for layered compounds [24,25]. The residual resistivity ratio, \( \rho_c(300\,K)/\rho_c(2\,K) \), in zero field
indeed often used for layered compounds [24,25]. The residual resistivity ratio, \( \rho_c(300\,K)/\rho_c(2\,K) \), in zero field
is about 120 and guarantees a high sample quality. The sample crystal was placed with its basal \( ab \) plane parallel
to the rotating stage of a single axis rotator, which controls
the azimuthal orientations of the magnetic field. To check
to the rotating stage of a single axis rotator, which controls
the possibility of a magnetic phase transition induced by
the magnetic field, the specific heat (\( C_p \)) and dc magnetic
the magnetic field, the specific heat (\( C_p \)) and dc magnetic
susceptibility (\( M/H \)) were measured with a commercial
susceptibility (\( M/H \)) were measured with a commercial
calorimeter and with a SQUID magnetometer on a group of
calorimeter and with a SQUID magnetometer on a group of
aligned crystals.

Figure 1 presents the temperature dependence of \( \rho_c \) for
Figure 1 presents the temperature dependence of \( \rho_c \) for
fields along the \([110]\) and \([110]\) directions. With magnetic
fields along the \([110]\) and \([110]\) directions. With magnetic
fields in the \( ab \) plane, \( \rho_c \) exhibits substantial enhancement
fields in the \( ab \) plane, \( \rho_c \) exhibits substantial enhancement
on cooling. In particular, for fields along the \([110]\)
don cooling. In particular, for fields along the \([110]\)
direction, \( \rho_c \) even turns into nonmetallic, exhibiting huge
direction, \( \rho_c \) even turns into nonmetallic, exhibiting huge
enhancement. It reaches 350 times larger than the zero-
enhancement. It reaches 350 times larger than the zero-
field resistivity value at 2 K in 14 T. This enhancement is
field resistivity value at 2 K in 14 T. This enhancement is
comparable to those of giant magnetoresistance and colos-
comparable to those of giant magnetoresistance and colos-
sal magnetoresistance materials [3,4]. However, in sharp
sal magnetoresistance materials [3,4]. However, in sharp
contrast with these materials, \( \text{PdCoO}_2 \) exhibits a positive
contrast with these materials, \( \text{PdCoO}_2 \) exhibits a positive
field response. Moreover, as confirmed by in-field mea-
field response. Moreover, as confirmed by in-field mea-
surements of \( C_p \) and \( M/H \), we did not observe any anom-
surements of \( C_p \) and \( M/H \), we did not observe any anom-
olies suggesting magnetic or charge order at temperatures
olies suggesting magnetic or charge order at temperatures
where the enhancement of \( \rho_c \) develops.

When rotating the field from the \([110]\) to \([110]\) directions,
When rotating the field from the \([110]\) to \([110]\) directions,
\( \rho_c \) sharply drops and switches to metallic, although
\( \rho_c \) sharply drops and switches to metallic, although
it slightly deviates from a typical \( T \)-linear dependence in
it slightly deviates from a typical \( T \)-linear dependence in
zero field. A small hump structure exists at temperatures
zero field. A small hump structure exists at temperatures
where \( \rho_c(H \parallel [110]) \) starts to increase. As is clear from
where \( \rho_c(H \parallel [110]) \) starts to increase. As is clear from
Figs. 1(c) and 1(d), the in-plane field anisotropy emerges at
Figs. 1(c) and 1(d), the in-plane field anisotropy emerges at
a temperature which depends on the field strength. In fact,
a temperature which depends on the field strength. In fact,
\( d\rho_c/dT \) for \( H \parallel [110] \) and \( H \parallel [110] \) starts to deviate from
\( d\rho_c/dT \) for \( H \parallel [110] \) and \( H \parallel [110] \) starts to deviate from
each other at about 100 K in 5 T, and about 180 K in 14 T.
each other at about 100 K in 5 T, and about 180 K in 14 T.
The anisotropy ratio, \( \delta \equiv \rho_c(H \parallel [110])/\rho_c(H \parallel [110]) \),
The anisotropy ratio, \( \delta \equiv \rho_c(H \parallel [110])/\rho_c(H \parallel [110]) \),
is up to 29 (2900%) at 2 K in 14 T. This value is larger
is up to 29 (2900%) at 2 K in 14 T. This value is larger
than the field anisotropy of ordinary nonmagnetic metals
than the field anisotropy of ordinary nonmagnetic metals
with simple sphere FSs, such as alkali metals (< 1%) [26].
with simple sphere FSs, such as alkali metals (< 1%) [26].
It is also noteworthy that this anisotropy value is even
It is also noteworthy that this anisotropy value is even
larger than a typical ratio of the anisotropic magnetoresis-
larger than a typical ratio of the anisotropic magnetoresis-
tance of magnetic metals (< 2%-3%) [27].
tance of magnetic metals (< 2%-3%) [27].

Figure 2 shows the field dependence of the MR ratio,
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\( \Delta \rho_c(H)/\rho_c(H=0) = \rho_c(H)/\rho_c(H=0) - 1 \), for the \([110]\)
\( \Delta \rho_c(H)/\rho_c(H=0) = \rho_c(H)/\rho_c(H=0) - 1 \), for the \([110]\)
and \([110]\) field directions. As observed in the temperature
and \([110]\) field directions. As observed in the temperature
dependence, \( \Delta \rho_c(H)/\rho_c(H=0) \) for the \([110]\) field
dependence, \( \Delta \rho_c(H)/\rho_c(H=0) \) for the \([110]\) field
direction exhibits steep increase with increasing field strength,
direction exhibits steep increase with increasing field strength,
reaching up to 350 at 2.5 K. \( \Delta \rho_c(H)/\rho_c(H=0) \) for the
reaching up to 350 at 2.5 K. \( \Delta \rho_c(H)/\rho_c(H=0) \) for the
\([110]\) field direction also increases, but it is an order of
\([110]\) field direction also increases, but it is an order of
magnitude smaller than that for the field along \([110]\). We
magnitude smaller than that for the field along \([110]\). We
note that even at 300 K the magnetoresistance amounts to
note that even at 300 K the magnetoresistance amounts to
6% in 9 T and shows the isotropic in-plane field depend-
6% in 9 T and shows the isotropic in-plane field depend-
dence [Fig. 2(b)]. The samples used for Figs. 2(a) and 2(b)
dependence [Fig. 2(b)]. The samples used for Figs. 2(a) and 2(b)
dependence [Fig. 2(b)]. The samples used for Figs. 2(a) and 2(b)
are different, but we confirmed that both samples show
different, but we confirmed that both samples show
effectively the same behavior. At lower in-plane fields,
effectively the same behavior. At lower in-plane fields,
at 2.5 K, \( \rho_c \) exhibits isotropic field response. It follows
at 2.5 K, \( \rho_c \) exhibits isotropic field response. It follows
the \( H^2 \) dependence expected from the orbital motion of
the \( H^2 \) dependence expected from the orbital motion of

FIG. 1 (color online). Temperature dependence of the interlayer
FIG. 1 (color online). Temperature dependence of the interlayer
resistivity \( \rho_c \) for several magnetic fields along the (a) \([110]\)
resistivity \( \rho_c \) for several magnetic fields along the (a) \([110]\)
and (b) \([110]\) directions. The strong enhancement with a nonmetallic
and (b) \([110]\) directions. The strong enhancement with a nonmetallic
upturn was observed for \( H \parallel [110] \). In contrast, the enhancement
upturn was observed for \( H \parallel [110] \). In contrast, the enhancement
for \( H \parallel [110] \) is an order of magnitude smaller. The left-hand inset
for \( H \parallel [110] \) is an order of magnitude smaller. The left-hand inset
of (b) is a schematic drawing of the FS with its relation to the crystal
of (b) is a schematic drawing of the FS with its relation to the crystal
axes and reciprocal lattice vectors. The right-hand inset shows
axes and reciprocal lattice vectors. The right-hand inset shows
the wire arrangement for the measurements of \( \rho_c \). (c) In-plane field
the wire arrangement for the measurements of \( \rho_c \). (c) In-plane field
anisotropy ratio \( \delta = \rho_c(H \parallel [110])/\rho_c(H \parallel [110]) \) for several
anisotropy ratio \( \delta = \rho_c(H \parallel [110])/\rho_c(H \parallel [110]) \) for several
field strengths. (d) Temperature derivative of resistivities for field
field strengths. (d) Temperature derivative of resistivities for field
along the \([110]\) and \([110]\) directions at 5 and 14 T.
along the \([110]\) and \([110]\) directions at 5 and 14 T.

FIG. 2 (color online). Field dependence of the magnetoresistance
FIG. 2 (color online). Field dependence of the magnetoresistance
\( \Delta \rho_c/H(=0) \) at (a) 2.5 K and (b) 300 K. The magnetoresistance at low temperature is much greater for field along
\( \Delta \rho_c/H(=0) \) at (a) 2.5 K and (b) 300 K. The magnetoresistance at low temperature is much greater for field along
the \([110]\) direction. At 300 K, the magnetoresistance is nearly
the \([110]\) direction. At 300 K, the magnetoresistance is nearly
independent of the in-plane field direction and exhibits an \( H^2 \)
independent of the in-plane field direction and exhibits an \( H^2 \)
dependence as indicated with the dotted curves.

dependence as indicated with the dotted curves.
conduction electrons [28]. However, in fields above 0.5 T, a clear anisotropy emerges with different field dependence: $\rho_c(H \parallel [110])$ varies as $H^{1.5}$, while $\rho_c(H \parallel [100])$ is proportional to $H^{0.5}$. This fact implies that $\omega_c\tau$, where $\omega_c$ is the cyclotron frequency and $\tau$ is the average relaxation time of charge carriers, becomes large in fields above 0.5 T (at 2.5 K), and the field response of the carrier mobility goes into an intermediate field region. In such a region, several orbital motions may contribute to the field dependence of the resistivity, leading to the super- or sublinear field dependence [28]. As another aspect of the field dependence, it is known that such a $H^{1.5}$ dependence can originate from the out-of-plane incoherent transport [29], in which a large number of in-plane scatterings of conduction electrons occur before electrons hop or tunnel to a neighboring plane.

Figure 3 is the so-called Kohler plot, in which $\Delta\rho_c(H)/\rho_c(H=0)$ is plotted against $\mu_0H/\rho_c(H=0)$. The universality among different temperatures is satisfied for each field direction; this indicates that the scattering process is well explained by a single relaxation rate $\tau$ and the dominant scattering process is not changed by field and temperature [28].

Figure 4(a) represents the in-plane field-angle dependence of $\rho_c$ at 2 K for several field strengths. The periodicity of $\rho_c$ is essentially 60° at and below 14 T. The observed asymmetry is attributed to a small misalignment of the magnetic field with respect to the $ab$ plane. In fact, we have confirmed that $\rho_c$ for high fields dramatically changes even with a few-degree misalignment of the magnetic field away from the $ab$ plane [30]. As we will explain later, the misalignment effect is also reproduced by the calculation [Fig. 4(b)]. The observed $\rho_c$ oscillation is consistent with the field-strength dependence: $\rho_c$ exhibits minima for $H \parallel [110]$ and maxima for $H \parallel [100]$.

In order to resolve the origin of the anomalously large MR effect in a simple metal, we have calculated the MR of PdCoO$_2$ by solving the semiclassical Boltzmann equation:

$$\begin{align*}
\sigma_{ij}(B) &= \frac{2e^2}{V} \sum_k \left( -\frac{d\phi_0(\varepsilon)}{d\varepsilon} \right) v_i(k(0)) \int_{-\infty}^{0} v_j(k(t)) e^{t/\tau} dt,
\end{align*}
$$

where $\sigma$ is the conductivity tensor, $\varepsilon$ is the elementary charge, $V$ is the volume of the sample, $\phi_0(\varepsilon)$ is the Fermi distribution function at $T = 0$, $v_i$ is the Fermi velocity along the direction $i = x, y, z$, and $\tau$ is the relaxation time. Then the resistivity tensor $\rho$ is obtained as the inverse of the conductivity tensor: $\rho = \sigma^{-1}$. Note that the assumption of a single relaxation time is justified by the fact that Kohler’s rule is satisfied in the present field range. The orbital motion of conduction electrons in the magnetic field causes the time evolution of $k(t)$, which is expressed as

$$\begin{align*}
\frac{dk(t)}{dt} &= -\frac{e}{\hbar} \mathbf{v}_f \times B = -\frac{e}{\hbar^2} \nabla_k e \times B.
\end{align*}
$$

The solution (1) is numerically calculated with $N \times N \times N$ ($N = 2^3$ or $2^6$) meshes in the reciprocal space. This process is essentially identical to that used for the quasi-one-dimensional conductor (TMTSF)$_2$X in Ref. [31].

We attempted to calculate the MR using a model dispersion relation $\varepsilon(k)$ that approximately reproduces the Fermi surface obtained from the first-principles calculations:
\(\varepsilon(k) = -2t_1 \{ \cos(k \cdot a) + \cos(k \cdot b) + \cos[-k \cdot (a + b)] \}
- 2t_2 \cos(k \cdot c) - 2t_3 \cos^2(k \cdot a) + \cos^2(k \cdot b)
+ \cos^2[-k \cdot (a + b)]\),

where \(t_1\) are tight-binding-like phenomenological hopping energies, \(a\), \(b\), and \(c\) are crystalline unit vectors of the hexagonal representation of the \(R\bar{3}m\) structure of \(\text{PdCoO}_2\). We chose \(t_1 \sim 1.0\) eV, \(t_2 \sim 0.01\) eV, and \(t_3 \sim 0.14\) eV, so that the resulting FS matches that obtained from the first-principles band calculation [32] as shown in the inset of Fig. 4(b). Note that \(t_1\) is the in-plane nearest-neighbor (i.e., Pd-Pd) hopping, \(t_2\) is the interlayer hopping energy for the \(c\)-axis unit cell length (i.e., three Pd layers), and \(t_3\) is the in-plane third-nearest-neighbor (i.e., Pd-Pd-Pd) hopping. Thus, \(t_1\), \(t_2\), and \(t_3\) should have approximate relations to the in-plane nearest-neighbor hopping \(t_{\text{NNN}}\), the in-plane next-nearest neighbor hopping \(t_{\text{NNNN}}\), and the \(c\)-axis nearest neighbor hopping \(t_{\text{zzz}}\) used in the recent de Haas–van Alphen study [20] as \(t_1 = t_{\text{NN}}\), \(t_2 \sim t_{\text{zz}}/3\), and \(t_3 \sim -\sqrt{3}t_{\text{NNN}}/2\). This is indeed the case since \(t_{\text{NN}} = 1\) eV, \(t_{\text{NNN}} = -0.23\) eV, and \(t_{\text{zz}} = 0.042\) eV is deduced from the de Haas–van Alphen results [33].

We comment here that effects of the spin-orbit interaction to the MR are probably negligible. This is because the spin-orbit interaction band splitting around the Fermi level is negligibly small although the coupling constant of the spin-orbit interaction band splitting around the Fermi level reduces the MR [see Fig. 4(a)]. The misalignment effect of the field out of the conducting plane: even a tiny misalignment of the field out of the conducting plane reduces the MR [see Fig. 4(a)]. The misalignment effect is also reproduced by the calculation, as shown with the broken curve in Fig. 4(b). According to the calculation, even \(3^\circ\) field misalignment results in a reduction of MR by 75%.

The calculation reveals that a long \(\tau\) is essential for the MR of \(\text{PdCoO}_2\). Indeed, it has been pointed out that the scattering rate \((\sim 1/\tau)\) of \(\text{PdCoO}_2\) is surprisingly small [20]. We can estimate the actual \(\tau\) using the observed in-plane mean free path \(l = (v_{F}^2)^{1/2} \sim 20\) \(\mu\)m from Refs. [20,33], and the mean Fermi velocity \(v_F\) is maximized, resulting in the large MR enhancement. Similar cases have been found in quasi-1D or -2D organic conductors [31,34,35]. In addition, closed orbits due to the FS warping along the \(k_c\) direction may also contribute to the MR. Indeed, the MR for fields nearly parallel to the [110] direction is quite sensitive to the field alignment along the conducting \(ab\) plane: even a tiny misalignment of the field out of the conducting plane reduces the MR [see Fig. 4(a)]. The misalignment effect is also reproduced by the calculation, as shown with the broken curve in Fig. 4(b). According to the calculation, even \(3^\circ\) field misalignment results in a reduction of MR by 75%.

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To summarize, we have discovered the extremely large magnetoresistance reaching \(\Delta \rho(H = 14\ T) / \rho(H = 0\ T) = 35\ 000\%\) in the quasi-two-dimensional, nonmagnetic metal \(\text{PdCoO}_2\). This MR is surprising since its electronic structure is very simple. Based on the semiclassical calculation, we demonstrate that the observed MR is closely related to the Lorentz-force-driven orbital motion of the high-mobility charge carriers. The present finding marks \(\text{PdCoO}_2\) as the first single-band simple metal exhibiting extremely large MR and furthermore opens a route to apply this oxide to industrial devices such as magnetic sensors.
We would like to thank N. Hussey, X. Xiaofeng, M. Kriener, S. Kittaka, C. Michioka, and K. Yoshimura for experimental advice and supports. We also acknowledge A. Mackenzie, C. Hicks, K. Ishida, Y. Ihara, Y. Nakai, H. Kadawaki, and R. Higashinaka for useful discussions. This work was supported by the MEXT Grants-in-Aid for Scientific Research 21340100, for Research Activity Start-up 22840036, for Young Scientists (B) 24740240, and for the Global COE program “The Next Generation of Physics, Spun from Universality and Emergence.”

Note added in proof.—We recently became aware of the work on PtSn$_4$ [37] reporting a magnetoresistance even greater than that which we report here. The Fermi surface of PtSn$_4$ is rather complicated and the mechanism of the large MR is attributed to the compensation of mobilities of different carriers. We thank P. Canfield for directing our attention to this work.

[30] From the polar angle $\theta$ dependence of $\rho_e$ between the [001] axis and the [110] axis, the misalignment of the angle in $\rho_e(\phi)$ was roughly estimated to be $\theta \approx 3^\circ$, assuming that the azimuthal angle $\phi$ was just in the [110] direction. For this estimation, we measured another sample with the single axis rotator.
[33] Supplemental Material of Ref. [20].
[36] It is noted that in Ref. [14] $\tau$ is obtained as $\tau \approx 10^{-2}$ ps. The discrepancy between this value and our estimation ($\tau \approx 10$ ps) mainly comes from a different value of the mean free path $l(=\nu_F \tau)$; i.e., $l = 60$ Å was used in Ref. [14], while $l = 20$ µm was used in our estimation. The latter value is quoted from Refs. [20,33] because the crystals used in our studies and in Refs. [20,33] are from an identical growth batch. We thus expect that the discrepancy of $\tau$ originates in the sample quality.