Emergence of a real-space symmetry axis in the magnetoresistance of the one-dimensional conductor \( \text{Li}_{0.9}\text{Mo}_6\text{O}_{17} \)

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We report on an emerging symmetry axis in the magnetoresistance of bulk single crystals of quasi-one-dimensional \( \text{Li}_{0.9}\text{Mo}_6\text{O}_{17} \) below \( T_{\min} = 25 \text{ K} \), the temperature at which the electrical resistivity experiences a minimum. Detailed angle-dependent magnetoresistance sweeps reveal that this symmetry axis is induced by the development of a negative magnetoresistance, which is suppressed only for magnetic fields oriented along the poles of the \( \text{Mo}_6\text{O}_{17} \) octahedra that form the conducting chains. We show that this unusual negative magnetoresistance is consistent with the melting of dark excitons, composed of previously omitted orbitals within the \( t_{2g} \) manifold that order below \( T_{\min} \). The unveiled symmetry axis in directional magnetic fields not only provides evidence for the crystallization of these dark excitons but also sheds new light on the long-standing mystery of the metal-insulator transition in \( \text{Li}_{0.9}\text{Mo}_6\text{O}_{17} \).

INTRODUCTION

Magnetoresistance (MR) effects have been of immense interest in the condensed matter community in recent years, with the observation of colossal or extreme MR in topological semimetals (1–3), negative longitudinal MR in Weyl semimetals (4, 5), and linear MR in a variety of systems (2, 6–8). In this work, we report the discovery of a previously unidentified phenomenon in the magnetoresistive response of a one-dimensional (1D) metal whereby the angle dependence of the MR decouples from the crystalline axes. In essence, this emergence of an asymmetric MR can be ascribed to a crossover from an angular MR that is determined by the itinerant plane waves associated with the 1D Fermi surface aligned with the reciprocal lattice vectors, to one in which the local atomic basis becomes the new paradigm.

The material in question is the purple bronze \( \text{Li}_{0.9}\text{Mo}_6\text{O}_{17} \) (LMO) (9), a quasi-1D metal hosting a particularly robust Tomonaga-Luttinger liquid (TLL) state—the 1D analog of the Fermi liquid for an interacting electron system (10). At high temperatures, both spectroscopic (11, 12) and transport (13) measurements have provided evidence for TLL physics, including signatures of spin-charge separation. Below a certain temperature \( T_{\min} \sim 25 \text{ K} \), the resistivity passes through a minimum (14) and begins to rise sharply before, finally, superconductivity sets in below around 2 K, possibly with an unconventional pairing symmetry (15). Despite three decades of experimental effort (16–22), the underlying mechanism of this resistive upturn remains elusive: There is no accompanying structural distortion (23), no signature of magnetic order (24), disorder can be ruled out by the presence of an anisotropic superconducting phase (at the lowest temperatures) (15), while the transition to a more insulating state excludes the simple dimensional crossover scenario (25). Moreover, the lack of any clear signal of a standard Peierls phase transition [e.g., in the specific heat (16, 22)] suggests an exotic, higher-order transition that involves only a small fraction of the carriers. All this makes LMO exceptional among the family of bronzes.

Many aspects of this unusual phenomenon, described in more detail below, are found to be consistent with a new theoretical framework (26) for LMO in which dark excitons (27), formed within the \( t_{2g} \) manifold close to the Fermi level, order at \( T_{\min} \) and, through their interaction with the charge carriers, induce the upturn in the resistivity. Both the anomalous angular dependence of the MR and the peculiar two-stage character of an underlying order-by-order phase transition suggest new physics involving multiorbital degrees of freedom.

RESULTS

LMO has a monoclinic crystal structure (\( P2_1/m \)) (16, 28), with a primitive unit cell comprising two stacks of four corner-sharing \( \text{Mo}_6\text{O}_{17} \) octahedra and two \( \text{Mo}_5\text{O}_{14} \) tetrahedra arranged along a chain (see Fig. 1A). Although it is 3D in structure, electronically (for a douplet of bands crossing the Fermi energy), it is 1D due to the very weak interchain dispersion is almost negligible. A cut along the P-K line is shown in the lower panel of Fig. 1C. Because of the strong crystal field effects, the two bands crossing the Fermi level (one for each stack) are of \( d_{xy} \) character, while the \( d_{xz} \) and \( d_{yz} \) orbitals within the same manifold are either empty or fully occupied, with a minimum gap of around 0.4 eV (highlighted in green in Fig. 1C) located midway along the P-K line. (This particular aspect of the band structure will become important

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Fig. 1. Crystal structure, band structure, and metal-insulator transition of LMO. (A) Projection of four unit cells onto the ac plane, where the MoO₆ octahedra hosting the conducting zigzag chains (oriented out of this plane along the b axis) are highlighted in purple. Blue sphere, Mo; red/pink, O; green, Li. (B) In each unit cell, there are double zigzag chains made of corner-sharing octahedra. The octahedra at the top of the figure have had all nonessential oxygen atoms removed. The corner oxygen shared by adjacent in-chain octahedra is denoted in red, while interchain oxygen is denoted in pink. (C) Simplified Fermi surface of LMO showing the weakly dispersive, quasi-1D bands along the a axis due to the weak interchain hopping energy (top panel), which, nevertheless, causes an energy gap around the Fermi surface (schematic green curves in the lower panel). The small energy gap easily allows excitation of electron-hole pairs, i.e., excitons. (D) In-chain resistivity versus temperature showing a metal-insulator transition around $T_{\text{min}} \sim 25$ K, below which the insulating form of the resistivity can be well fitted by a power law (see section S4C and fig. S11). Crystallographic drawings were produced using VESTA (44).

Later.) Last, Fig. 1D shows a typical in-chain resistivity curve $\rho_{\text{ac}}(T)$. The upturn in $\rho_{\text{ac}}(T)$ occurs around $T_{\text{min}} \sim 25 \pm 5$ K, irrespective of whether the ground state is superconducting. Above $T_{\text{min}}$ the metallic behavior is usually ascribed to TLL physics, whereas the nonmetallic state below $T_{\text{min}}$ has remained a mystery.

To gain more insight into the nature of the electronic state below $T_{\text{min}}$, we have investigated the angle-dependent MR (ADMR) in LMO for different current (I) and magnetic field (B) configurations (details of the resistivity measurements can be found in Materials and Methods). The angles of orientation of B within the three principal planes colored in Fig. 2A are specified by $\theta$, $\alpha$, and $\beta$ throughout the manuscript. Figure 2B shows the temperature evolution of the angular dependence of the a axis MR (i.e., $I//a$) as a constant field of 13 T is rotated within the ac plane. Above $T_{\text{min}} \sim 25$ K, the MR has minima and maxima wherever the field is oriented along a or c, respectively. As shown in the left-hand panel of Fig. 2C, upon reflecting the MR trace about $B//c$, the two traces are perfectly superimposed, implying a fully symmetric response. With decreasing temperature however, the positions of the maxima (and, to a lesser extent, the minima) begin to shift away from the crystallographic axes. The developing asymmetry is most illustrated in the reflection plot, shown in the right-hand panel of Fig. 2C for $T = 4.2$ K. (Corresponding reflection plots for a number of intermediate temperatures are displayed in fig. S1.)

In general, the MR of a monoclinic crystal is asymmetric, with respect to the crystallographic axes (32). However, the inclined angle in LMO is so small (~0.61°) that the system can be taken to be orthorhombic within experimental uncertainty. This agrees well with the experimentally observed mirror symmetry of MR at high temperature and with the theoretically calculated Fermi surface (30). The unexpected breaking of the mirror symmetry at low temperature, nonetheless, points to a marked change in the electronic structure that can be enhanced by strong magnetic fields.

To quantify this unusual asymmetric response, we plot in Fig. 2D the integrated area (shaded area in the right-hand panel of Fig. 2C) between the two reflections normalized to the magnitude of the MR (i.e., the difference between the resistivity values at $B//c$ and $B//a$). The inset in Fig. 2D shows a blowup of the region between 10 and 40 K. A clear onset in the asymmetry is revealed at a temperature $T_{\text{asym}} = 24 \pm 1$ K, implying that it is intimately connected to the resistivity upturn at $T_{\text{min}}$. Moreover, the growth in the asymmetry near $T_{\text{min}}$ resembles that of an order parameter. Below 10 K however, the asymmetry grows much more steeply (see the main panel of Fig. 2D for $I//a$, and fig. S2 for corresponding data for $I//b$).

This asymmetric MR is found to be independent of current direction. Figure 2E shows the evolution of the ADMR within the ac plane for $I//b$ (similar data for $I//c$ are shown in fig. S3). Above $T_{\text{min}}$, the mirror symmetry is preserved with respect to the crystallographic axes. Below $T_{\text{min}}$, the asymmetric behavior is observed at all temperatures. The magnitude of the asymmetry, however, does depend on the field strength. Fig. 2F shows the enhanced asymmetry with increasing field up to 30 T measured on a different LMO crystal at $T = 1.2$ K (see almost identical behavior in Fig. 3B for another insulating crystal and in fig. S6b for a second, superconducting crystal). As the field increases, the position of the maximum MR shifts away from the crystallographic axes. Notably, when the magnetic field is rotated within the ab or bc plane, there is no discernible shift in the position of the maxima/minima across $T_{\text{min}}$ (see also figs. S3 and S4). No shift is found either with increasing field strength (fig. S5). Such a dependence of the asymmetric MR on the specific plane of rotation places strong constraints on any effective theory put forward to explain this effect.

To gain deeper insight into how the asymmetry develops with increasing B-field, we have performed a series of MR sweeps as a function of magnetic field oriented at multiple angles within the ac plane. Fig. 3A shows results for a nonsuperconducting sample mounted with $I//a$ at $T = 1.2$ K. (Again, almost identical behavior in a superconducting sample is plotted in fig. S6). In the low-B regime ($B < 5$ T), $\Delta \rho(B)$ follows a quadratic dependence in field and scales approximately as $B \cos \theta$ ($\theta$ measured from the c axis), albeit with a small negative offset, suggesting that it originates from the effects of...
With increasing field, the MR starts to deviate from its quadratic dependence and passes through a maximum at $B_{\text{peak}} \approx 14$ T (for $B//c$), indicating the emergence of a negative MR component to $\Delta \rho$(B).

As the field is tilted away from the $c$ axis, the low-field MR decreases in magnitude. Despite this decrease however, the peak in the MR actually shifts to a higher field, leading to a cascade of crossing points as displayed in Fig. 3A. As a consequence, the angle at which $\Delta \rho$ peaks (at constant field strength) shifts progressively away from the $c$ axis with increasing field (Fig. 3B). Comparison of Fig. 3B with the ADMR data plotted in Fig. 2F confirms that the asymmetric MR in LMO arises because of the emergence of a negative component whose magnitude increases as the field is tilted away from a specific “critical angle” $\theta_{\text{CR}}$ within the $ac$ plane. One might argue that a positive component peaking around $\theta_{\text{CR}}$ can also lead to the observed asymmetry. As discussed in our previous work (17), an anisotropic positive MR must arise because of the field-induced one-dimensionalization (i.e., decreased Fermi velocity along the $b$ axis and, hence, enhanced scattering rate). Nevertheless, it is expected to preserve the mirror symmetry with respect to crystallographic axes rather than at an intermediate angle like $\theta_{\text{CR}}$. As a result, this anisotropic positive MR, although being important to shape MR curves, cannot account for the observed asymmetry (see section S5 for further discussion of the anisotropic positive MR).

In Fig. 3C, the MR data are rescaled to remove the $|\cos \theta|$ dependence (as explained in section S2 and fig. S7). In this way, all the initial slopes collapse onto a single curve, as shown in the inset. Although the field scale has been substantially renormalized, the peak value $B_{\text{peak}}$ plotted in Fig. 3D still exhibits its own maximum at a specific critical angle of $\theta_{\text{CR}} = -50^\circ \pm 2^\circ$ from the $c$ axis. At this critical angle, the influence of the negative MR is, thus, at its weakest.

Inspection of the atomic configuration within the unit cell in Fig. 4A reveals that $\theta_{\text{CR}}$ corresponds very closely (in one of its two possible orientations) to an axis that passes directly through the poles of the MoO$_6$ octahedra [i.e., the interchain direction, conventionally denoted by $[-2 0 1]$ as acknowledged previously in (23)]. From the
analysis presented in Fig. 3, we conclude that the negative MR contribution is minimized when the field is oriented along this specific polar axis, implying that the MR response of this bulk 3D crystal is governed by the real-space alignment of the atoms within the crystal lattice. Such a deviation in the symmetry of the MR from the crystal main axes may take place, for instance, in materials that are susceptible to a charge density wave (CDW) instability that is not necessarily aligned with the crystal main axis. In this case however, the asymmetry can be deduced almost entirely from the nesting conditions defined by the Fermi surface (33). Another example is a material with strong (e.g., Hubbard type) localization where, because electrons or holes spend most of their lifetime in the local atomic orbitals, it makes sense that the characteristics of local orbitals in real space become dominant. The uniqueness of LMO thus stems from the fact that in this system, which is nonetheless described well by itinerant electron physics, a signal is detected that can be ascribed directly to the local system of coordinates of atomic orbitals. Below, we introduce a model that can qualitatively explain this unprecedented effect.

**DISCUSSION**

In previous theoretical treatments of the TLL physics of LMO, only the pure $d_{xy}$ orbitals were considered as a key starting ingredient (an assumption justified by the fact that only these cross $E_F$) (30, 34). Recently however, other orbital degrees of freedom, in particular the interorbital interactions of the $d_{xz}$ and $d_{yz}$ orbitals, were incorporated into the many-body formalism and shown to affect the low-energy physics (26) (further details of the model are expounded in section S4). In particular, the specific arrangement of the $d_{xz}$-derived bands provides favorable conditions for the formation of excitons along the P–K line (highlighted in green in the lower panel of Fig. 1C). This exciton resides in adjacent octahedra within the two zigzag chains. The interband exciton has a spin triplet configuration that strongly suppresses the electron-hole recombination probability through photon emission and gives the resulting dark exciton a long lifetime (and allows for spin exchange scattering with the $d_{xy}$ fermions). At high temperatures, excitons behave as free particles (with a hopping energy of the order 1 to 2 meV), while at low temperatures, the physics of these emergent particles is governed...
by a quantum order-by-disorder transition (35), which leads to a Wigner-like periodic arrangement of the excitons. The \( d_{xy} \) fermions that remained mobile in the presence of the (slightly incommensurate) Umklapp processes associated with quarter-filling now localize on this additional potential, leading to an upturn in the resistivity below \( T_{\text{min}} \).

Crucially, the angular momentum quantization axis for the dark excitons is the \( z \) axis of the local coordination system shown in Fig. 1B and indicated by the purple arrows in Fig. 4, A to C. Any component of the \( B \)-field perpendicular to this quantization axis will lead to a mixing of the \( d_{xy} \) and \( d_{yz} \) orbitals. This breaks the conservation of angular momentum, thereby admixing bright (via electron-hole recombination) and dark excitons and opening up their fast decay channel. Once the dark exciton—the essential building block of the Wigner crystal—acquires a finite lifetime, the crystal itself must melt. Ultimately, it is this melting of the excitonic order that suppresses the additional Umklapp process below \( T_{\text{min}} \), giving rise to the negative MR and restoring the metallic state in the presence of a strong magnetic field (17). Since the crystal melting is strongly influenced by the angular offset between the magnetic field vector and the quantization axis, this model provides a natural explanation for the location of \( B_{\text{peak}} \) at \( \theta = \theta_{\text{CR}} \) and the resulting asymmetric MR response; with decreasing temperature, the unique angular dependence is determined by the orientation of the \( B \)-field with respect to the quantization axis rather than to the main reciprocal axes of the lattice.

The fact that \( B_{\text{peak}} \) does not diverge at \( \theta = \theta_{\text{CR}} \) is attributed within this picture to the slight canting of the \( \text{MoO}_6 \) octahedra within the unit cell \( \theta_{\text{crt}} \approx 6^\circ \), as illustrated in fig. S8 (and section S3), which ensures that there is always a residual component of the \( B \)-field that is orthogonal to the quantization axis and, thus, the condition for perfect alignment is never satisfied. When the magnetic field is rotated within the \( ab \) or \( bc \) plane, the angle between the \( B \)-field and the quantization axis is always maximized when \( B//b \), and so is the negative MR (17, 26). In addition, \( B//a \) and \( B//c \) have a higher positive MR than \( B//b \) due to the enhanced Lorentz force as well as a propensity for field-induced one-dimensionalization whenever the \( B \)-field is perpendicular to the atomic chains (36). Thus, one expects the maxima (minima) to be located at \( B//a \) or \( B//c \) (\( B//b \)), and no asymmetry in the ADMR is expected in these rotation planes (see section S5).

According to the theoretical model outlined above, the resistivity upturn in LMO is caused by a reentrance of Umklapp interactions between the remnant mobile \( d_{xy} \) fermions and this periodic [Wigner-like (37)] arrangement of excitons (fig. S10). In one sense, the periodic arrangement of excitons helps to “complete” the Mott transition in near-commensurate LMO, i.e., the many body system uses its own multi-orbital degrees of freedom to open a correlation gap and, hence, gain energy. Recalling that the formation of excitons is favored by long-range interactions and given that a 1D metal (in contrast to a metal in higher dimensions) is unable to screen long-range interactions, we conclude that this novel localization phenomenon can only take place in a 1D (Luttinger liquid) system—like LMO.

Now we return to discuss the unusual temperature dependence of the asymmetry parameter shown in Fig. 2B. According to the theory, not one single transition, but a sequence of two transitions (38, 39), is expected. The first transition at \( T = T_{\text{min}} \) is into a partially ordered state, while the second transition (into a fully frozen lattice) is expected to take place at \( \sim T_{\text{min}}/3 \), i.e., around 8 K. Furthermore, the magnitude of the order parameter associated with the intermediate phase order should be much smaller than the lowest-temperature fully ordered state. Both features are in agreement with the data presented in Fig. 2B.

Last, we turn to discuss various other mechanisms proposed for the resistivity upturn around \( T_{\text{min}} \) in the context of our new findings. The first is the CDW, possibly electronically driven (23), a suppression of which by Zeeman splitting in magnetic fields would lead to the giant negative MR (17). This suppression is expected to lead to an isotropic negative MR, however an expectation that is incompatible with the emerging symmetry axis unveiled by more detailed angular characterization in the present study. In the case of disorder-induced localization (22), the MR is also expected to display mirror symmetry within the \( ab \) plane, in notable contrast to the mirror symmetry-breaking response of the dark excitons that naturally correlates with the emerging symmetry axis. The dark exciton theory is, thus, expected to be an essential part of any mechanism accounting for the mysterious resistivity upturn, although how superconductivity develops with itinerant residual carriers in the presence of crystalized (localized) dark excitons is yet to be explored.

Mutual interplay between localized and itinerant degrees of freedom is a key issue in strongly correlated systems such as heavy fermions (40) and Kondo insulators (41). What happens when these two types of correlated carriers coexist, however, is largely uncharted territory, with no obvious guiding principles. Our experimental result, where both real and reciprocal space manifest in a single measurement, is likely to lay a new research direction in this field. The influence of the excitonic Wigner crystal on the magnetoresistive response of metallic LMO reported here is profound and is particularly notable given that it develops without a concomitant distortion of the lattice, suggesting that it is a purely electronically driven phenomenon. It turns out that the mysterious physics at \( T_{\text{min}} \) is driven by a type of order-by-disorder phase transition that has fascinated theorists for three decades. So far, however, the focus has been on spin systems. No one really looked at excitons as hardcore bosons, and this makes our result—a realization of a correlation gap and, hence, gain energy. Recalling that the formation of excitons is favored by long-range interactions and given that a 1D metal (in contrast to a metal in higher dimensions) is unable to screen long-range interactions, we conclude that this novel localization phenomenon can only take place in a 1D (Luttinger liquid) system—like LMO.

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MATERIALS AND METHODS
Crystal growth and crystallographic characterization
High-quality single crystals of LMO were grown using a temperature gradient flux method, and their axes were identified by a Bruker...
**SUPPLEMENTARY MATERIALS**

Supplementary material for this article is available at http://advances.sciencemag.org/cgi/content/full/5/7/eaar8027/DC1

Section S1. Similar observation in a superconducting sample (fig. S6)

Section S2. Scaling factor used in Fig. 3C and fig. S6C

Section S3. Canted angle between adjacent MoO₃ octahedra

Section S4. Theory of dark excitons and its contribution to resistivity and MR along the a axis

Section S5. Decoupling of the anisotropic MR in LMO

Table S1. Location of the maxima and minima in the angular MR in LMO above Tₘ₉₉

Fig. S1. Mirror reflection of the ADMR curves at various temperatures about the c axis.

Fig. S2. The magnitude of asymmetry as a function of temperature for MR curves of current along the b axis.

Fig. S3. Contrast in the asymmetric MR response upon rotation within the three crystallographic planes.

Fig. S4. Normalized ADMR curves obtained at various temperatures as a constant magnetic field of 13 T, rotated within the bc plane.

Fig. S5. Absence of asymmetric MR (II/III) with increasing field strength by B rotated within the bc plane.

Fig. S6. Origin of the asymmetric MR and determination of the critical angle for a second, superconducting LMO crystal.

Fig. S7. Scaling factor used in Fig. 3C and fig. S6C, respectively.

Fig. S8. A close view of the canted MoO₃ octahedra in LMO.

Fig. S9. Configurations of electric contacts applied to our LMO crystals.

Fig. S10. Schematic of crystallization of dark excitons.

**REFERENCES AND NOTES**


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