Possible co-existence of local itinerancy and global localization in a quasi-one-dimensional conductor

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In the chain compound PrBa$_2$Cu$_4$O$_8$ localization appears simultaneously with a dimensional crossover in the electronic ground state when the scattering rate in the chains exceeds the hopping rate between the chains. Here we report the discovery of a large, transverse magnetoresistance (T-MR) in PrBa$_2$Cu$_4$O$_8$ in the localized regime. This result suggests a novel form of localization whereby electrons retain their metallic (quasi-one-dimensional) character over a microscopic length scale despite the fact that macroscopically, they exhibit localized (one-dimensional) behavior.

The physics of purely one-dimensional (1D) conducting chains is a rich and fascinating area of solid state research due primarily to the emergence of exotic ground states and the availability of exact solutions [1,2]. Once coupling is introduced between chains however, as in real electronic systems, our understanding is far from complete, especially when disorder and/or interactions are considered. While it is established that all states are localized in strictly 1D systems containing arbitrarily weak disorder [3,4,5,6], in quasi-1D conductors, the onset of localization [7,8] and the transition to the Tomonaga-Luttinger liquid (TLL) state [9,10] remain theoretical controversies, prompting the search for chain-containing compounds whose effective dimensionality can be controlled by tuning the interchain coupling.

PrBa$_2$Cu$_4$O$_8$ (Pr124), with its network of double CuO layers sandwiched between insulating CuO$_2$ bilayers (see Fig. 1), is one such system. In clean Pr124 crystals, a highly anisotropic 3D Fermi-liquid state develops at low temperatures with $\rho(T) \sim T^2$ along all three crystallographic axes ($\rho_a, \rho_b, \rho_c \sim 1000-13000 \Omega\cdot m$) [11,12]. Due to the small interchain hopping energies ($2t_{a,c} \sim 3-5$meV), coherent interchain tunneling is extremely fragile to external perturbations and dimensional crossover phenomena have been observed both as a function of temperature [11] and magnetic field $H$ [12]. It was also shown recently that when the residual in-chain resistivity $\rho_0$ exceeds a certain threshold value, $\rho_b(T)$ develops an upturn at low $T$ [13]. This upturn occurs in a regime where the interchain elastic scattering rate $h/\tau_0 > 2t_{a,c}$, suggesting a close correlation between interchain coherence and localization of in-chain states. The possibility to vary both dimensionality and the nature of the metallic ground state makes Pr124 a potentially ideal system with which to resolve experimentally the controversies outlined above.

In this Letter, we probe the in-chain charge dynamics in Pr124 across the localization threshold via a detailed and systematic in-chain magnetotransport study. In the absence of any charge-density-wave (CDW) formation, a unique form of localization is implied, whereby the chain network becomes fragmented into metallic islands sandwiched between strong back-scattering impurities. In contrast to existing models [8,14,15] however, the data imply that locally, finite coherent interchain hopping survives despite the fact that globally, $h/\tau_0 > 2t_{a,c}$.

All four needle-like crystals used in this study (and in Ref. [13]) were grown by a self-flux method in MgO crucibles under high oxygen pressures [16]. Elemental analysis revealed the presence of Mg as the most abundant impurity element with a content that scaled approximately with (extrapolated) residual resistivities. The $\rho_b(T,H)$ measurements were carried out for 1.4K $\leq T < 300$K using a standard four-probe ac lock-in technique in a 14 Tesla magnet. For a detailed description of the mounting and measurement procedure, in particular the isolation of the in-chain current response and the absolute determination of $\rho_b$, please see Ref. [11,13]. Our estimate of the uncertainty in the stated values is $\pm 20\%$.

Fig. 1 shows $\rho_b(T)$ of the four crystals, labelled b1-b4.
in order of increasing resistivity at $T = 300K$. Only $b1$ remains metallic down to $T = 0.3K$. All other crystals have a resistivity minimum at $T = T_{\text{min}}$. For sample $b2$, close to the localization threshold ($T_{\text{min}} = 27K$), the nominal zero-temperature mean-free-path $\ell_0 \sim 200A$ equivalent to $k_F\ell_0 \sim 50$ ($k_F$ being the in-chain Fermi wave vector). The kinks observed in $\rho_0(T)$ in samples $b2 - b4$ are due to Néel ordering of the Pr ions at $T_N(\text{Pr}) = 17.5K$. Since Pr ordering occurs in all crystals (with or without upturns) at around the same temperature, the resistivity upturns, that appear only in more disordered crystals and at varying $T_{\text{min}} > T_N(\text{Pr})$, are clearly of a different origin.

The panels of Fig. 2 display the change in resistivity $\Delta\rho_0 = \rho_0(\mu_0H) - \rho_0(0)$ as a function of applied field for $b1$-$b4$ at $T = 20K$ [12]. Let us consider first the metallic sample $b1$: the T-MR for $H/\parallel a$ is large, positive and varies quadratically with $\mu_0H$ up to 13T. By contrast, the T-MR for $H/\parallel c$ is small, negative and similar in size to the longitudinal MR. This striking anisotropy in the MR response indicates that it is related to carriers on the CuO chains rather than the insulating planes. The marked difference in the T-MR for $H/\parallel c$ and $H/\parallel a$ is surprising though given the Fermi surface topology and the fact that the warping of the quasi-1D Fermi sheets is comparable in both interchain directions [12].

One possible explanation for this anisotropy is that the orientation of the zig-zag double chain network along the c-axis (see Fig. 1) gives rise to a larger hopping integral within the bi-chains and hence a larger orbital MR for $H/\parallel a$. This conjecture is not consistent however with the observation of a large T-MR for $I/\parallel a$ and $H/\parallel c$ [18]. The more likely origin for the T-MR anisotropy for $I/\parallel b$ is a near-perfect cancellation of the magnetoconductance $\Delta\sigma$ for $H/\parallel c$ by the Hall conductivity $\sigma_{xy}$. The general expression for $H/\parallel z$ is $\Delta\rho_{xx}/\rho_{xx}(0) = -\Delta\sigma_{xx}/\sigma_{xx}(0) - \sigma_{xy}^2/\sigma_{xx}(0)\sigma_{yy}(0)$. In our case, $x = b$ and $y = a(c)$ for $H/\parallel c(a)$. In a perfectly isotropic metal, the two terms in the sum are equivalent and correspondingly, the T-MR is zero. In an anisotropic (quasi-1D or quasi-2D) metal, such cancellation is also possible, but only for in-chain(plane) currents and isotropic mean-free-paths. The observation of finite T-MR for $H/\parallel a$ requires the condition $\sigma_{ba} > \sigma_{bc}$ and thus some additional momentum-dependence in the integral for $\sigma_{ba}$ that is absent for $\sigma_{bc}$.

The most surprising feature of Fig. 2 is the observation of similarly strong anisotropy in the MR response of the more disordered samples. In the top panel of Fig. 3 we compare directly $\Delta\rho_0(\mu_0H)$ at $T = 20K$ and $H/\parallel a$ for...
each crystal. Remarkably, $\Delta \rho_b(\mu_0 H)$ for $b1$ is more than one order of magnitude larger than for $b1$. Indeed, the T-MR is smallest in $b1$ even when $\Delta \rho_b(\mu_0 H)$ is scaled by the corresponding zero-field value $\rho_b(0)$, as shown in the inset to the top panel of Fig. 3.

Before discussing possible origins of the large T-MR in the more disordered crystals, let us first highlight two aspects of their T-MR response that differ from that of the metallic sample; namely the field dependence and Kohler’s scaling. The bottom panel of Fig. 3 shows $\partial \rho_b/\partial (\mu_0 H)$ for $b1$-$4$ at $T$=$20K$ (scaled for clarity). Below $5T$, the derivative is linear (i.e. $\Delta \rho_b \propto H^2$) for all four crystals and remains so for $b1$ at all field strengths studied. In the three other samples however, $\partial \rho_b/\partial (\mu_0 H)$ shows kinks around $5T$ and $10T$. These kinks are only observed for $H // a$ and are present above and below both $T_{\text{min}}$ and $T_N(\text{Pr})$. This peculiar field-dependence in fact mirrors that observed for $I // c$ and $H // a$ in clean crystals that remain metallic at low $T$ [12]. We reproduce one such curve (at $T = 20K$) in the inset to the bottom panel of Fig. 3 for comparison. For $I // c$, these two kinks are attributed to a field-induced renormalization of the $c$-axis warping and provide estimates for $2t_c$ on the individual Fermi sheets [12]. The increased sensitivity of the (conduction) electrons to small perturbations is probably an effect due to weaker screening of the carriers in a state of lower dimensionality (i.e. since $\hbar/\tau_0 > 2t_{a,c}$) and is also manifest in the appearance of the kink in $\rho_b(T)$ of the more disordered crystals at $T_N(\text{Pr})$ (see Fig. 1), a feature that is universally observed in $\rho_b(T)$ [11].

Kohler’s rule states that $\Delta \rho/\rho \propto (H/\rho)^2$ for $\Delta \rho(H) \propto H^2$. Thus by plotting $\Delta \rho/\rho(0)$ vs. $(H/\rho(0))^2$, all data should collapse onto a single curve independent of temperature. Such scaling, which is only observed in metallic systems, implies that the lifetime controlling the cyclotron motion is the same as that which determines $\rho(T)$ [19]. Fig. 4 shows Kohler plots for samples $b1$ and $b2$. For $b1$, Kohler’s scaling is reasonably well obeyed below $20K$ with small deviations from full scaling thereafter. In contrast, for $b2$, Kohler’s rule is violated at all $T$ (in fact the product $\Delta \rho_b.\rho_b(0)$ decreases exponentially with $T$). This difference in Kohler’s scaling between $b1$ and $b2$ is striking given the very similar $\rho_b(T)$ behavior of the two crystals (Fig. 1) and suggests that a Boltzmann-type approach with its associated relaxation time approximation is no longer valid once $\hbar/\tau_0 \geq 2t_{a,c}$. Such violation of Kohler’s rule is reminiscent of that seen in the high-$T_c$ cuprates where it has been attributed to the presence of two scattering times possibly associated with spin-charge separation, a hallmark of the TLL state [20].

The presence of such a large positive T-MR appears to rule out Kondo scattering as the origin of the low-$T$ upturn in $\rho_b(T)$ in Pr124. A positive quadratic MR may occur in localized quasi-1D compounds due to strong spin-orbit scattering (anti-localization) [21] or field-enhanced spin-flip scattering [22] but these effects tend to be isotropic and in the case of the latter, scale as $(H/T)^2$ [22], in marked contrast to what is observed here. Finally, neither the $T$-dependence of $\rho_b$ nor the $H$-dependence of $\Delta \rho_b$ are compatible with variable-range-hopping [23].

In partially-gapped CDW compounds such as NbSe$_3$ [24] and AMo$_6$O$_{17}$ ($A = K, Tl, Na$) [25], a large positive MR is observed for fields perpendicular to the principal conduction axis due to field-enhanced nesting of the Fermi sheets [26] and resultant two-band effects which lead to a violation of Kohler’s rule [23, 27]. One might consider a similar process occurring in Pr124 whereby disorder progressively pins the slowly fluctuating charge order [28]. It is well known however that disorder also tends to randomize the phases of the CDW and so disturb the coherence needed for achievement of the phase transition. Moreover this scenario is difficult to reconcile with the fact that $\Delta \rho_b(H)$ in samples $b2$-$4$ is qualitatively the same as that seen for $I // c$ and $H // a$ in crystals showing no resistivity upturn and therefore no pinned CDW [12]. In the latter, the low-field T-MR is entirely consistent with Boltzmann transport theory. We conclude therefore that whilst the electronic states in disordered Pr124 are showing clear signatures of localization, $\Delta \rho_b(H)$ has a similar orbital origin to that seen in the low-$T$ metallic state; the enhanced T-MR and violation of Kohler’s scaling in the more disordered crystals resulting from a change in dimensionality and subsequent re-balancing of the two T-MR components $\Delta \sigma$ and $\sigma_{xy}$ [20].

The essential point here is that even though samples...
implies that the (disorder-induced) dimensional crossover and induces singular (strong back-scattering) effects akin at the interrupted segments. In Pr124, (occasional) coherent defects are assumed to be perfectly insulating (in the alternating organometallic complex [14]. In the ISM however, Drude tail in the optical response of an apparently insulating organometallic complex [14, 15] which was originally developed to explain the observation of a ‘interrupted strand’ model (ISM) [14, 15] which remains long-lived and subject to an orbital Lorentz force. This picture bears certain similarities to the so-called ‘interrupted strand’ model (ISM) [14, 15] which was originally developed to explain the observation of a Drude tail in the optical response of an apparently insulating organometallic complex [14]. In the ISM however, defects are assumed to be perfectly insulating (in the strictly 1D sense) with no coherent tunneling between the interrupted segments. In Pr124, (occasional) coherent tunnelling between chains must occur for the Lorentz force to be active. The key point of our proposed model is that in the regime where $\hbar/\tau_0 \sim \ell_{a,c}$ interchain hopping still remains a relevant perturbation, and electrons can still hop coherently, despite not being the extended Bloch wavefunctions of the more metallic crystal. Crucially it seems disorder is more effective at modifying the slope of $\rho_0(T)$ than it is at disrupting the electron’s orbital motion. In this respect, the behavior resembles more that of the ‘hopschotch’ model of Phillips [30] in which carriers can tunnel to an adjacent chain via interchain scattering.

In conclusion we have uncovered an anomalously enhanced anisotropic T-MR in Pr124 crystals exhibiting localized behavior that suggests charge carriers maintain an itinerant character over microscopic length scales. This unique phenomenon occurs in Pr124 due to the low levels of disorder required to induce a dimensional crossover in the electronic system and may therefore be absent in other quasi-1D metals where hopping integrals tend to be larger. Whether aspects of TLL physics are manifest in Pr124 is still an interesting open question [31, 32], though the strong violation of Kohler’s rule in sample 52 is one possible indicator of such an exotic state.

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