Crossed Luttinger liquid hidden in a quasi-two-dimensional material

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Although the concept of the Luttinger liquid (LL) describing a one-dimensional (1D) interacting fermion system collapses at higher dimensions, it has been proposed to be relevant to enigmatic problems in condensed matter physics including the normal state of cuprate superconductors, unconventional metals and quantum criticality. Here we investigate the electronic structure of quasi-2D η-Mo₄O₁₁, a charge-density wave material, using high-resolution angle-resolved photoemission spectroscopy and ab initio calculations. We show a prototypical LL behaviour originating from the crossed quasi-1D chain arrays hidden in the quasi-2D crystal structure. Our results suggest that η-Mo₄O₁₁ materializes the crossed LL phase in its normal state, where the orthogonal orbital components substantially reduce the coupling between intersecting quasi-1D chains and therefore maintain the essential properties of the LL. Our finding not only presents a realization of a 2D LL, but also provides a new angle to understand non-Fermi liquid behaviour in other 2D and 3D quantum materials.

Non-Fermi liquids are unconventional metallic systems that cannot be described by Landau's Fermi liquid theory. This is due to the strong electron correlation, which was suggested to be the key to understanding some of the mysterious phases in condensed matter physics, such as the normal state of cuprate high-temperature superconductors, heavy-fermion materials and the normal state of itinerant-electron ferromagnets. As an example, due to the reduction of scattering phase-space and electronic screening, 1D electronic systems can harbour the LL state, which exhibits spin-charge separation and power-law scaling of physical properties.

Although the LL theory was developed to describe 1D electronic systems, it is believed to be highly relevant to many long-standing mysteries in 2D and 3D quantum materials, such as the normal state of cuprate high-temperature superconductors, unconventional metals and quantum criticality. Therefore, it is of fundamental importance to generalize the LL phenomenology to higher dimensions. Many theoretical models suggest that the essential features of an LL can be retained in specifically coupled 1D chains (Fig. 1a). This can be understood by a coupled LL model to form the sliding LL phase. Various quasi-1D materials with weak inter-chain coupling have been experimentally verified to be LL materials with a possible dimensional crossover. Such coupled quantum wires are further proposed to host fascinating properties such as non-Abelian quantum Hall states, topological phases and quantum spin liquids.

Particularly, a novel 2D LL phase can be established by weaving 1D atomic chains as proposed in the crossed sliding LL model (Fig. 1b), which is highly relevant to the stripe phase of the cuprates. However, the realization of this intuitive theoretical proposal is challenging, as it requires a delicate construction of ordered intercrossing chain arrays. Indeed, except for the pioneering effort to pattern carbon nanotubes...
Fig. 1 | Physics in arrays of 1D chains and basic properties of η-Mo4O11.
a. Schematic of a quasi-1D crystal consisting of parallel chains. b. Schematic of crossed arrays of planar parallel chains, which may host a crossed sliding LL even with inter-array interaction. c. Overall crystal structure of η-Mo4O11, showing slabs consisting of MoO6 octahedra separated by MoO4 tetrahedra. The light grey plane indicates the (100) cleavage plane. Four inequivalent Mo atoms are marked. d. Projected view of a single slab in c. Selected chains along three different directions (b, b + c, b – c) are highlighted (see also Supplementary Fig. 1). e. Laue diffraction pattern along the (100) plane. The inset shows a photograph of a typical sample with three natural crystalline directions corresponding to the b, b + c and b – c directions. f. 3D plot of the band structure measured at 30 K, using 50 eV photons. The yellow rectangle is the surface Brillouin zone.

with only one crossing\textsuperscript{12,33}, no compelling evidence of a genuinely 2D LL has been identified yet.

In this Letter, combining high-resolution angle-resolved photoemission spectroscopy (ARPES) and ab initio calculations, we systematically investigate the electronic structure of a quasi-2D molybdenum oxide, η-Mo4O11. Remarkably, we discover an LL phase that is manifested by a robust power-law scaling behaviour of ARPES spectra near the Fermi energy (E_F), which is independent of the temperature and momentum direction. By constructing an effective tight-binding model, the low-energy electronic structure can be reproduced using three chain arrays featuring orthogonal Mo 4d t_{2g} orbitals, revealing its nature of crossed 1D electronic structure hidden in the quasi-2D crystal structure, in excellent agreement with our experiment. The inter-array coupling in η-Mo4O11 is weak due to the orthogonality between the orbital components of different chain arrays, which naturally materializes the geometry of the crossed sliding LL model. Thus, this system not only demonstrates a novel LL phase in a quasi-2D material but will also provide a new angle to understand non-Fermi liquid behaviour in other 2D and 3D quantum materials.

η-Mo4O11 crystallizes in a monoclinic structure with space group P2_1/c. As shown in Fig. 1c, it exhibits a quasi-2D structure consisting of MoO6 slabs separated by MoO4 tetrahedra\textsuperscript{34}. The slabs are composed of corner-sharing MoO6 octahedra, forming chain-like structures along three different directions, as highlighted in Fig. 1d (Supplementary Fig. 1) and shown by the three natural crystalline directions of the sample (inset of Fig. 1e). It was found that η-Mo4O11 possesses two successive charge-density wave (CDW) transitions at T_{CDW1} = 109 K and T_{CDW2} = 30 K with partial gap opening and periodic lattice distortion\textsuperscript{35,36}. These were attributed to the hidden 1D Fermi surface (FS) nesting (see Supplementary Fig. 7 for the comparison between previous results\textsuperscript{37,38} and our data regarding the CDW transition)\textsuperscript{39,40}. Previous tight-binding calculations also suggested the possibility of nesting-induced FS reconstruction in the CDW state. This was adopted on the understanding of the quantum oscillation measurements\textsuperscript{39,40} but has not yet been observed by ARPES (ref. 39). The Laue diffraction pattern along the (100) plane (Fig. 1e) and the X-ray diffraction pattern (Supplementary Fig. 2) confirm the crystal structure and the high quality of our samples\textsuperscript{41,42}. The 3D plot of the experimental band structure in Fig. 1f shows highly dispersive bands crossing E_F and parallel straight lines on the FS, consistent with previous ARPES measurements\textsuperscript{37,43}.

Figure 2 shows the detailed electronic structure of η-Mo4O11. The measured FS (Fig. 2a) consists of three groups of straight lines running along different directions (Supplementary Fig. 3). The angle between the C1 and C2 (C2 and C3) FS sheets is 51° (78°), consistent with the natural crystalline directions (inset of Fig. 1e) and implying 1D physics hidden in the quasi-2D crystal structure\textsuperscript{39}. The different FS sheets hybridize with each other, inducing gaps at the crossing points, as shown by the zoomed-in plots and momentum distribution curves (MDCs) in Fig. 2b. In Fig. 2c, we observe two highly dispersive bands along ΓZ (C2 and C3 bands) that are nearly degenerate near E_F, while the C1 band is nearly dispersionless around −650 meV, suggesting its 1D nature. Along ΓM, the C3 band becomes nearly dispersionless, while the other two bands disperse strongly (Supplementary Fig. 4). The MDCs of the band structure along ΓZ in Fig. 2d highlight the hybridization-induced band splitting between the C2 and C3 bands. These results suggest that the low-energy electronic structure is built of coupled 1D chains. The measured electronic structures are perfectly reproduced by the surface-projected ab initio calculations of both the FS (Fig. 2e) and band dispersions (Fig. 2g), where the effect of weak inter-chain coupling is also well captured by the anti-crossings between the calculated FS sheets (Fig. 2f) and band splittings (Fig. 2h) (Supplementary Figs. 3 and 4).
Fig. 2 | Comparison between the experimental and calculated electronic structure of η-Mo₄O₁₁. a, Measured FS showing three sets of quasi-1D sheets along different directions. The yellow rectangle indicates the first surface Brillouin zone. b, Magnification of the crossings between different FS sheets as indicated by the red and purple circles in a, shown in b(i) and b(ii), respectively. The dashed grey lines are guides to the eye for FS sheets. The MDCs (black circles) along the white lines show the avoided crossings of FS sheets. The MDCs fit well to two Lorentzians (blue and orange curves) on top of constant backgrounds (green curves). c, Experimental band structure along ΓZMΓM. The yellow rectangle indicates the first surface Brillouin zone (green curves). d, Measured FS showing three sets of quasi-1D sheets along different directions. The yellow rectangle indicates the first surface Brillouin zone (green curves). e, ARPES measurement directions (dashed coloured lines) and schematic FS sheets (solid black lines) marked within the first surface Brillouin zone (yellow rectangle). f, EDCs measured along different directions as marked in e and the fits to the LL model. g, Value of α extracted from data fitting at various temperatures in the measurements along different directions. h, Value of α extracted from scaling plot in the measurements along different directions (see also Supplementary Fig. 10). In g and h, the colours of circles correspond to the colours of dashed lines in e, the dashed lines are the guides to the eye for the temperature independence of α and the purple areas highlight the scattering of α. The error bars show the confidence intervals of the data fitting under the 95% confidence level. Data in a–d were measured with 21.2 eV photons. Data in f were measured at temperatures between 120 and 135 K. Data in f–h were measured with photon energies of 21.2 eV (green, cyan, purple), 32 eV (red), 33 eV (orange, yellow), 50 eV (dark blue) and 55 eV (light blue).

Fig. 3 | LL behaviour in η-Mo₄O₁₁. a, Comparison of integrated EDCs of polycrystalline gold, η-Mo₄O₁₁ and Li₀.₉Mo₆O₁₇. b, EDCs of η-Mo₄O₁₁ measured at different temperatures and the fits to the LL model. c, Scaling plot of the EDCs in b with a scaling factor of 10⁻⁴. The dashed line plots the simulated LL spectral function with anomalous exponent α = 0.36. d, log–log plot of the EDC intensity at $E_F$ versus temperature. The black line is a linear fit to the data with α = 0.38. e, ARPES measurement directions (dashed coloured lines) and schematic FS sheets (solid black lines) marked within the first surface Brillouin zone (yellow rectangle). f, EDCs measured along different directions as marked in e and the fits to the LL model. g, Value of α extracted from data fitting at various temperatures in the measurements along different directions. h, Value of α extracted from scaling plot in the measurements along different directions (see also Supplementary Fig. 10). In g and h, the colours of circles correspond to the colours of dashed lines in e, the dashed lines are the guides to the eye for the temperature independence of α and the purple areas highlight the scattering of α. The error bars show the confidence intervals of the data fitting under the 95% confidence level. Data in a–d were measured with 21.2 eV photons. Data in f were measured at temperatures between 120 and 135 K. Data in f–h were measured with photon energies of 21.2 eV (green, cyan, purple), 32 eV (red), 33 eV (orange, yellow), 50 eV (dark blue) and 55 eV (light blue).
We note that the band dispersions, despite crossing $E_F$, exhibit strongly suppressed spectral weight near $E_F$ (Fig. 2c). This is reminiscent of the power-law spectral function of an LL taking the hidden-1D electronic structures into account. In addition, the bands disperse almost linearly near $E_F$, fulfilling the prerequisite for an LL phase (Supplementary Fig. 5). Figure 3a compares the integrated energy distribution curves (EDCs) of $\eta$-Mo$_4$O$_{11}$ with prototypical Fermi liquid and LL materials. The spectrum is clearly distinct from that of a Fermi-liquid system (for example polycrystalline gold) with a Fermi edge but similar to that of the prototypical quasi-1D LL material Li$_{0.9}$Mo$_6$O$_{17}$ (ref. 25) (Fig. 3a). The analyses suggest an LL nature of $\eta$-Mo$_4$O$_{11}$ residing in its quasi-2D crystal structure.

For a quantitative description of the LL behaviour, we fit the experimental data to the finite-temperature LL model:

$$
\rho(\epsilon, T) \propto T^\alpha \text{Re} \left[ (2\pi)^{d+1} B \left( \frac{d-1+\alpha/2}{2}, -\alpha \right) \right], \tag{1}
$$

where $\epsilon = (E - E_F)/k_B T$ is the scaled energy with $k_B$ the Boltzmann constant, $B$ is the beta function and $\alpha$ represents the LL anomalous exponent. The analyses suggest $\alpha = 0$ for gold as expected for a Fermi liquid and $\alpha = 0.69$ for Li$_{0.9}$Mo$_6$O$_{17}$, the same as in a previous report (Fig. 3a). Regarding $\eta$-Mo$_4$O$_{11}$, the ARPES spectra in a wide temperature range (Supplementary Figs. 6 and 7) fit perfectly to the LL model with a temperature independent $\alpha = 0.4 \pm 0.1$ (Fig. 3b and Supplementary Fig. 8). In addition, as indicated by equation (1), the LL nature can be independently demonstrated by the scaling behaviour with temperature. That is, the ARPPES intensity scaled by $T^\alpha$ is temperature independent, which is established excellently in Fig. 3c with $\alpha = 0.36$ (Supplementary Fig. 9). Moreover, the LL model suggests a linear dependence of $\log(I(E_F))/\log(T)$ (I is the intensity) with the slope $\alpha$, as plotted in Fig. 3d, from which we extract $\alpha = 0.38$, consistent with that obtained from the model fitting and scaling plot. We note that the LL analysis was attempted in a previous ARPES measurement with a slightly different exponent of 0.2, which could result from the different experimental conditions. Compared to the previous work, our findings compellingly prove the LL nature of $\eta$-Mo$_4$O$_{11}$ residing in its quasi-2D crystal structure.

To reveal the anisotropy of the unexpected LL behaviour in the quasi-2D material, we measure ARPES spectra along different momentum directions, as indicated by the coloured dashed lines in Fig. 3e. The integrated EDCs along different directions show the same line shape and conform exactly to the LL model in all the measurements (Fig. 3f). Moreover, the spectra measured above the CDW transition temperature in different measurements can be perfectly scaled to a single curve, despite the distinct experimental conditions (Supplementary Fig. 10). Figure 3g,h summarizes the $\alpha$ values extracted independently from the model fitting and scaling plot, respectively. Despite the high resolution of our experiment, we cannot reliably extract the CDW transition due to the lack of a well-defined Fermi edge in the LL system, unlike in previous work. As a result of the minor impact of CDW transitions on our data (Supplementary Fig. 6), the spectra can be well fitted and scaled at temperatures from the normal state down to 10 K. Within experimental error, $\alpha$ is independent of temperature and measurement direction, suggesting an in-plane isotropic LL behaviour in the system.
To understand the novel LL behaviour established above, which is the focus of this work, we further investigate the electronic structure of \(\eta\)-Mo\(_4\)O\(_{11}\) by combining ab initio calculations and Wannier orbital construction in Fig. 4. The orbital-projected calculations demonstrate that the low-energy bands consist almost exclusively of 4d orbitals of inner Mo atoms in the Mo\(_2\)O\(_3\) slabs, that is, Mo III and Mo IV in Fig. 1c (Supplementary Figs. 11 and 12). Specifically, the C1 and C2/C3 bands consist mainly of the Mo\(_d_x^0\) and \(d_y^0/d_z^0\) orbitals, respectively (Fig. 4a).

For further exploration of the nature of these bands, a low-energy effective model based on Wannier orbitals is constructed, where the six bands that cross \(E_F\) can be accurately described by the tight-binding model with six Wannier orbitals in a unit cell (Fig. 4a). The resulting Wannier orbitals form pairs, lying on either of the two slabs in a unit cell. The three pairs of Wannier orbitals feature different orientations and fall into two groups, as depicted from different viewing angles in Fig. 4b,c (Fig. 4d,e). All the orbitals are molecular-like and can be regarded as \(d_x^0\) and \(d_y^0/d_z^0\) orbitals of Mo atoms. The Wannier orbital with a \(d_x^0\) component fades out at Mo II and almost disappears at Mo I (the Mo tetrahedron layer), in agreement with the ab initio calculations projected onto inequivalent Mo atoms (Supplementary Fig. 12), and thus builds a 1D chain along \(b\), namely the C1 chain. The Wannier orbitals with \(d_x^0\) and \(d_y^0/d_z^0\) components also form chain-like structures extending along the \(b + c\) and \(b - c\) directions, which correspond to C2 and C3 chains and are related to each other by symmetry. In short, the Wannier orbitals feature identical orbital characters to those in the ab initio results (Fig. 4a) and extend along exactly the directions demanded within the hidden-1D scenario.

The observed hidden-1D electronic structure provides a natural explanation for many interesting properties of the quasi-2D \(\eta\)-Mo\(_4\)O\(_{11}\), such as the periodic distortion in the lattice structure, the FS nesting and the similarity with quasi-1D materials in optical spectra. However, the robust LL behaviour revealed in the current work is beyond the scope of canonical LL theory restricted to (quasi-)1D systems. Given the three arrays of chain-like structures in \(\eta\)-Mo\(_4\)O\(_{11}\) that can host the 1D LL physics separately, the crossed-chain geometry naturally materializes the theoretical proposal of a crossed sliding LL phase (Fig. 1b), where a 2D LL phase exists in the phase space, especially at finite temperatures and for low-energy excitations.

It is noteworthy that the original proposal of a crossed sliding LL is established in crossed chain arrays that are spatially separated. However, in \(\eta\)-Mo\(_4\)O\(_{11}\) all hidden-1D chains are constructed from the same octahedra in the Mo\(_2\)O\(_3\) slab (Fig. 4b–c) and thus cross through each other. To understand why the LL phase survives in such a 2D system, we conducted detailed analysis of the hopping amplitudes in the Wannier-orbital-based tight-binding Hamiltonian, which reveals minor inter-chain coupling. Within the parallel chain array, the intra-chain nearest-neighbour hopping amplitude is 327 (322) meV for \(d_x^0\) (\(d_y^0/d_z^0\)) Wannier orbitals along the \(b\) (\(b + c/b - c\)) direction, compared to the inter-parallel-chain and inter-crossed-chain hopping amplitudes of only several tens of meV (Supplementary Table 1 and Supplementary Tables 1a and 2). The large ratio between the intra- and inter-chain hopping amplitudes suggests a dimensional crossover energy scale of only 3–5 meV (ref. 17) (Supplementary Note 15), beyond which the LL behaviour can be well retained. Considering the energy resolution of the measurement (15 meV) and the thermal broadening during measurement (\(k_B T = 12\) meV at 135 K), the dimensional crossover effect can be ignored in our measurement and the conclusion about the LL behaviour in \(\eta\)-Mo\(_4\)O\(_{11}\) is not affected. We also note that the hopping amplitudes within the two slabs in a unit cell differ from each other by several meV due to the inter-slab coupling, leading to the slight band splitting of the three band pairs in the ab initio calculations (Fig. 4a).

This is absent in a previous calculation based on the extended Hückel method using the single-slab geometry. Combining our experiment and calculations, we emphasize that the 1D properties of the crossed chains, despite their overlapping in real space, are preserved due to their orthogonal orbital components. This ingeniously realizes the intriguing crossed LL phase in \(\eta\)-Mo\(_4\)O\(_{11}\) as schematically depicted in Fig. 4f (Supplementary Fig. 14).

The discovery of a crossed LL hidden in a quasi-2D material \(\eta\)-Mo\(_4\)O\(_{11}\), not only materializes the long sought-after phase, but also sheds light on the understanding of non-Fermi liquid behaviour in other 2D and 3D materials.

**Online content**

Any methods, additional references, Nature Research reporting summaries, source data, extended data, supplementary information, acknowledgements, peer review information; details of author contributions and competing interests; and statements of data and code availability are available at https://doi.org/10.1038/s41567-022-01829-z.

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Methods
Sample growth and characterization
Single crystals of $\eta$-Mo$_4$O$_{11}$ were grown by the chemical vapour transport technique\(^5\). High-purity reagents MoO$_3$ powder (Alfa Aesar, 99.0%) and MoO$_2$ powder (Alfa Aesar, 99.998%) mixed with a molar ratio of about 1:3 were used as the starting materials. TeCl$_4$ (Alfa Aesar, 99.9%) was used as the transport agent. The mixtures were thoroughly ground and sealed into a quartz tube under a vacuum of $10^{-5}$ mbar and subsequently placed in a two-zone furnace with a temperature profile of 560–510 °C to grow crystals. After maintaining this condition for 15 days, millimetre-sized $\eta$-Mo$_4$O$_{11}$ single crystals were obtained. For the X-ray diffraction slice cuts, a Bruker D8 single-crystal X-ray diffractometer with Mo Kα radiation ($\lambda = 0.71073$ Å) was used at 150 K. A back-reflection Laue detector (MWL120, Multiwire Laboratories) was also used to check the quality of the obtained single crystals and to determine the crystallographic orientation at room temperature.

Angle-resolved photoemission spectroscopy
High-resolution ARPES measurements were performed at beam line I05 of the Diamond Light Source (DLS), beam line APE of the Elettra Synchrotron Radiation Source, beam line 13U of the National Synchrotron Radiation Laboratory (NSRL), beam line 03U of the Shanghai Synchrotron Radiation Facility (SSRF), beam line 5–2 of the Stanford Synchrotron Radiation Lightsource (SSRL) and Tsinghua University. Data were collected with Scienta 4000 electron analysers at DLS, NSRL and SSRF; a Scienta SES2002 electron analyser at APE and Scienta DA30L electron analysers at SSRL and Tsinghua University. The overall energy and angle resolutions were set to 15 meV and 0.2°, respectively. The samples were cleaved in situ and measured under ultra-high vacuum below $1.0 \times 10^{-10}$ mbar.

Ab initio calculations
Ab initio band-structure calculations were performed using the QUANTUM ESPRESSO code package with the Perdew–Burke–Ernzerhof (PBE) type generalized gradient approximation (GGA)\(^5\)\(^1\)\(^2\). Experimental structural parameters were relaxed with a force threshold of 0.01 eV Å$^{-1}$. The cutoff energy for the plane-wave basis set was set to 600 eV and a $\Gamma$-centred Monkhorst–Pack \(k\)-point mesh of $11 \times 14 \times 3$ was used to obtain a self-consistent charge density. The Fermi surface and (projected) densities of states were calculated with a denser mesh of $22 \times 28 \times 6$. The Wannier orbitals were constructed using the tight-binding-based calculation supplied by the Wannier90 code\(^5\)\(^6\)\(^7\), based on which the surface-projected and tight-binding band structures were calculated using the WannierTools package\(^5\).

Data availability
The datasets that support the findings of this study are available from the corresponding authors on reasonable request.

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Author contributions
L.X.Y. conceived the experiments. X.D. and L.K. carried out ARPES measurements with the assistance of J.S.Z., X.G., R.Z.X., Q.Q.Z., Z.X.Y., W.X.Z., Y.D.L., S.M.H., D.P., M.X.W. and Z.K.L. Data analyses on the ARPES results and the ab initio calculations were performed by X.D. Single crystals were synthesized and characterized by Y.Y.L. and Y.B.C. The first draft of the paper was written by X.D.; L.X.Y. and Y.L.C. contributed to the revision of the manuscript. All authors contributed to the scientific planning and discussion.

Competing interests
The authors declare no competing interests.

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