New Luttinger-liquid physics from angle-resolved photoemission on a paradigm material

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Abstract

Li\textsubscript{0.9}Mo\textsubscript{6}O\textsubscript{17} is a paradigm material for studying Luttinger-liquid physics in the solid state. This paper summarizes recent and new photoemission studies directed at the quantum critical behavior that is expected for such a system. A theoretical description of the results requires a two-band Tomonaga–Luttinger model augmented by marginal interactions.

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The one-dimensional (1D) Luttinger liquid (LL), as idealized [1] by the Tomonaga–Luttinger model (TLM) [2], is a quantum critical (QC) system [3] in which competing fluctuations are frustrated from order by the one-dimensionality. There are no quasi-particle excitations, only independent collective excitations of the charge and spin density, known as holons and spinons, respectively, that disperse with different velocities $v_c$ and $v_s$. Electron removal or addition results entirely in holon/spinon generation and the system is said to display spin–charge separation and electron fractionalization. In correlation functions, such as that for the single-particle spectrum, QC behavior is manifested in power laws that show an absence of length and energy scales and display anomalous dimensions. Because temperature ($T$) is the only scale, critical “$E/T$” and “$vk/T$” scaling results, where $E$ is the energy relative to the Fermi energy $E_F$, $k$ is the momentum relative to the Fermi momentum, and $v$ is a constant with units of velocity. In approaching $E_F$, the energy dependence of the momentum-summed single-particle spectrum displays a power-law decay with an anomalous exponent $\alpha$, i.e. $|E|^\alpha$.

Li\textsubscript{0.9}Mo\textsubscript{6}O\textsubscript{17} is a quasi-1D metal, superconducting at low $T$ [4,5], having highly anisotropic electronic properties [4,6] and linearity of the dispersion of the underlying bands [7] crossing $E_F$ over a substantial energy range >0.1 eV away from $E_F$. The latter two properties favor LL physics and we have studied the single-particle spectrum of this material extensively by angle-resolved photoemission spectroscopy (ARPES) [8–13] with the purpose of establishing it as a paradigm for the study of LL physics in the solid state. The $k$-dependent spectra reveal a band structure in good general agreement with that of tight-binding band calculations [7], and detailed comparison to TLM spectral theory [14] shows [11,12] that the ARPES line shape has the expected holon and spinon features moving with different velocities. The momentum-integrated spectrum shows a clear power-law suppression near $E_F$. The power-law suppression has also been observed in scanning tunneling spectroscopy, however, with a different value of $\alpha$ [15].

In this paper, we summarize recent [16] and new work on the $T$-dependence of ARPES in the range 15–300 K to

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study and demonstrate the QC behavior of the system. In another work [17] not described here, we found that more bulk sensitive ARPES at high photon energy is essentially the same as that at low photon energy, consistent with our understanding from the crystal structure that the quasi-1D chains lie well below the likely cleavage plane and hence are locally in a bulk environment.

Experiments at the Wisconsin Synchrotron Radiation Center (SRC) were performed on the U1-NIM undulator beamline using an ARPES chamber equipped with an SES2002 Scienta electron analyzer. Single-crystal Li$_{0.9}$Mo$_6$O$_{17}$ samples grown using the temperature-gradient flux method were cleaved in situ to expose fresh surfaces for measurement. The sample temperature was maintained by a closed-cycle helium refrigerator and a resistance heater. Photon energies between 20 and 30 eV were used. The angular resolution in the experiment is 0.3° and the energy resolution is estimated to be 15–18 meV.

Figs. 1(a) and (b) are typical ARPES spectra showing the band structure of Li$_{0.9}$Mo$_6$O$_{17}$ for two directions parallel to the 1D chains. The intensity plot (a) is along $I$–$Y$ and the stack plot (b) is along a parallel line half-way between $I$–$Y$ and the zone boundary. Two of the four valence bands (C and D in Fig. 1(b)) merge together at binding energy $\sim$0.13 eV, disperse to cross $E_F$, and define the 1D Fermi surface. The D-band is not visible along the $k$-path of (a), probably due to a matrix element effect, but is clearly observed along the $k$-path of (b).

$T$-dependent spectra were $k$-summed along the chain direction to give the momentum-integrated spectra plotted in Fig. 1(c). A clear sharpening can be seen, and analysis shows it to be well beyond a simple thermal effect. To quantitatively assess the $T$-dependence, we fit the spectra using the finite-$T$ TLM spectral function [14]. The calculated line shape was convolved with a Gaussian representing the energy resolution and compared to experimental spectra. As presented in Ref. [16], essentially perfect agreement between experiment and theory at all temperatures could be obtained, but with a changing $\alpha$-value. It is noteworthy that the LL line shape for non-zero $T$ is not a perfect power law and that the good agreement between theory and experiment confirms this aspect of the TLM theory. The extracted $\alpha$~0.9 at high temperature and ~0.6 at low temperature is in good agreement with both previous ARPES and the low-temperature tunneling results.

As is also described in Ref. [16], this renormalization of $\alpha$ can be understood to result from marginal interactions among charge neutral modes present explicitly because of the two-band nature of Li$_{0.9}$Mo$_6$O$_{17}$. Invoking these extra modes is important. The resistivity of Li$_{0.9}$Mo$_6$O$_{17}$ shows a single power-law behavior over almost the same temperature range as that of our experiment. For the single-band TLM, this would contradict our observation in ARPES of a $T$-varying $\alpha$. However, the single-band TLM has only two modes, the holon ($\rho$) and spinon ($\sigma$), whereas having two bands gives four modes that are fluctuations of the

![Fig. 1. (a) Intensity plot of ARPES data along $I$–$Y$ at $T = 200$ K. (b) Stack plot of ARPES data along a line parallel to $I$–$Y$ but offset by 1/4 the Brillouin zone width. (c) Spectra $k$-integrated along $I$–$Y$ for multiple $T$. Inset shows wide energy range spectrum at 30 K.](image-url)
sups and differences of the densities of the two bands \((\rho_+, \rho_-, \sigma_+, \sigma_-)\). Marginal interactions for the charge neutral mode \((\rho_-)\) allow the renormalization of the \(\alpha\)-value while keeping the \(T\)-independence of the resistivity exponent. Quantitative renormalization group calculations give a good description of the measured \(\alpha(T)\), showing that \(\alpha\) renormalizes downwards because interactions in the \(\rho\)-sector become screened.

Figs. 2(a) and (b) show [11,12], respectively, overplots of ARPES data along the \(\Gamma-Y\) direction at \(T = 250\) K compared to TLM spectra calculated for the same \(T\), for \(\alpha = 0.9\), and with a ratio of holon to spinon velocity of 2. The combined effects of non-zero \(T\) and of including the experimental resolutions render unobservable the details of the sharp singularities and power laws of the \(T = 0\) theory curves. Nonetheless, the moving peak (holon) and leading edge (spinon) of the data and the theory are very similar and we have shown [11,12] that velocity ratios of 1.5 or 2.5 yield noticeably worse agreement.

The QC form of the TLM ARPES spectral function can be written as a universal function \(T^{(\alpha-1)}A(E/T,kv/T)\). To study QC scaling in Li\(_{0.9}\)Mo\(_6\)O\(_{17}\), we have examined the spectra along \(\Gamma-Y\) for multiple temperatures for \(k=0\) and \(k=cT\), where \(c\) is a constant. Fig. 2(c) gives a brief preliminary introduction to the nature of the findings. This figure shows the result of normalizing the \(k=0\) spectra to the spectral weight at \(E_F\) and plotting vs. \(E/k_B T\) on the horizontal axis. We see that for the leading spinon edge all the spectra fall together. The holon peaks are not completely shown, but it is evident that both their positions and amplitudes do not scale. For the leading edges above \(E_F\), we observe slight deviations from the scaling behavior at temperatures below 80 K and we can show that this is largely due to the finite energy resolution, which is increasingly important for a QC system as \(T\) is lowered.

The result of Fig. 2(c) suffices to show the basic result that Li\(_{0.9}\)Mo\(_6\)O\(_{17}\) is a QC system. In a more detailed paper to be published elsewhere, we present the results also for \(k=cT\), determine the prefactor of the QC scaling form, show the details of the behavior for the holon peak, and relate both the perfect and imperfect scaling behavior qualitatively to the same physics that underlies our understanding of the \(T\)-dependence of \(\alpha\). This connection is supported by the finding that the prefactor of the QC scaling form, although different from that of the theory, picks out the low \(T\)-value of \(\alpha\).

In summary, high-resolution \(T\)-dependent ARPES performed on the quais-1D metal Li\(_{0.9}\)Mo\(_6\)O\(_{17}\) shows LL and QC behaviors over a wide range of temperature. The particulars of the data require the consideration of a two-band TLM augmented by marginal interactions. Such studies of Li\(_{0.9}\)Mo\(_6\)O\(_{17}\) are revealing how LL physics plays out in an actual solid material.

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Fig. 2. (a) Overplotted ARPES spectra [10,11] along \(\Gamma-Y\) at 250 K. (b) Calculated TLM spectra for 250 K with experimental resolutions accounted for. (c) \(E/T\) scaling of \(k_F\) spectra at various \(T\). Inset shows same spectra before scaling.
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