Metal-Mott insulator interfaces

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Motivated by the direct observation of electronic phase separation in first-order Mott transitions, we model the interface between the thermodynamically coexisting metal and Mott insulator. We show how to model the required slab geometry and extract the electronic spectra. We construct an effective Landau free energy and compute the variation of its parameters across the phase diagram. Finally, using a linear mixture of the density and double-occupancy, we identify a natural Ising order parameter which unifies the treatment of the bandwidth and filling controlled Mott transitions.

INTRODUCTION

First-order transitions exhibit phase separation, and the real-space structure of the interface between the two thermodynamic phases contains information about the free energy functional [1]. Specifically, the thickness of the interface allows direct access to the ratio of the potential to kinetic energy terms in the free energy, which is related to the barrier height between the two minima of the double-well. A widely observed first-order transition in solid state systems is the Mott transition (reviewed in [2] for a large class of materials). Here, temperature, pressure or chemical doping drives a transition between a metal and a Mott insulator, a state where electrons cannot conduct due to the large ratio of the local Coulomb repulsion relative to the kinetic energy. While phase separation at the Mott transition is theoretically well-studied [3–8], the interface between the thermodynamically coexisting metal and Mott insulator is not. The recent development of experimental probes with nanometer-scale spatial resolution [9–11] has allowed the direct observation of the real-space structure of these interfaces.

As a first step towards characterizing the metal-Mott interface, we compute the real-space structure of the interfaces for the canonical example of a correlated system, the single-band Hubbard model. We use techniques in the spirit of work on correlated surfaces [12, 13] and heterostructures [14–18]. We extract the evolution of the density, double-occupancy and spectral features across the interface, allowing us to determine the parameters of the underlying free energy across the phase diagram.

LANDAU FREE ENERGY

The Mott transition can be tuned by two parameters besides temperature: the chemical potential $\mu$ and correlation strength $U$. At half-filling, extensive work has shown the first-order transition is analogous to the liquid-gas transition, placing the Mott transition within the Ising universality class [19–26]. In this work, we extend the construction away from half-filling into the $\mu$-$U$ plane [27]. Since we are interested in the metal-Mott interface, we work at temperatures below the critical point to construct our Landau theory.

We choose our fields to be the quantities conjugate to the external parameters ($\mu, U$), namely the density $n = \langle n \rangle$ and double occupancy $d = \langle n_\uparrow n_\downarrow \rangle$, a construction hinted at in [28]. The transition between the metal and paramagnetic Mott insulator does not break any symmetries [2, 29], so the terms in the free energy functional $F[n, d]$ are unconstrained. The free energy generically will have one global minimum, and should a tran-
FIG. 2. Generic Mott phase diagram, as realized by the single-band Hubbard model on a cubic lattice at \( T = 0.005 D \), where \( D = 6 \delta \) is the half-bandwidth. Dots label values of \((\mu, U)\) lying on first-order Mott transition line used for interface calculations. Lines mark the spinodals \( U_{c1} \) and \( U_{c2} \) where the insulating and metallic solutions respectively vanish. The diagram is symmetric about \( \mu/(U/2) = 1 \).

sition exist, it will occur via the switching between two discrete minima as no symmetry forces a locus of states to simultaneously lower in energy. We will explicitly construct the scalar order parameter in the following.

Along the Mott transition line in the \( \mu-U \) plane, the two minima will have the same energy. To facilitate analytic calculation, we take the two minima to be symmetric, an assumption certainly not justified by symmetry, but which will prove to be a good approximation. Writing the fields as \( \vec{\phi} = (n, d) \), the free energy functional takes a double-well form,

\[
\mathcal{F}[\vec{\phi}] = \frac{1}{2} D (\nabla \vec{\phi})^2 + \lambda (\vec{\phi} - \vec{\phi}_i)^2 (\vec{\phi} - \vec{\phi}_m)^2, \tag{1}
\]

where \( \vec{\phi}_i = (n_i, d_i) \) and \( \vec{\phi}_m = (n_m, d_m) \) are the insulating and metallic minima. A note on units: we work on a discrete lattice to easily connect with computation and omit an overall (dimensionless) normalization to the free energy.

A domain wall is given by the standard solution used, e.g. in the theory of instantons [30],

\[
\vec{\phi}(x_j) = \frac{\vec{\phi}_m + \vec{\phi}_i}{2} + \frac{\vec{\phi}_m - \vec{\phi}_i}{2} \tanh \left( \frac{x_j - x_0}{2\lambda} \right) \tag{2}
\]

where \( x_j \) is the coordinate of the \( j \)th site and the wall thickness is \( t^2 = 2(\lambda/D)(\vec{\phi}_m - \vec{\phi}_i)^2 \). Note the fields \( \vec{\phi} \) do not transform as a vector and the notation is for convenience. Determining the dependence of \( \vec{\phi}_i, \vec{\phi}_m \) and \( \lambda/D \) on \((U, T)\) requires microscopic modeling.

**MODELING THE INTERFACE**

The Hubbard Hamiltonian is the “standard model” of correlated electrons. Its two terms describe the competition between kinetic energy, which delocalizes electrons to promote metallic behavior, and mutual electron repulsion, which tends to localize electrons onto sites and drive the transition to a Mott insulator. We work with the simplest one-band case on a cubic lattice,

\[
H = \sum_{k\sigma}(\epsilon_k - \mu)n_{k\sigma} + U \sum_{j} n_{j\uparrow} n_{j\downarrow}, \tag{3}
\]

where we take \( \epsilon_k = -2t(\cos k_x + \cos k_y + \cos k_z) \) and use the half bandwidth \( D = 6\delta \) as the unit of energy in all the following. We will index the sites by \( j = (n_1, n_2, n_3) \) in the following. Ignoring ordered phases, which is a reasonable assumption at intermediate temperatures or in the presence of frustration, the phase diagram generically consists of a Mott insulating region for large \( U \) and a range of \( \mu \) corresponding to half-filling, and a Fermi liquid everywhere else. To find the first-order transition line, we use standard single-site dynamical mean-field theory (DMFT) [31–33] with a continuous-time quantum monte carlo (CTQMC) hybridization expansion impurity solver [34–36]. The phase diagram at \( T = 0.005 D \) is plotted in Fig. 2, along with the two spinodals \( U_{c1} \) and \( U_{c2} \) between which both solutions exist.

To model the interface in the coexistence regime, we fix our parameters to a point on the first-order line (dots in Fig. 2), then partition the lattice into three regions along the \( x \)-axis (see Fig. 3): metal \((n_1 \leq 0)\), insulator \((n_1 \geq N+1)\), and a transition region \((1 \leq n_1 \leq N)\). Here \( n_1 \) is the site index along the \( x \)-axis and we take \( N = 20 \) large enough to capture the interface. We perform an inhomogenous DMFT calculation by setting the self-energy of the lattice \( \Sigma_{n_1, n_1'} = \delta_{n_1, n_1'} \Sigma_{n_1} \),

\[
\Sigma_{n_1} = \begin{cases} 
\Sigma_{\text{metal}} & n_1 \leq 0 \\
\Sigma_{n_1} & 1 \leq n_1 \leq N \\
\Sigma_{\text{ins}} & n_1 \geq N + 1 
\end{cases} \tag{4}
\]
Only the self-energies $\Sigma_{n_1}$ in the transition region are updated, while $\Sigma_{\text{metal}}$ and $\Sigma_{\text{ins}}$ are fixed boundary conditions taken from the single-site DMFT solution. Our setup assumes the interface is perpendicular to one of the crystal directions ($x$) and the system is translationally invariant in the other two ($y$ and $z$) so self-energies are independent of $n_2$ and $n_3$.

To render the equations soluble in the transition region, we compute the lattice Green’s function and use its local component $G_{n_1 n_1}$ to map the system to a chain of $N$ auxiliary impurity problems [18],

$$G_{n_1 n_1}(i\omega_n) = \frac{1}{i\omega_n - E_{\text{imp}} - \Delta_{n_1}(i\omega_n) - \Sigma_{n_1}(i\omega_n)}.$$  \hspace{1cm} (5)

Using the extracted impurity levels $E_{\text{imp}}$ and hybridization functions $\Delta_{n_1}$, we obtain the new local self-energies $\Sigma_{n_1}$ and iterate to convergence. The procedure for computing the local Green’s function is provided in the Supplementary Material.

FIG. 4. Variation of the density (top) and double occupancy (bottom) across the interface at several points along the Mott transition line at $T = 0.01D$. Thin lines are fits to the standard solution for a double-well potential $a + b\tanh((x_i - x_0)/2l)$, allowing extraction of the parameters for the underlying free energy. Curves are shifted horizontally by varying amounts for clarity. The chemical potential is in units of $U/2$, as detailed in the right-hand table of Table I.

FIG. 5. Trajectory in $(n,d)$ space as the system evolves across the interface from the insulating to the metallic minima at $\mu = 0.95(U/2)$, $U = 2.05D$, $T = 0.005D$. The extracted Landau parameters are used to plot the contours of the double-well potential. The dotted lines trace the shift of the minima along the Mott transition line at $T = 0.005D$.

RESULTS

The evolution of the density $n$ double occupancy $d$ for along the Mott transition line is displayed in Fig. 4 at a temperature of $T = 0.005D$. At the particle-hole symmetric point, there is no jump in density between the metal and Mott insulator, while the change in double-occupancy is maximal. As we progress along the transition line (which we parameterize with the chemical potential $\mu$) towards the hole-doped side, the density difference between the metal and insulator increases. Additionally, the density of the insulator drops below unity because we are at finite temperature. In contrast, the jump in double occupancy decreases.

The variation of both quantities fit well to Eq. 2 for the double well potential, albeit with slightly different length scales, and we use the average of the two wall widths to compute $\lambda$. The small difference in length scales implies the potential is not perfectly symmetric (as expected), and that the path in $(n,d)$ space between the two minima is close to, but not exactly, a straight line (see Fig. 5). The extracted parameters for the Landau free energy are presented in Table I.

We also analytically continue the Matsubara self-energies produced by the impurity solver to the real axis to compute the variation of the spectral density across the interface. We plot in Fig. 1 the spectra for parameters $\mu = 0.95(U/2)$, $U = 1.97D$ and $T = 0.01D$, which is slightly on the hole-doped side. Starting from the metallic solution, we find that the quasiparticle peak shift slightly downwards and disappears into the lower Hubbard band as we progress to the Mott insulator. The gap between the Hubbard bands slightly narrow.

The extracted parameters combined with our ansatz
(Eq. 1) allow us to reconstruct the free energy. Shown in Fig. 5 is a representative case for \( \mu = 0.95, U = 2.05, T = 0.005D \). We have plotted the trajectory in \((n, d)\) space as the system evolves from the metallic to insulating minima, superimposed with contour lines of the potential constructed using the extracted parameters. The movement of the two minima as we step along the Mott transition line is shown in the dotted lines.

As promised, we explicitly construct the order parameter field \( \Delta \) as a linear combination of \( n \) and \( d \), owing to the fact that the trajectory is almost straight. The construction is essentially geometric: we take the line segment joining the two minima and parameterize it with an angle \( \alpha \):

\[
\Delta = (n - \bar{n}) \sin \alpha + (d - \bar{d}) \cos \alpha
\]

where \( \bar{n} = (n_i + n_m)/2 \) and \( \bar{d} = (d_i + d_m)/2 \). The angles are tabulated in Table I. At particle-hole symmetry, the angle is zero and the variation of the order parameter is entirely driven by the double occupancy. Increasing angles imply the density becomes a larger component of the order parameter, which occurs as we progress to larger correlation strengths.

### SUMMARY

In this work, we have taken a first step towards characterizing the metal-Mott interface by modeling its spatial properties, constructing a Landau free energy and identifying an Ising order parameter. The key parameter of the free energy which could not be obtained by previous solutions in homogenous geometries is the interface width \( l \), which is directly related to the double-well barrier height via \( \lambda/D \). We also comment that while in general for first-order transitions which do not possess an organizing symmetry, any number of fields can be chosen to construct the free energy [20, 21, 26], the choice of the quantities conjugate to the physical tuning parameters \( \mu \) and \( U \) allow for an especially transparent construction of the order parameter which can uniformly treat both the bandwidth and filling controlled transitions.

We want to point out the simplifying assumptions used: (1) we took the interface to be perpendicular to a crystallographic axis, (2) we only included nearest-neighbor hopping to simplify the formulae, (3) we made the slow-varying approximation, assuming each site was an independent impurity affecting the others only via the hybridization, and (4) we have ignored the long-range Coulomb interaction. Relaxing these assumptions to capture more realistic scenarios warrant further investigation.

We expect that future calculations on realistic systems will provide quantitative results for comparison with near-field optics and STM observations, and more speculatively, could provide a new constraint on the value of \( U \) in these compounds. Finally, while we have guessed the form of the Landau free energy and numerically determined its parameters, especially satisfying for future work would be a microscopic derivation from the appropriate mean-field theory.

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SUPPLEMENTARY: CALCULATION OF LOCAL GREEN’S FUNCTION

The Green’s function of the lattice is given by

\[ G_{RR'} = [(i\omega + \mu)\delta_{RR'} - t_{RR'} - \Sigma_{RR'}]^{-1} \tag{7} \]

where \( R \) is a lattice vector \( R = (n_1, n_2, n_3) \) with the cubic primitive lattice vector and \( t_{RR'} \) denotes the nearest neighbor hopping. To see the spatial variation across the two different phases, we divide the lattice into three regions: metallic \((M : -\infty < n_1 \leq 0)\), insulating \((I : N + 1 \leq n_1 < \infty)\) and transition \((T : 1 \leq n_1 \leq N)\) region. So \( T \) is sandwiched by \( M \) and \( I \). Then we assign to each site the localized self-energy \( \Sigma_{n_1n_1'} = \delta_{n_1n_1'}\Sigma_{n_1} \) with

\[ \Sigma_{n_1} = \begin{cases} \Sigma_{\text{metal}} & (n_1 \in M) \\ \Sigma_{n_1} & (n_1 \in T) \\ \Sigma_{\text{ins}} & (n_1 \in I) \end{cases} \tag{8} \]

Note that in the metallic and insulating regimes, the self-energy is fixed to \( \Sigma_{\text{metal}} \) and \( \Sigma_{\text{ins}} \) respectively, while we allow the local self-energy in the transition regime to vary across the sites.

The Fourier transformation of Eq. (7) along \( y \) and \( z \) directions gives the following matrix form of Green’s function in the mixed representation \((n_1; k_y, k_z)\) \((n_1 \) is the site index of \( x)\):

\[ [G(k_y, k_z; i\omega)]_{n_1n_1'} = \begin{bmatrix} (i\omega + \mu - \varepsilon(k_y, k_z) \\ -\Sigma_{n_1}(i\omega)) [\hat{t} - i \hat{t}'] \end{bmatrix}^{-1} \tag{9} \]

where \( \hat{t} = -(\delta_{n_1,n_1'} + \delta_{n_1,n_1'} - 1) \) and \( \varepsilon(k_y, k_z) = -2t\cos(k_ya) + \cos(k_za) \). To apply DMFT to the transition regime, we must calculate the local component of the Green’s function at each site and map each onto an auxiliary impurity.

We can rewrite Eq. (9) in a block matrix divided into the three regimes \( M, T \) and \( I \), that is,

\[ [G(k_y, k_z; i\omega)]_{n_1n_1'} = \begin{bmatrix} F_M & t & F_T \\ t & F_T & t \\ F_T & t & F_I \end{bmatrix}^{-1} \tag{10} \]

where we define the three block matrices by

\[ [F_M]_{n_1n_1'} = (i\omega + \mu - \varepsilon(k_y, k_z) - \Sigma_M(i\omega)) \delta_{n_1n_1'} - t_{n_1n_1'} \equiv z_M \]

\[ [F_T]_{n_1n_1'} = (i\omega + \mu - \varepsilon(k_y, k_z) - \Sigma_n(i\omega)) \delta_{n_1n_1'} - t_{n_1n_1'} \equiv z_T \]

\[ [F_I]_{n_1n_1'} = (i\omega + \mu - \varepsilon(k_y, k_z) - \Sigma_{\text{ins}}(i\omega)) \delta_{n_1n_1'} - t_{n_1n_1'} \equiv z_I \]

Note that \( z_M \) and \( z_T \) are fixed while \( z_I \) varies across the sites.

Using block matrix inversion

\[ \begin{bmatrix} A & B \\ C & D \end{bmatrix}^{-1} \bigg|_{\varepsilon A} = [A - BD^{-1}C]^{-1} \tag{11} \]

we obtain the complete form of Green’s function in the transition regime \( T \) (a \( N \times N \) matrix) into which all the degrees of freedom of metallic and insulating regions are incorporated:

\[ [G(k_y, k_z)]_{n_1n_1'} = \begin{bmatrix} F_T \end{bmatrix} - (t_{\text{T,M}}[F_M]^{-1}t_{\text{T,M}} + t_{\text{T,I}}[F_I]^{-1}t_{\text{T,I}})^{-1} \]

\[ = \begin{bmatrix} z_{11} - t^2R_M & t & 0 \\ t & z_{22} & t \\ t & \ddots & \ddots \\ t & \ddots & \ddots & \ddots & t \\ 0 & t & z_{N-1,N-1} & t & t \\ & \vdots & \vdots & \ddots & \ddots & \ddots & \ddots \end{bmatrix}^{-1} \tag{12} \]

where \( R_M \equiv [F_M^{-1}]_{00}, R_I \equiv [F_I^{-1}]_{N+1,N+1} \) and \( t_{\text{T,M}(I)} \) is the overlap between \( T \) and \( M(I) \). The effect of integrat-
ing out the degrees of freedom in $\mathcal{M}$ and $\mathcal{I}$ is captured by $t^2 R_{\mathcal{M}}$ and $t^2 R_{\mathcal{I}}$ at the $(1, 1)$ and $(N, N)$ components respectively.

To compute $R_{\mathcal{M}}$ and $R_{\mathcal{I}}$, we again rely on Eq. (11). Since $[F_{\mathcal{M}}]$ takes a symmetric tridiagonal matrix form equal to

$$F_{\mathcal{M}} = \begin{bmatrix} z_M & t & & & \\ t & z_M & t & & \\ & t & z_M & \ddots & \\ & & \ddots & \ddots & t \\ & & & t & z_M \end{bmatrix} = \begin{bmatrix} z_M & t \\ t & F_{\mathcal{M}} \end{bmatrix}$$

we see the matrix repeats itself inside. As a direct consequence of (11), we obtain the following recursive equation:

$$[F^{-1}_{\mathcal{M}}]_{00} = R_{\mathcal{M}} = \frac{1}{z_M - t^2 R_{\mathcal{M}}}$$

where the solution is

$$R_{\mathcal{M}} = \frac{z_M - \sqrt{(z_M)^2 - 1}}{t}.$$  (15)

$R_{\mathcal{I}}$ is obtained by the same procedure.

Finally, we need to convert the mixed representation form (10) into the pure real-space representation. Performing the inverse Fourier transformation with respect to $k_y$ and $k_z$, we can obtain the local Green’s function at the site $n_1$

$$[G]_{n_1 n_1} = \frac{d^2 k}{(2\pi)^2} [G(k_y, k_z)]_{n_1 n_1} = \int d\varepsilon [G(\varepsilon)]_{n_1 n_1} D^{2D}(\varepsilon)$$

where the $\varepsilon$ dependence of $G$ comes from $\varepsilon = \varepsilon(k_y, k_z)$. Here, $D^{2D}(\varepsilon)$ is the density of states of non-interacting 2D square lattice whose analytic expression is known and the integration (16) is performed numerically.