Extremely Large Magnetoresistance in the Nonmagnetic Metal $PdCoO_2$

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Extremely large magnetoresistance is realized in the nonmagnetic layered metal $PdCoO_2$. 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 $[1\bar{1}0]$ 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 $PdCoO_2$. 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 $PdCoO_2$ has a layered hexagonal structure with the space group $R\bar{3}m$ 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 [11,13,15–17,20]. The electronic specific-heat coefficient is $\gamma \simeq 1\,\mathrm{mJ/molK^2}$, indicating that electron correlation in $PdCoO_2$ is not strong. The carrier density n estimated from the Hall coefficient is $n_{\mathrm{obs}} = 1.6 \times 10^{22}\ \mathrm{cm}^{-3}$, 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 $3d^6$ low-spin state of Co^{3+} with zero total spin, $PdCoO_2$ 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 ρ_c is surprisingly enhanced by the application of the in-plane magnetic field along the $[1\bar{1}0]$ 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 ρ_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 $PdCoO_2$ 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 $PdCoO_2$ 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 ρ_c was measured on single-domain crystals with the hexagonal plate shape having an area of about $5\,\mathrm{mm}^2$ 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 ($\phi = 25\,\mu\mathrm{m}$) 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 electrodes, as shown in the inset of Fig. 1(b). This wire arrangement is justified for anisotropic metals, and is indeed often used for layered compounds [24,25]. The residual resistivity ratio, $\rho_c(300\ \mathrm{K})/\rho_c(2\ \mathrm{K})$, 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 the possibility of a magnetic phase transition induced by a magnetic field, the specific heat (C_P) and dc magnetic susceptibility (M/H) were measured with a commercial calorimeter and with a SQUID magnetometer on a group of aligned crystals.

Figure 1 presents the temperature dependence of ρ_c for fields along the $[1\bar{1}0]$ and [110] directions. With magnetic fields in the ab plane, ρ_c exhibits substantial enhancement on cooling. In particular, for fields along the $[1\bar{1}0]$ direction, ρ_c even turns into nonmetallic, exhibiting huge enhancement. It reaches 350 times larger than the zero-field resistivity value at 2 K in 14 T. This enhancement is comparable to those of giant magnetoresistance and colossal magnetoresistance materials [3,4]. However, in sharp contrast with these materials, $PdCoO_2$ exhibits a positive field response. Moreover, as confirmed by in-field measurements of C_P and M/H, we did not observe any anomalies suggesting magnetic or charge order at temperatures where the enhancement of ρ_c develops.

When rotating the field from the $[1\bar{1}0]$ to [110] directions, ρ_c sharply drops and switches to metallic, although it slightly deviates from a typical T-linear dependence in zero field. A small hump structure exists at temperatures where $\rho_c(H \parallel [1\bar{1}0])$ starts to increase. As is clear from Figs. 1(c) and 1(d), the in-plane field anisotropy emerges at a temperature which depends on the field strength. In fact, $d\rho_c/dT$ for $H \parallel [1\bar{1}0]$ and $H \parallel [110]$ starts to deviate from each other at about 100 K in 5 T, and about 180 K in 14 T. The anisotropy ratio, $\delta \equiv \rho_c(H \parallel [1\bar{1}0])/\rho_c(H \parallel [110])$, is up to 29 (2900%) at 2 K in 14 T. This value is larger than the field anisotropy of ordinary nonmagnetic metals with simple sphere FSs, such as alkali metals ($< 1\%$) [26]. It is also noteworthy that this anisotropy value is even larger than a typical ratio of the anisotropic magnetoresistance of magnetic metals ($< 2\%$–3%) [27].

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

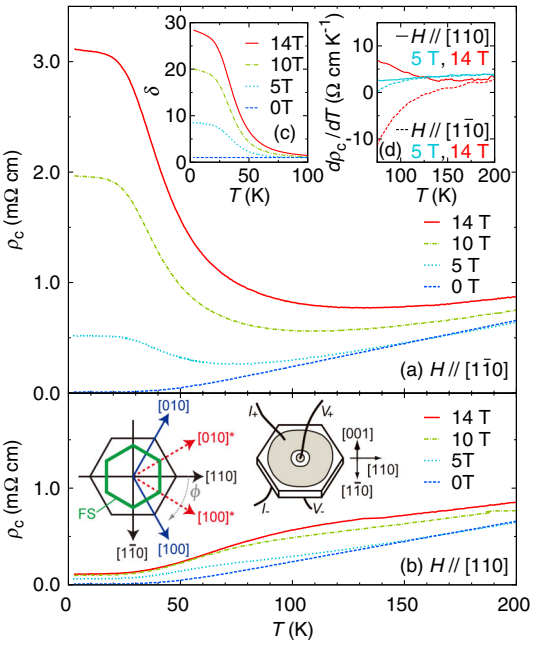

FIG. 1 (color online). Temperature dependence of the interlayer resistivity ρ_c for several magnetic fields along the (a) $[1\bar{1}0]$ and (b) [110] directions. The strong enhancement with a nonmetallic upturn was observed for $H \parallel [1\bar{1}0]$. In contrast, the enhancement for $H \parallel [110]$ 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 axes and reciprocal lattice vectors. The right-hand inset shows the wire arrangement for the measurements of ρ_c. (c) In-plane field anisotropy ratio $\delta = \rho_c(H \parallel [1\bar{1}0])/\rho_c(H \parallel [110])$ for several field strengths. (d) Temperature derivative of resistivities for field along the $[1\bar{1}0]$ and [110] directions at 5 and 14 T.

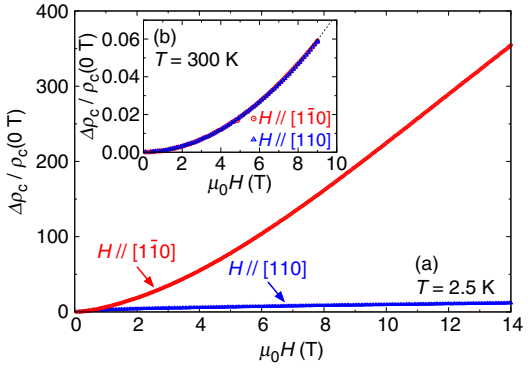

FIG. 2 (color online). Field dependence of the magnetoresistance $\Delta\rho_c/\rho_c(H=0)$ at (a) 2.5 K and (b) 300 K. The magnetoresistance at low temperature is much greater for field along the $[1\bar{1}0]$ direction. At 300 K, the magnetoresistance is nearly independent of the in-plane field direction and exhibits an H^2 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: $\rho_c(H \parallel [1\bar{1}0])$ varies as $H^{1.5}$, while $\rho_c(H \parallel [110])$ is proportional to $H^{0.5}$. This fact implies that $\omega_c\tau$, where ω_c is the cyclotron frequency and τ 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 $\mu_0 H/\rho_c(H=0)$. 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 τ and the dominant scattering process is not changed by field and temperature [28].

Figure 4(a) represents the in-plane field-angle ϕ dependence of ρ_c at 2 K for several field strengths. The periodicity of ρ_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 ρ_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 ρ_c oscillation is consistent with the field-strength dependence: ρ_c exhibits minima for $\boldsymbol{H} \parallel [110]$ and maxima for $\boldsymbol{H} \parallel [1\bar{1}0]$.

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:

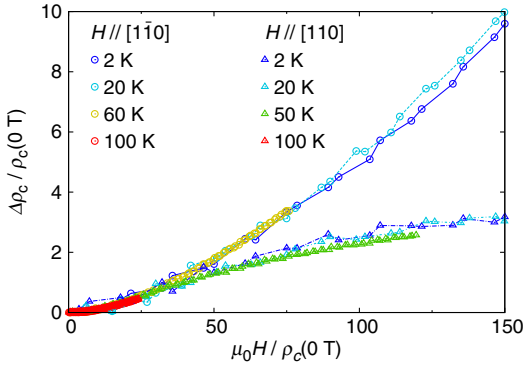

FIG. 3 (color online). The so-called Kohler plot: the magnetoresistance ratio versus field divided by the zero-field resistivity.

$$\sigma_{ij}(\boldsymbol{B}) = \frac{2e^2}{V}\sum_{\boldsymbol{k}}\left(-\frac{df_0(\varepsilon)}{d\varepsilon}\right)v_i(\boldsymbol{k}(0))\int_{-\infty}^{0} v_j(\boldsymbol{k}(t))e^{t/\tau}dt, \tag{1}$$

where σ is the conductivity tensor, e is the elementary charge, V is the volume of the sample, $f_0(\varepsilon)$ is the Fermi distribution function at $T=0$, v_i is the Fermi velocity along the direction $i = x, y, z$, and τ is the relaxation time. Then the resistivity tensor ρ 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 $\boldsymbol{k}(t)$, which is expressed as

$$\frac{d\boldsymbol{k}(t)}{dt} = -\frac{e}{\hbar}\boldsymbol{v}_{\mathrm{F}} \times \boldsymbol{B} = -\frac{e}{\hbar^2}\nabla_{\boldsymbol{k}}\varepsilon \times \boldsymbol{B}. \tag{2}$$

The solution (1) is numerically calculated with $N \times N \times N$ ($N = 2^5$ or 2^6) meshes in the reciprocal space. This process is essentially identical to that used for the quasi-one-dimensional conductor $(TMTSF)_2X$ in Ref. [31].

We attempted to calculate the MR using a model dispersion relation $\varepsilon(\boldsymbol{k})$ that approximately reproduces the Fermi surface obtained from the first-principles calculations:

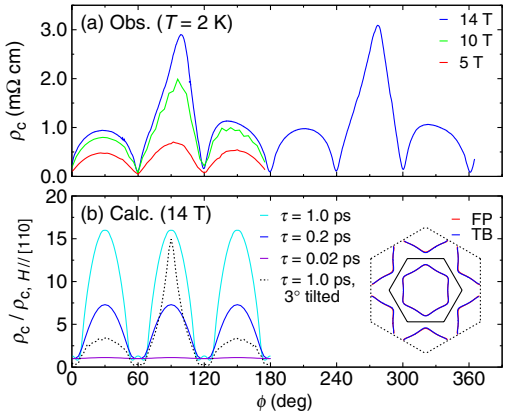

FIG. 4 (color online). (a) Observed azimuthal field angle ϕ dependence of the interlayer resistivity ρ_c of $PdCoO_2$ at 2 K, for $\mu_0 H = 5$, 10, and 14 T. (b) Calculated ϕ dependence of ρ_c at $\mu_0 H = 14$ T and $\tau = 1.0$, 0.2, and 0.02 ps, normalized by ρ_c for $H \parallel [110]$. We also present ρ_c at 1.0 ps for fields slightly tilted from the ab plane with the broken curve. For this calculation, the field-rotation plane is assumed to be tilted by 3° from the $[1\bar{1}0]$ direction in the ab plane to the [001] direction. The inset of (b) presents the Fermi surface cross section at $k_z = 0$ for the first-principles (FP) band calculation and the tight-binding (TB) model. The solid hexagon indicates the first BZ for the hexagonal representation and the broken hexagon the first BZ for the rhombohedral representation. The latter is shown to present a larger portion of the FS.

$$\varepsilon(\boldsymbol{k}) = -2t_1\{\cos(\boldsymbol{k}\cdot\boldsymbol{a}) + \cos(\boldsymbol{k}\cdot\boldsymbol{b}) + \cos[-\boldsymbol{k}\cdot(\boldsymbol{a}+\boldsymbol{b})]\} - 2t_2\cos(\boldsymbol{k}\cdot\boldsymbol{c}) - 2t_3\{\cos^2(\boldsymbol{k}\cdot\boldsymbol{a}) + \cos^2(\boldsymbol{k}\cdot\boldsymbol{b}) + \cos^2[-\boldsymbol{k}\cdot(\boldsymbol{a}+\boldsymbol{b})]\},$$

where t_i are tight-binding-like phenomenological hopping energies, $\boldsymbol{a}$, $\boldsymbol{b}$, and $\boldsymbol{c}$ are crystalline unit vectors of the hexagonal representation of the $R\bar{3}m$ structure of $PdCoO_2$. 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_{NN}, the in-plane next-nearest neighbor hopping t_{NNN}, and the c-axis nearest neighbor hopping t_{zz} used in the recent de Haas–van Alphen study [20] as $t_1 = t_{NN}$, $t_2 \sim t_{zz}/3$, and $t_3 \sim -\sqrt{3}t_{NNN}/2$. This is indeed the case since $t_{NN} = 1$ eV, $t_{NNN} = -0.23$ eV, and $t_{zz} = 0.042$ 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, ζ, is relatively large at the Pd site ($\zeta \sim 0.22$ eV) from our band calculation. The absence of the band splitting around the Fermi level is attributed to the fact that the FS consists of one band of Pd d electrons, being well separated in energy from the other bands [32].

Figure 4(b) represents the calculated $\rho_c = \rho_{zz}$ for $\mu_0 H = 14$ T and $\tau = 1.0, 0.2$, and 0.02 ps. The calculated qualitatively reproduces the observed behavior: i.e., minima in ρ_{zz} for $\boldsymbol{H} \parallel [110]$. We should also point out that the in-plane MR anisotropy grows rapidly as τ becomes longer: $\rho(0°)/\rho(90°)$ reaches 15 for $\tau = 1$ ps. We also calculated the field dependence of $\Delta\rho_c/\rho_c(H=0)$ for $\tau = 1$ ps as plotted in Fig. 5. This calculation also captures the enhancement of ρ_c by several hundred times for $\boldsymbol{H} \parallel [1\bar{1}0]$ (i.e., $\phi = 90°$). These agreements strongly indicate that the observed large MR is closely related to the band structure near the Fermi energy and essentially ascribable to the orbital motions of conduction electrons. In particular, the in-plane hexagonal anisotropy of the FS (i.e., the t_3 term in the dispersion or the k_{31} and k_{60} terms in Ref. [20]) plays an important role in the anisotropy in the MR: For $\boldsymbol{H} \parallel [110]$, two planes of the FS are nearly perpendicular to the magnetic field. Since $\boldsymbol{v}_F \propto -\nabla_{\boldsymbol{k}}\varepsilon(\boldsymbol{k}_F)$ is perpendicular to the FS, the Lorentz force $\boldsymbol{F}_L \propto \boldsymbol{v}_F \times \boldsymbol{B}$ is nearly zero for carriers on this part of the FS. Thus, roughly speaking, $2/6$ of the carriers do not contribute to the enhancement of the MR. In contrast, for $\boldsymbol{H} \parallel [1\bar{1}0]$, almost all conduction electrons on the FS feel the Lorentz force and contribute to the MR enhancement. In particular, for the carriers on the two FS planes parallel to the magnetic field, $|\boldsymbol{F}_L|$ 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_z direction may also contribute to the MR. Indeed, the MR for fields nearly parallel to the $[1\bar{1}0]$ 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° field misalignment results in a reduction of MR by 75%.

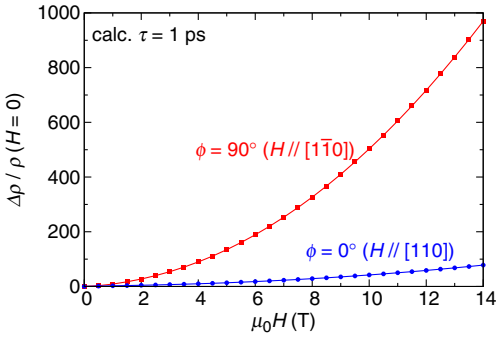

FIG. 5 (color online). Calculated field dependence of the out-of-plane magnetoresistance $\Delta\rho_c/\rho_c(H=0)$ for $\tau = 1$ ps. The solid lines just connect the discrete calculated data points. The observed large anisotropy in $\Delta\rho_c/\rho_c(H=0)$ is well reproduced.

The calculation reveals that a long τ is essential for the MR of $PdCoO_2$. Indeed, it has been pointed out that the scattering rate ($\sim 1/\tau$) of $PdCoO_2$ is surprisingly small [20]. We can estimate the actual τ using the observed in-plane mean free path $l = \langle \boldsymbol{v}_F^2 \rangle^{1/2}\tau \sim 20$ μm from Refs. [20,33], and the mean Fermi velocity $\langle \boldsymbol{v}_F^2 \rangle^{1/2} \sim 1.5 \times 10^6$ m/s from our calculation: we obtain $\tau \sim 10$ ps, which is even larger than our calculation limit [36]. Thus, the unusual suppression of scattering is the key ingredient of the observed large MR. Note that the extremely large MR in a simple metal has been overlooked for many years. The reason is probably that a layered material with such a high mobility has been quite rare so far.

To summarize, we have discovered the extremely large magnetoresistance reaching $\Delta\rho(H = 14\text{ T})/\rho(H = 0\text{ T}) = 35\,000\%$ in the quasi-two-dimensional, nonmagnetic metal $PdCoO_2$. 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 $PdCoO_2$ 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.

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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.

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