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Stoichiometry, structure, and transport in the quasi-one-dimensional metal $\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$

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A correlation between lattice parameters, oxygen composition, and the thermoelectric and Hall coefficients is presented for single-crystal $\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$, a quasi-one-dimensional (Q1D) metallic compound. The possibility that this compound is a compensated metal is discussed in light of a substantial variability observed in the literature for these transport coefficients.

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I. INTRODUCTION

$\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$, known as “lithium purple bronze” (LiPB), is a low-temperature superconductor ($T_c \approx 2$ K) first synthesized and studied in the 1980s.^{1–3} It has attracted interest more recently for its quasi-one-dimensionality and Luttinger-liquid candidacy.^{4–9} It is distinguished among quasi-one-dimensional (Q1D) compounds by the absence of a conventional density-wave transition^{10,11} (either charge or spin) throughout a broad temperature range, $T \geq T_c$. An upturn in its resistivity below $T_M \sim 30$ K may be associated with localization, dimensional crossover, or the development of unconventional (e.g., electronically driven) charge density-wave order.^{10,11}

Values reported for the chain-axis (*b*-axis) electrical resistivity of LiPB^{2,10,12–17} vary by more than an order of magnitude, from 0.4 mΩcm to more than 10 mΩcm at 300 K, and appear to be responsible for similar differences in reported values for the anisotropy ratio, ρ_c/ρ_b (ρ_c is the resistivity along the next most conducting direction). Some of the differences may be associated with inadequate shorting,¹⁷ for current along the chains, of the voltage drops in directions transverse to the current flow, a delicate matter in highly anisotropic conductors. Increases in ρ_b for crystals¹⁸ annealed in air at 200 °C suggest that a variable oxygen stoichiometry also contributes to the reported differences. Recent thermoelectric and Hall measurements¹⁹ support such a view, indicating small differences in structure and stoichiometry that correlate with transport. This is an important issue given the renewed attention being paid to LiPB and the sensitivity of conduction in such low-dimensional materials to defects or impurities that may alter the low-energy electronic structure. A variable stoichiometry that allows for control of charge-carrier density could prove useful in the study of LiPB, which is proposed as a model, bulk system exhibiting Luttinger-liquid physics.

The principal finding reported here is a correlation between oxygen content and the *c* lattice parameter for a number of as-grown LiPB crystals. Seebeck (thermopower) and Hall coefficient measurements on LiPB crystals from our own work¹⁹ and those of others,^{15,16,20,21} are then presented together. Measured under open circuit conditions, the thermopower is not prone to the same challenges of transverse shorting that may complicate comparison of resistivity measurements. Hall measurements are also less susceptible to such errors given proper averaging with reversed current and magnetic field directions. The low-*T* chain-axis thermopower varies

considerably among the crystals measured,¹⁹ and correlates with their *c*-axis lattice parameters. Results for the Hall coefficient show considerable variability but the role of stoichiometry is less definitive as there are not enough data available on individual crystals for which structure, stoichiometry, and transport coefficients have all been measured. The possibility that LiPB is a compensated metal, motivated by the behavior of the thermoelectric coefficients,¹⁹ is examined in light of the Hall data.

II. CRYSTAL STRUCTURE AND STOICHIOMETRY

Single-crystal growth of $\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$ using a temperature-gradient flux method is described in detail elsewhere.^{2,11} The crystals grow as thin platelets, with $a \simeq 0.05$ – 0.20 mm and 0.5 – 2 mm for *b* and *c*. A Philips X’Pert x-ray diffractometer (Cu K_α , $\lambda = 1.54056$ Å) was employed in determining the crystallographic structure. Lattice parameters for the monoclinic unit cell²² were determined by from high-angle extrapolation²³ of reflections from various lattice planes (see Fig. 1) and angles β (between the *a* and *c* axes) from differences in *d*-spacings for the (*h*0*h*) and (*h*0 \bar{h}) reflections. Some crystals exhibit resolution-limited rocking curve widths, FWHM $\lesssim 0.05^\circ$ (without a monochromator), while others (typically larger crystals) exhibit a modest mosaicity, FWHM ~ 0.1 – 0.2° (see inset in Fig. 1). For the latter, rocking curves exhibit a convolution of two or more resolution-limited peaks, indicative of separate domains misaligned by small angles. A total of ten crystals were studied and their *a* and *b* lattice parameters were found to be the same within uncertainties, 12.752(2) Å and 5.520(2) Å, respectively. These agree with those determined from recent neutron scattering studies²⁴ on a powder specimen ground from thousands of similarly prepared crystals from our group.

The *c* lattice parameters were found to vary among the as-prepared crystals, correlating with oxygen content for seven of the crystals that were examined with x-ray energy-dispersive spectroscopy (EDS) on a system equipped with a polymer window suitable for light-element detection. Such variations, observed in the present study for crystals from the same growth batch and from different batches, is common in flux-grown crystals. The results from three or more EDS scans (with area $1 \mu\text{m} \times 1 \mu\text{m}$) from different locations on the surface of each crystal were averaged to determine the oxygen content. Li, not

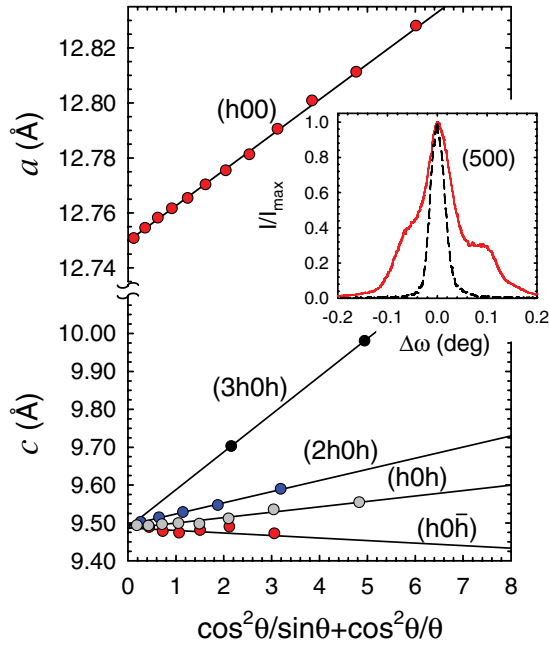


FIG. 1. (Color online) Examples of high-angle extrapolations of XRD reflections used to determine the a and c lattice parameters for $\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$ single crystals. Solid lines are least-squares fits. (Inset) Rocking curves of the (500) reflections for two crystals.

detectable in these scans, was not included in the compositional normalization [chemical analysis of the neutron scattering powder specimen²⁴ indicated a Li content 0.924(9)]. To ensure that the crystal surfaces (probed to a depth of several μm by EDS) were representative of the bulk, pieces of two crystals were polished to half their original thickness (corresponding to a depth of 50–100 μm) and examined by EDS for comparison. The average oxygen contents determined from measurements at several locations on the interior surfaces of these crystals differed from those of the original surfaces by no more than variations between different locations on each surface.

Figure 2 shows the correlation between c and oxygen content for these crystals. Standardless oxygen determinations from EDS typically show small systematic discrepancies due to inadequate accounting for absorption. As a measure of this discrepancy, we use the neutron scattering results,²⁴ which indicate full oxygenation (17 O/f.u., corresponding to 32.09 wt% ignoring Li) for $c = 9.4909(2)$ Å. Comparing our data for the same c values suggests that the EDS oxygen results are ≈ 2.5 wt% too low. The right (left) ordinates of Fig. 2 correspond to the EDS (corrected: EDS + 2.5 wt%) oxygen contents.

The maximum variation in oxygen content for the crystals examined, ~ 2 wt%, corresponds to ~ 1 oxygen atom per f.u. The corrected EDS oxygen scaling in Fig. 2 implies that some crystals have excess oxygen, presumably accommodated as interstitials. On the other hand, the fact that four of the crystals have the same measured oxygen content within uncertainties but differing c parameters suggests that other factors may influence c , e.g., variations in the Li concentration² or inhomogeneity of the oxygen distribution (on a scale smaller than the 1 μm). Though prior work² suggests a very narrow

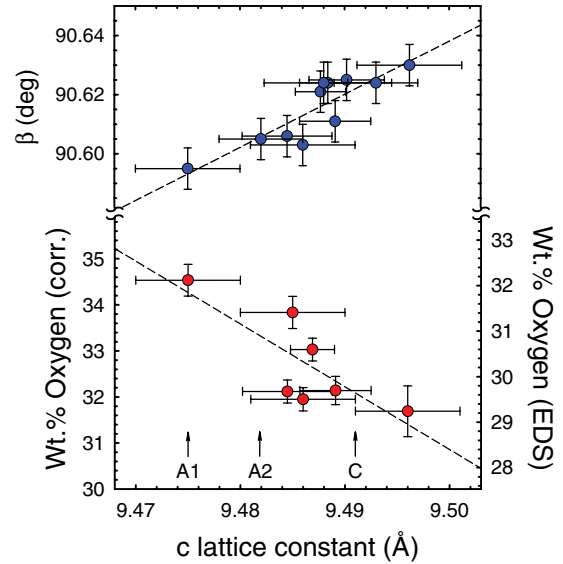


FIG. 2. (Color online) (Bottom) Oxygen content vs c lattice constant for $\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$ crystals. Right (left) ordinate scales correspond to EDS (EDS + 2.5 wt%) oxygen contents (see text). Error bars represent the standard deviation of EDS oxygen contents from multiple locations and of high-angle c -parameter extrapolations from multiple planes. Arrows labeled A1, A2, and C indicate the c lattice parameters for specimens discussed in the text. (Top) β vs c for all crystals studied.

range of Li nonstoichiometry (0.87–0.93 per f.u.), small Li concentration variations between crystals may exist and might correlate with those observed here for oxygen. The c parameter is thus a surrogate indicator for stoichiometric variation in oxygen and possibly Li. We note that a crystal with oxygen excess (labeled “A1” in Fig. 2) was remeasured after 18 months stored in a desiccator and found to have larger c (labeled “A2” in Fig. 2), suggesting a tendency of crystals with oxygen excess to slowly lose oxygen or for oxygen to redistribute, via diffusion, over long periods of time. Small increases in the angle β with increasing c parameter are shown in Fig. 2.

Interstitial oxygen in other oxides (e.g., cuprates) may affect the electronic structure through local charge doping, lattice distortion, or magnetic moment formation. The quasi-one-dimensional, conducting Mo-O double chains of LiPB may be especially susceptible to perturbations from such effects. The LiPB band structure^{3–5,7,8} is composed of two nearly-degenerate, Q1D bands crossing the Fermi energy (with $k_F \approx \pi/2b$), yielding two pairs of slightly warped Fermi surface (FS) sheets in the b^*-c^* planes with very little dispersion along a^* [see Fig. 3(a)]. The small splitting predicted for the bands near E_F from density functional theory^{5,8} (~ 30 – 40 meV) has not been observed in photoemission.^{4,6} The splitting is associated with an opposite warping of the FS sheets, attributable within tight-binding parametrizations^{7,8} to opposite signs (and comparable magnitudes) for the hopping integrals between constituent chains of a ladder ($t_{\perp 1}$) and between ladders ($t_{\perp 2}$) in neighboring unit cells [see Figs. 3(b) and 3(c)]. These transverse hopping parameters may be sensitive to variations in the c parameter.

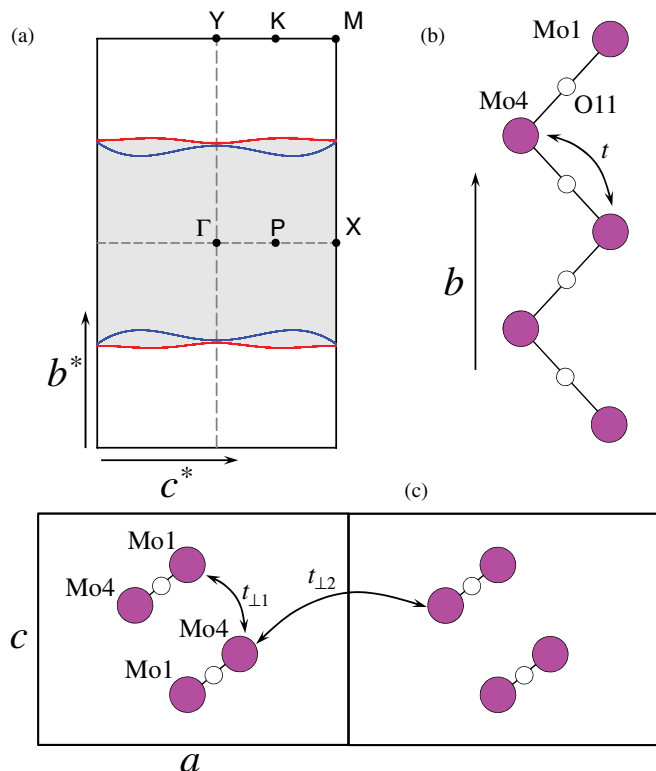


FIG. 3. (Color online) (a) Fermi surface from Ref. 5, (b) schematic of the conducting Mo-O-Mo zigzag chains along the crystallographic b axis, and (c) the arrangement of double chains within the ac plane.

III. THERMOPOWER

The b -axis thermopower of LiPB, $S_b(T)$ (see Fig. 4) was first reported in Ref. 20. More recently,¹⁹ $S_b(T)$ was reported for several crystals having different c parameters (letter labels in Fig. 4). Data for crystal D have been extended in the present work to 385 K. The inset of Fig. 4 shows S_b as a function of c . While the room-temperature thermopower is similar for all crystals, the dramatic low- T peak in S_b near $T = 50$ K correlates with the c parameter. The very large value (200 $\mu\text{V}/\text{K}$) found for specimen A1 with the smallest c parameter and highest oxygen content (see Fig. 2) is unusual for a metal. The largest Nernst coefficients (ν), with maxima near $T = 20$ K, were found¹⁹ for specimens C and D having the smallest values for S_b . This favors a two-carrier (ambipolar) conduction scenario in which very large hole and electron partial thermopowers, S^h and S^e ($S^e < 0$), nearly cancel due to compensation: $S_b \propto S^h + S^e$, $\nu \propto \mu(S^h - S^e)$ (μ is the carrier mobility). A departure from the compensation condition in specimens with larger c parameter could explain the appearance of their large low- T values for S_b . The mechanism underlying such very large partial thermopowers at low T remains to be established, but phonon-drag appears to be a plausible candidate.¹⁹

Though all specimens have positive 300 K thermopowers, their T dependencies approach a linear-in- T form with negative slope that results in negative values for S_b above room temperature (specimen D, Fig. 4). This implies a negative

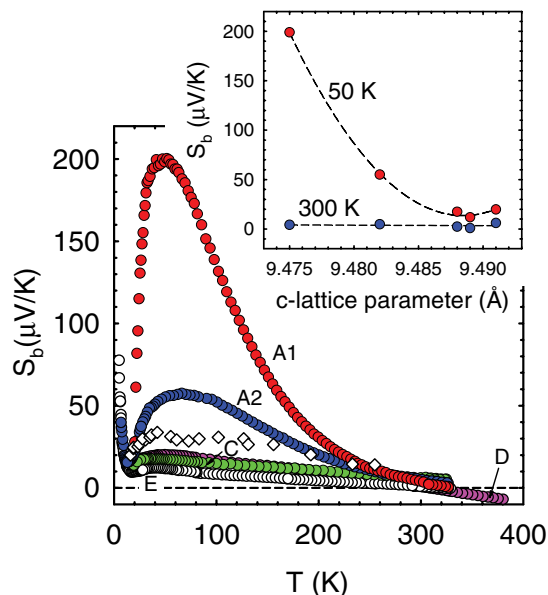


FIG. 4. (Color online) $S_b(T)$ for crystals from Ref. 19 (circles with letter labels) and Ref. 20 (diamonds). (Inset) S_b at $T = 50$ and 300 K vs c lattice parameter for crystals from Ref. 19.

(electron-like) carrier diffusion thermopower along the chains that is independent of the low- T behavior.

IV. HALL COEFFICIENT

The Hall coefficients of crystals A2 and C having $c = 9.482(4)$ and $9.491(4)$ \AA , respectively (arrows in Fig. 2), were reported elsewhere.¹⁹ Their oxygen contents were not measured; their Hall data are shown in Fig. 5. Also shown are data from three other sources in the literature.^{15,16,21} All of the

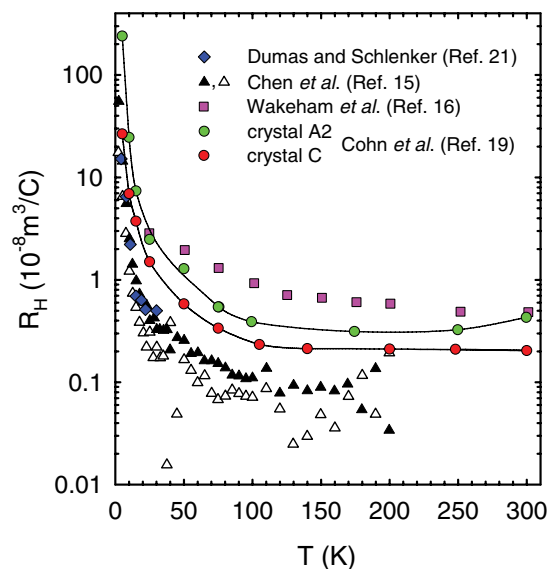


FIG. 5. (Color online) Hall coefficient for crystals A2 and C (see Ref. 19) and others from the literature. Data for Ref. 16 were computed by interpolation from their reported σ_{xy}/B , ρ_b , and ρ_c data as $R_H = (\sigma_{xy}/B)\rho_b\rho_c$.

data show very similar T dependencies, but magnitudes vary by a factor of 10. Note that $R_H > 0$ throughout the T range for all measurements.

At present it is not possible to draw conclusions about a dependence of R_H on c parameter with complete data sets only for specimens A2 and C (c was not reported along with the other data reproduced in Fig. 5). In a more recent work,¹⁷ studying crystals from the same source as that of Ref. 16, lattice parameters $a = 12.73 \text{ \AA}$ and $c = 9.51 \text{ \AA}$ where reported, both differing significantly from those reported here. Thus in addition to a stoichiometric variability observed for the present crystals, there may well be other differences between specimens prepared by different groups that have yet to be identified.

The Hall data might be reconciled with the compensation scenario motivated by the thermoelectric data. That R_H is always positive (a sign opposite to that of the diffusion thermopower) and varies substantially in magnitude among those crystals measured tends to support such a view. To examine this issue in more detail, first note that for Q1D systems with open Fermi surfaces the relation between the low-field Hall coefficient and carrier density differs from the free-electron expression, and for constant mean-free-path is generalized to²⁵ $R_H = \eta/(n|e|)$, where η measures the nonlinearity of the dispersion along the conducting chains, $\eta = -(\hbar k_F/v_F)\partial^2\epsilon/\partial k_b^2|_{\epsilon_F}$, and $v_F = \partial\epsilon/\partial k_b|_{\epsilon_F}$.

From the perspective of correlation effects, LiPB is one-quarter filled (approximately half an electron per band crossing E_F) with a tight-binding dispersion,^{7,8} $\epsilon(k_b) = -t \cos(k_b b/2)$. The Hall coefficient corresponding to $k_F = \pi/(2b)$ of LiPB⁴ would then be^{25–27} $R_H = -(\pi/4)/(n|e|) \simeq -1.7 \times 10^{-9} \text{ m}^3/\text{C}$ assuming $1.9e^-/\text{unit cell}$ as dictated by the chemistry and bonding. Though this value is in the middle of the experimental magnitudes near room temperature, the sign is opposite to those observed. The Hall coefficient for coupled Luttinger chains differs from the noninteracting case only by small power-law-in T corrections.²⁸ Theoretical treatments of the thermopower for single Luttinger chains with impurity scattering²⁹ indicate a linear temperature dependence for the carrier diffusion contribution similar to the noninteracting case.

A positive sign for R_H can arise from negative curvature in the dispersion ($\eta > 0$) along one or both of the FS sheets (e.g., the density functional bands^{3,8} suggest this is the case for the band that crosses at higher momentum along the P - K direction), or from a strong momentum dependence for the mean-free path³⁰ due to electron-electron^{25,31} or electron-phonon interactions, though such scattering anisotropy tends to be weaker at high T . Two-carrier fits¹⁹ to the R_H data for crystals A2 and C yield hole and electron partial Hall coefficients that are an order of magnitude or more larger

than R_H , implying carrier densities substantially smaller than expected. A reduced mobile carrier density is suggested by observations of localized charge in optical studies,¹⁰ as might be anticipated for carriers confined to chain fragments isolated by disorder.

An alternate, intriguing possibility is that strong correlations modify the FS from that predicted within density functional theory [see Fig. 3(a)]. The Mott gap (Δ) in LiPB appears to be quite small⁸ such that $t_{\perp} > \Delta$. In such systems, having strong on-site and longer-range Coulomb repulsion, calculations reveal^{32,33} a regime intermediate between 1D insulator and 2D metal where the suppression of the Mott gap via transverse hopping leads to the formation of a FS broken into narrow electron and hole pockets. This physics was recently invoked as a possible explanation for Fermi arcs found in photoemission studies of Y-124 due to the CuO chains.³⁴ Such a picture is appealing for LiPB as it offers a possible explanation for two-carrier physics apparent in the thermoelectric coefficients, positive values for R_H (provided there is a higher weighting of hole-like pockets), and carrier densities inferred from a two-carrier analysis of R_H that are much smaller than expected. Whether this scenario can be consistent with photoemission observations of the FS awaits a more complete and higher-resolution mapping throughout the Brillouin zone.⁹

V. SUMMARY

Variations in the c -lattice parameter of $\text{Li}_{0.9}\text{Mo}_6\text{O}_{17}$ crystals, attributable to stoichiometric variations in oxygen and possibly Li, correlate with the low- T thermopower. The implication is that the low-energy electronic structure is sensitive to small variations in stoichiometry in as-grown crystals. Thus the c parameter is revealed by this work to be an important metric with which to compare specimens. The data suggest formation of interstitial oxygen during growth as a possible source of oxygen variability. The available Hall data are less conclusive as to trends with c , but the observed variations in magnitude may be consistent with a two-carrier picture motivated by thermoelectric data.

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