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Surface dead layer for quasiparticles near a Mott transition

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Electron quasiparticles are progressively weakened by correlations upon approaching a continuous Mott metal insulator transition in a bulk solid. We show that corresponding to the bulk weakening, a dead layer forms below the surface of the solid, where quasiparticles are exponentially suppressed. The surface dead layer depth is a bulk property, and diverges when the Mott transition is approached. We describe this phenomenon in a Hubbard model within a self-consistent Gutzwiller approximation. Photoemission data of Rodolakis *et al.* in V_2O_3 appear to be in accord with this physical picture.

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The Mott transition[1] where a lattice of atoms or molecules abandons the metallic state and turns insulating due to electron-electron repulsion, has a very intuitive physical explanation. Electron motion in the lattice is caused by kinetic energy, and favored by electron-ion energy because the same electron can feel in this way the attraction of more than one nucleus. It is opposed by Coulomb repulsion, higher for itinerant electrons due to the higher chance of collision during motion. When the first two terms (which form the band energy) prevail, the system is a band metal; otherwise the electrons localize, and we have an insulator. Despite that conceptual simplicity, properties of Mott insulators and especially of the strongly correlated metallic state close to a Mott transition remain quite difficult to capture both theoretically and experimentally. Theoretically, the reason is that the Mott transition is a collective phenomenon, which escapes single-particle or mean field theories such as Hartree-Fock or DFT-LDA approximations. Experimentally, complications such as magnetism, lattice distortions, etc., often conspire to mask the nature of metal insulator transitions.

Fresh progress on this problem has come in the last two decades with dynamical mean field theory (DMFT)[2], which in the standard Hubbard model showed that, as the electron-electron repulsion parameter U increases, the initial band-metal evolves first to a strongly correlated metal well before the Mott transition. In the strongly correlated metal the electron spectral function undergoes a profound change exhibiting well formed, localized Mott-Hubbard bands coexisting with delocalized, propagating quasiparticles – the latter narrowly centered in energy near the Fermi level. Only successively do the quasiparticles disappear as the Mott transition takes place when U is increased to reach $U = U_c$. This intriguing prediction – simultaneous metallic and insulating features, though on well separated energy scales – has stimulated a considerable experimental effort to reveal coexisting quasiparticles and Mott-Hubbard bands

in strongly correlated metals[3, 4, 5, 6, 7, 8, 9, 10, 11, 12]. A large amount of work has been done on V_2O_3 , the prototype compound where a Mott transition was first discovered[13] and studied theoretically[14, 15]. At the metal-insulator transition of $(V_{1-x}Cr_x)_2O_3$, early photoemission experiments[16, 17, 18, 19] failed to reveal the sharp quasiparticle peak predicted by DMFT. The electronic spectrum was simply dominated by the lower Mott-Hubbard band with barely a hint of metallic weight at the Fermi energy. A similar puzzle was actually reported much earlier in f -electron materials[20], and soon ascribed to large surface effects in the presence of strong correlations[21]; the same conclusion reached by more recent photoemission experiments[3, 4, 6, 7, 11, 12, 22]. In V_2O_3 , using higher kinetic energy photo-electrons, whose escape depth is larger, a prominent quasiparticle peak coexisting with incoherent Mott-Hubbard bands was eventually observed [5, 10, 23]. Quasiparticle suppression in surface-sensitive probes was attributed[23] to surface-modified hamiltonian parameters, the reduced atomic coordination pushing the surface closer to the Mott transition than the underlying bulk. Larger electronic correlations at the surface have been discussed by several authors through *ad-hoc* formulations of DMFT[25, 26, 27]. There is general agreement on intrinsically different quasiparticle properties near a surface, even if all hamiltonian parameters were to remain identically the same up to the outermost atomic layer[25].

This conclusion, although not unexpected, raises a more fundamental question. A metal does not possess any intrinsic length-scale at long distances other than the Fermi wavelength. Thus an imperfection like a surface can only induce at large depth a power-law decaying disturbance such as that associated with Friedel's oscillations. Since one does not expect Luttinger's theorem to break down, even in a strongly correlated metal these oscillations should be controlled by the same Fermi wavelength as in the absence of interaction, irrespectively of the proximity of the Mott transition. However, a

strongly correlated metal does possess an intrinsic energy scale, the parametric distance of the Hamiltonian from the Mott transition, where that distance could be associated with a length scale. The surface as a perturbation should alter the quasiparticle properties within a depth corresponding to that length, a bulk property increasing near the Mott transition, unlike the Fermi wavelength that remains constant. In this respect, it is not *a priori* clear whether the recovery of bulk quasiparticles spectral properties with increasing depth should be strictly power-law, compatible with the common view of a metal as an inherently critical state of matter, or whether it should be exponential, as one would expect by regarding the Mott transition as any other critical phenomena where power laws emerge only at criticality. We find here in the simple half-filled Hubbard model that the quasiparticle spectral weight below the surface is actually recovered exponentially inside the bulk with a length-scale that depends only on the bulk properties and diverges approaching the continuous Mott transition.

To address the generic surface features of a strongly correlated metal, we study the simplest Hamiltonian exhibiting a Mott transition, namely the Hubbard model at half-filling

$$H = -t \sum_{\langle \mathbf{R}\mathbf{R}' \rangle \sigma} c_{\mathbf{R}\sigma}^\dagger c_{\mathbf{R}'\sigma} + H.c. + \sum_{\mathbf{R}} U_{\mathbf{R}} n_{\mathbf{R}\uparrow} n_{\mathbf{R}\downarrow}, \quad (1)$$

where $\langle \mathbf{R}\mathbf{R}' \rangle$ are nearest neighbor sites, $c_{\mathbf{R}\sigma}^\dagger$ creates an electron at site \mathbf{R} with spin σ and $n_{\mathbf{R}\sigma} = c_{\mathbf{R}\sigma}^\dagger c_{\mathbf{R}\sigma}$. Conventionally, the Mott transition of the half-filled Hubbard model is studied restricting to the paramagnetic sector of the Hilbert space[2, 14, 15] so as to avoid spurious effects due to magnetism. We assume a cubic lattice of spacing a with periodic boundary conditions in x and y directions and open boundary conditions in the z direction, in an N -layer slab geometry with two surfaces at $z = 0$ and $z = Na$. The Hubbard electron-electron interaction parameter $U_{\mathbf{R}}$ is U everywhere except at the top surface layer ($z = 0$), where it takes a generally higher value $U_s > U$. In this way we can compare effects at the ideal lower surface ($z = Na$), where $U_{Na} = U$, with the more correlated upper surface ($z = 0$). DMFT[2] offers an ideal tool to attack this model in the paramagnetic sector, assuming a local self-energy that depends on the layer index z [25, 26, 27]. However, a full DMFT calculation of this sort is numerically feasible only for a small number of layers, e.g. $N = 20$ as in Ref.[28], making the critical regime near the Mott transition hard to access. As a useful approximate alternative, one can resort to the so-called linearized DMFT[25, 29] to treat moderately larger sizes. We decided to adopt a different method altogether, the Gutzwiller variational approximation[30]. Despite its

limitations (static mean field character; inability to describe the insulating phase) it is known to provide a good description of quasiparticle properties close to the Mott transition[2] with very little size-limitations, and great simplicity and flexibility (it may treat intersite interactions, any kind of lattice, etc.). We study (1) by means of a Gutzwiller type variational wavefunction

$$|\Psi\rangle = \prod_{\mathbf{R}} \mathcal{P}_{\mathbf{R}} |\Psi_0\rangle, \quad (2)$$

where $|\Psi_0\rangle$ is a paramagnetic Slater determinant. The operator $\mathcal{P}_{\mathbf{R}}$ has the general expression

$$\mathcal{P}_{\mathbf{R}} = \sum_{n=0}^2 \lambda_n(z) |n, \mathbf{R}\rangle \langle n, \mathbf{R}|, \quad (3)$$

where $|n, \mathbf{R}\rangle \langle n, \mathbf{R}|$ is the projector at site $\mathbf{R} = (x, y, z)$ onto configurations with n electrons, and $\lambda_n(z)$ are layer-dependent variational parameters. We calculate average values on $|\Psi\rangle$ using the so-called Gutzwiller approximation[31, 32], (for details see e.g. Ref.[30], whose notations we use hereafter), and require that

$$\langle \Psi_0 | \mathcal{P}_{\mathbf{R}}^2 | \Psi_0 \rangle = 1, \quad \langle \Psi_0 | \mathcal{P}_{\mathbf{R}}^2 n_{\mathbf{R}\sigma} | \Psi_0 \rangle = \langle \Psi_0 | n_{\mathbf{R}\sigma} | \Psi_0 \rangle. \quad (4)$$

Because of particle-hole symmetry, $\langle \Psi_0 | n_{\mathbf{R}\sigma} | \Psi_0 \rangle = 1/2$, from which it follows that Eq. (4) is satisfied if $\lambda_2(z) = \lambda_0(z)$, $\lambda_1(z)^2 = 2 - \lambda_0(z)^2$. The average value of (1) is then[30, 33]

$$E = \frac{\langle \Psi | H | \Psi \rangle}{\langle \Psi | \Psi \rangle} = \sum_{\mathbf{R}} \frac{U_{\mathbf{R}}}{4} \lambda_0(z)^2 - t \sum_{\langle \mathbf{R}\mathbf{R}' \rangle \sigma} R(z) R(z') \langle \Psi_0 | c_{\mathbf{R}\sigma}^\dagger c_{\mathbf{R}'\sigma} + H.c. | \Psi_0 \rangle, \quad (5)$$

where $R(z) = \lambda_0(\mathbf{R}) \sqrt{2 - \lambda_0(\mathbf{R})^2}$ plays the role of a wavefunction renormalization factor. Its square is the actual quasiparticle weight, $Z(z) = R^2(z)$, since quasiparticle creation renormalizes into $R(z) c_{\mathbf{R}\sigma}^\dagger$ in Fermi liquid theory. One can invert this equation to express $\lambda_0(z)$ as function of $R(z)$, which become the actual variational parameters together with the Slater determinant $|\Psi_0\rangle$. In order to minimize E in Eq. (5) we assume that the Slater determinant $|\Psi_0\rangle$ is built with single-particle wavefunctions that, because of the slab geometry, have the general expression $\phi_{\mathbf{e}\mathbf{k}_{||}}(\mathbf{R}) = \sqrt{1/A} e^{i\mathbf{k}_{||} \cdot \mathbf{R}} \phi_{\mathbf{e}\mathbf{k}_{||}}(z)$, where A is the number of sites per layer and $\mathbf{k}_{||}$ the momentum in the x - y plane. The stationary value of E with respect to variation of $\phi_{\mathbf{e}\mathbf{k}_{||}}(z)$ and $R(z)$ corresponds to the coupled equations

$$\epsilon \phi_{\epsilon \mathbf{k}_{||}}(z) = R(z)^2 \epsilon_{\mathbf{k}_{||}} \phi_{\epsilon \mathbf{k}_{||}}(z) - t R(z) \sum_{p=\pm} R(z+pa) \phi_{\epsilon \mathbf{k}_{||}}(z+pa), \quad (6)$$

$$R(z) = \frac{4\sqrt{1-R(z)^2}}{U(z)A} \sum_{\epsilon \mathbf{k}_{||}}^{\text{occupied}} \left[-2R(z) \epsilon_{\mathbf{k}_{||}} \phi_{\epsilon \mathbf{k}_{||}}(z)^2 + t \phi_{\epsilon \mathbf{k}_{||}}(z) \sum_{p=\pm} R(z+pa) \phi_{\epsilon \mathbf{k}_{||}}(z+pa) \right], \quad (7)$$

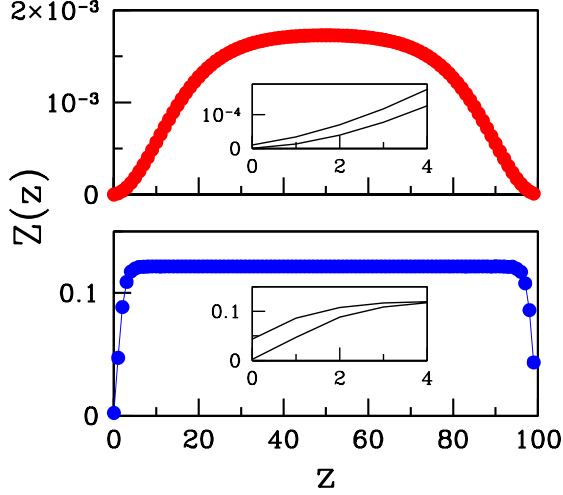


Figure 1: (Color online) The quasiparticle weight $Z(z) = R^2(z)$ as function of the coordinate z perpendicular to the surface (in units of the lattice spacing) for a 100-layer slab. The interaction parameter at $z = 0$ is $U_s = 20t$, while the bulk U is $15.98t$ in the upper panel and $15t$ in the lower one (while $U_c = 16$). The insets show the behavior of Z close to the two surfaces; the highest curve corresponding to the bulk-like surface, the other to $U_s = 20t$.

where $\epsilon_{\mathbf{k}_{||}} = -2t(\cos k_x a + \cos k_y a)$ and the sum in Eq. (7) runs over all pairs of $(\epsilon, \mathbf{k}_{||})$ that are occupied in the Slater determinant $|\Psi_0\rangle$. The first equation has the form of a Schrödinger equation that the single-particle wavefunctions $\phi_{\epsilon \mathbf{k}_{||}}(z)$ must satisfy, depending parametrically on $R(z)$. The second equation has been intentionally cast in the form of a map $R_{j+1}(z) = F[R_j(z), R_j(z+a), R_j(z-a)]$ whose fixed point we have verified to coincide with the actual solution of (7) in the parameter region of interest. Eqs. (6) and (7) can be solved iteratively as follows. First solve the Schrödinger equation at fixed $R_j(z)$; next find the new $R_{j+1}(z)$ using the old $R_j(z)$ and the newly determined wavefunctions $\phi_{\epsilon \mathbf{k}_{||}}(z)$. With the new $R_{j+1}(z)$, repeat the above steps and iterate until convergence. Because of the large number of variational parameters, this iterative scheme is much more efficient than – while fully equivalent to – a direct minimization of E , Eq. (5).

In Fig. 1 we plot $Z(z) = R^2(z)$, experimentally the total spectral weight carried by quasiparticles, calculated

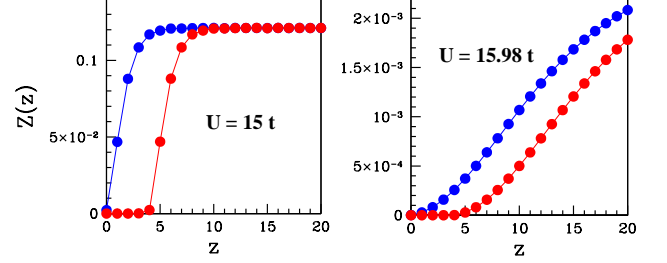


Figure 2: Quasiparticle weight dependence on the distance z from the surface for two different bulk U values and for two cases: one where only the first layer has $U_s = 20t > U$ (upper curve in each panel), the other where five surface layers have $U_s = 20t$.

as function of z (in units of the lattice spacing a) for $U_s = 20t$, for two different bulk values $15t$ and $15.98t$ of U below the critical Mott-transition value $U_c = 16t$. Coming from the bulk, the quasiparticle weight $Z(z)$ decreases monotonically on approaching both surfaces, where it attains much smaller values than in bulk. As expected, the more correlated surface has a smaller quasiparticle weight, $Z(0) < Z(N)$. Note however that so long as the slab interior (the “bulk”) remains metallic, the surface quasiparticle weight never vanishes no matter how large U_s [25]. Mathematically, this follows from Eq. (7), which is not satisfied by choosing $R(0) = 0$ while $R(z > 0) \neq 0$. Physically, some metallic character can always tunnel from the interior to the surface, so long as the bulk is metallic. The quasiparticle weight approaches the surface with upward curvature when U is closest to U_c , upper panel in Fig. 1, whereas the behavior is linear well below U_c , as found earlier within linearized DMFT[25]. We note that an upward curvature is in better accord with photoemission spectra of Rodolakis *et al.* on V_2O_3 [34]. The curvature becomes more manifest if the number of surface layers where $U_s > U$ is increased, as shown in Fig. 2. Next, we analyse the dependence of $R(z)$ at large distance $1 \ll z \ll N/2$ below the surface. As Fig. 3 shows, we find no trace of a power law, and R is best fit by an exponential $R(z) = R_{\text{bulk}} + (R_{\text{surf}} - R_{\text{bulk}}) e^{-z/\lambda}$, where R_{bulk} is the bulk value (a function of U only) and $R_{\text{surf}} < R_{\text{bulk}}$. R_{surf} now depends on both U and on U_s , and vanishes only when R_{bulk} does at $U > U_c$. A detailed study by varying U and U_s shows that the surface

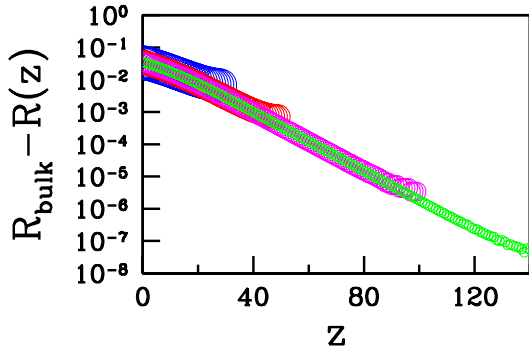


Figure 3: Log scale plot of $R_{\text{bulk}} - R(z)$ versus z for $U = 15.99$, $U_s = 20t$ and for different thicknesses of the slab $N = 60, 100, 200, 400$.

“dead layer” thickness λ depends only on bulk properties and diverges at the Mott transition as $\lambda \propto (U_c - U)^{-\nu}$. Numerically we find $\nu = 0.53 \pm 0.3 \simeq 0.5$, a typical mean field exponent[28]. The same conclusion can actually be drawn by analysing Eqs. (6) and (7) deep inside the bulk. We note that the precise behavior at the outermost surface layers would in a real system depend on details, such as lack of electron-hole symmetry and/or surface dipoles, not included in our model. However, we believe that the exponential behavior and its divergence at a continuous Mott transition should be generic and universal, and thus independent of these and other details. In conclusion, we have shown in a simple approximation the existence in the Hubbard model of strongly correlated metals of a “dead layer” below the crystal surface. Within this layer – whose depth is a bulk property and not a surface property of the metal – the quasiparticle weight decays exponentially on approaching the surface. The dead layer thickness λ inversely depends on the distance in parameter space to the bulk continuous Mott transition, where it diverges critically. The physical significance of λ is that of a correlation length of the bulk metallic state, where the quasiparticle weight acts as an order parameter, critically vanishing at a continuous Mott transition. Like other features of the Hubbard model, this result should we believe carry over to real systems with an ideal Mott transition, not obscured by e.g., symmetry breaking phenomena like magnetic order, provided that the critical region is not preempted by a strong first order jump, like that in the α - γ transition of Ce. It could therefore apply to high temperature V_2O_3 near the paramagnetic metal-insulator weakly first order line, notwithstanding complications including orbital degeneracy, Hund’s rules, and coupling to the lattice (see e.g. Ref.[35] and references therein). We thus expect a surface dead layer in the metal phase of V_2O_3 , with thickness increasing (although not diverging because of the first order transition) on approaching the Mott transition line. The associated paper by Rodolakis *et al.* reports photoemission

evidence which lends some support to this picture. It is also interesting to note that an anomalously thick sub-surface dead layer has long been observed in mixed valent YbInCu_4 [36], with a depth not smaller than 60\AA [37].

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