Tunneling spectroscopy of highly correlated systems

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In the course of tunneling between two highly correlated systems, Coulomb repulsion causes a substantial renormalization of the tunneling matrix element. At half-filling (in the insulating phase), this renormalization leads to a cutoff of the tunneling current, regardless of the sign of the voltage. For an arbitrary filling, a tunneling conductivity exists in the insulating phase only to the extent that the filling differs from half-filling. Experimental implications for STM images are discussed in the particular case of EuX systems with f shells. © *1994 American Institute of Physics.*

Descriptions of scanning tunneling microscopy and spectroscopy start from the Bardeen–Tersoff–Hamann formula, [1,2] which gives us the following expression for the tunneling current between two weakly linked subsystems (the microscope tip and the crystal):

$$I=\frac{2\pi e}{\hbar}\int d\varepsilon|T_{LR}|^2\rho_L(\varepsilon)\rho_R(\varepsilon)[f_L(\varepsilon)-f_R(\varepsilon)]. \tag{1}$$

Here T_{LR} is the tunneling matrix element [by convention, between the left system (L) and the right one (R)], $\rho_{L,R}(\varepsilon)$ are the local densities of states for the systems, without allowace for the interaction between them, and $f_{L,R}$ are Fermi distribution functions with chemical potentials $\mu_L-\mu_R=eV$, where V is the applied voltage.

We show below that, if the tunneling occurs between systems in which there is a strong electron–electron interaction (in one system or both), there is a substantial renormalization of the tunneling matrix element (in addition to the change in the density of states from the one-particle value when the interaction is taken into account). This effect may lead to a cutoff of the tunneling current.

To describe the tunneling it is convenient to use the strong-coupling method. We take the electron–electron interaction into account by the "slave-boson" approach.

We describe the spectrum in the isolated L and R systems by means of the Hubbard Hamiltonian[3]

$$\hat{H}=\sum_{i\sigma}\varepsilon_{i\sigma}\hat{f}^{+}_{i\sigma}\hat{f}_{i\sigma}+\sum_{ij\sigma}t_{ij}\hat{f}^{+}_{i\sigma}\hat{f}_{j\sigma}+U\sum_{i}\hat{n}_{i\sigma}\hat{n}_{i-\sigma}. \tag{2}$$

To keep the equations simple, we omit the subscripts L and R wherever we can do so without causing any confusion. The operators $\hat{f}^+_{i\sigma}$ create electrons at sites, $\varepsilon_{i\sigma}$ is the site energy, t_{ij} is the integral for a jump within the L or R system, and U is the energy of the Coulomb repulsion at a site.

The tunneling coupling of the systems is described by the Hamiltonian

$$T=\sum_{ij\sigma} (T_{LRij}\hat{f}^+_{i\sigma}\hat{f}_{j\sigma}+\text{H.a.}), \tag{3}$$

where T_{LRij} is the tunneling matrix element between the L and R systems, the subscript i refers to the sites of L, and the subscript j refers to the sites of R.

The operator representing the tunneling current is written in the standard form:

$$\hat{I}=\frac{ie}{\hbar}\sum_{ij\sigma} (T_{LRij}\hat{f}^+_{i\sigma}\hat{f}_{j\sigma}-\text{H.a.}). \tag{4}$$

To take the interaction into account, it is convenient to use a version of the slave-boson method proposed in Ref. 4. In this method, some slave (auxiliary) boson fields are introduced in order to describe four atomic states of a site. The physical states at the sites, $|\sigma i\rangle$, are found by applying boson operators and new fermion operators to the vacuum states $|\text{vac}\rangle$. The physical states are found in the following way:

(1) states of a vacant site, $|\sigma i\rangle=\hat{e}^+_i|\text{vac}\rangle$,
(2) states of a singly filled site, $|\sigma i\rangle=\Sigma_{\sigma'}\hat{c}^+_{i\sigma'}\hat{p}^+_{i\sigma'\sigma}|\text{vac}\rangle$,
(3) states of a doubly filled site, $|\uparrow\downarrow i\rangle=\hat{c}^+_{i\uparrow}\hat{c}^+_{i\downarrow}\hat{d}^+_i|\text{vac}\rangle$.

With an eye on subsequent applications to spin-dependent tunneling, we will use a rotationally invariant formulation of the slave-boson method.[5]

The boson operators $\hat{e}^+$, $\hat{p}^+$, and $\hat{d}^+$ obey commutation relations, and the new fermion operators $\hat{c}^+$ obey anticommutation relations. To eliminate extraneous states and to preserve the equivalence of the Hamiltonian in the new representation and the original Hamiltonian, we introduce some auxiliary constraints on the boson and fermion operators:[5]

$$\hat{Q}=\hat{e}^+_i\hat{e}_i+\text{tr}(\hat{p}^+_i\hat{p}_i)+\hat{d}^+_i\hat{d}_i, \tag{5}$$

$$\text{tr}(\hat{\tau}_\mu\hat{p}^+_i\hat{p}_i)+2\delta_{\mu,0}\hat{d}^+_i\hat{d}_i=\sum_{\sigma\sigma'}\hat{c}^+_{i\sigma}\hat{\tau}^{\sigma\sigma'}_\mu\hat{c}_{i\sigma'},$$

where $\hat{\tau}_\mu$ are 2×2 basis matrices [the unit matrix ($\mu=0$) and three Pauli matrices, $\mu=1,2,3$].

In the slave-boson representation, the initial Hamiltonians become quadratic in the new Fermion operators:

$$\hat{H}=\sum_{i\sigma}\varepsilon_{i\sigma}\hat{c}^+_{i\sigma}\hat{c}_{i\sigma}+\sum_{ij\sigma\sigma'\sigma_1}t_{ij}(\hat{c}^+_{i\sigma}\hat{Z}^+_{i\sigma\sigma_1})(\hat{Z}_{j\sigma_1\sigma'}\hat{c}_{j\sigma'})+U\sum_i\hat{d}^+_i\hat{d}_i. \tag{6}$$

The tunneling-current operator becomes

$$\hat{I}=\frac{ie}{\hbar}\sum_{ij\sigma\sigma_1\sigma'}[T_{LRij}(\hat{c}^+_{i\sigma}\hat{Z}^+_{Li\sigma\sigma_1})(\hat{Z}_{Rj\sigma_1\sigma'}\hat{c}_{j\sigma'})-\text{H.a.}]. \tag{7}$$

The operators $\hat{Z}_{i,j\sigma_1\sigma}$ are introduced to preserve the equivalence of the operators in the new representation. They are given by[4,5]

$$\hat{Z}_i=[(1-\hat{d}^+_i\hat{d}_i)\hat{\tau}_0-\hat{p}^+_i\hat{p}_i]^{-1/2}(\hat{e}^+_i\hat{p}_i+\tilde{\hat{p}}^+_i\hat{d}_i)[(1-\hat{e}^+_i\hat{e}_i)\hat{\tau}_0-\hat{p}^+_i\hat{p}_i]^{-1/2}. \tag{8}$$

The numerator describes conservation of the number of bosons in the course of jumps from one site to another and is determined unambiguously (it does not contain any arbitrariness operator). The first term corresponds to the process (singly occupied site) $\rightarrow$(empty site), and the second to the process (doubly occupied site) $\rightarrow$(singly occupied site). The normalization factors contain an arbitrariness operator and are defined in such a way that the slave-boson method yields the exact result in the free-electron approximation ($U=0$) and for isolated centers (the atomic limit: $t_{ij}=0$, with U arbitrary).

The tunneling current is calculated as the expectation value of current operator (7). The physical expectation values are given by

$$\langle ...\rangle=\int[D\hat{c}^+][D\hat{c}][D\hat{p}_\mu][D\hat{e}][D\hat{d}][D\lambda^{(1)}][D^{(2)}_\mu], \tag{9}$$

$$(...)\exp\left\{i\int_K dt[L_L(t)+L_R(t)-H_L(t)-H_R(t)-T(t)]\right\}.$$

For brevity we have introduced $[D\hat{c}^+]\equiv[D\hat{c}^+_L][D\hat{c}^+_R]$, etc. The quantities $\lambda^{(1)}$ and $\lambda^{(2)}_\mu$ are Lagrange multipliers, which incorporate constraints (5) (see Refs. 4 and 5 for details). The quantities $\hat{L}_{L,R}(t)$ are the Lagrangians of the noninteracting L and R systems, given by

$$\hat{L}_{L,R}(t)=\sum_{i\sigma\sigma'}\hat{c}^+_{L,Ri\sigma}(t)\left\{\left[i\frac{\partial}{\partial t}-\mu_{L,R}+\lambda^{(2)}_{i0}\right]\delta_{\sigma\sigma'}+\lambda^{(2)}_i\hat{\tau}^{\sigma\sigma'}_\mu\right\}\hat{c}_{L,Ri\sigma'}(t). \tag{10}$$

We assume that the applied voltage is incorporated in the chemical potentials $\mu_{L,R}$; we therefore have $\mu_L-\mu_R=eV$. The integration in the exponential function is carried out over a closed time contour:[6–9]

$$\int_K dt=\int_{-\infty}^{+\infty}dt+\int_{+\infty}^{-\infty}dt=\eta_\alpha\int_{-\infty}^{+\infty}dt_\alpha,$$

where $\alpha=1,2$; $\eta_1=1$; and $\eta_2=-1$.

Expanding the exponential function in the operator representing the tunneling between systems, and retaining up to the first order, we find the following expression for the tunneling current:

$$I=\frac{e}{\hbar}\sum_{\substack{ij\sigma\sigma_1\sigma'\\ i'j'\sigma_2\sigma_3\sigma_4}}\int ds T_{LRij}T_{RLj'i'}[G^{+-}_{\substack{Lii'\\ \sigma\sigma_1\\ \sigma_3\sigma_4}}(t,s)G^{-+}_{\substack{Rjj'\\ \sigma_1\sigma'\\ \sigma_2\sigma_3}}(s,t)-\text{H.a.}. \tag{11}$$

The superscripts on the Green's functions refer to the time contours; the Green's functions themselves are given by

$$\overset{+-}{G}_{\substack{Lii' \\ \sigma\sigma_1 \\ \sigma_3\sigma_4}}(t,s) = -\langle \hat{c}^+_{Li\sigma}(t)\hat{Z}^+_{Li\sigma\sigma_1}(t)\hat{Z}_{Li'\sigma_3\sigma_4}(s)\hat{c}_{Li'\sigma_4}(s)\rangle,$$

$$\overset{-+}{G}_{\substack{Rjj' \\ \sigma_1\sigma' \\ \sigma_2\sigma_3}}(s,t) = \langle \hat{Z}_{Rj\sigma_1\sigma}(t)\hat{c}_{Rj\sigma}(t)\hat{c}^+_{Rj'\sigma_2}(s)\hat{Z}^+_{Rj'\sigma_2\sigma_3}(s)\rangle. \tag{12}$$

Expression (11) can be written in the more compact form

$$I = \frac{e}{\hbar}\int ds \ \mathrm{tr}\{\hat{T}_{RL}\hat{G}^{+-}_L(t,s)\hat{T}_{LR}\hat{G}^{-+}_R(s,t) - \mathrm{H.a.}\}, \tag{13}$$

where tr means a summation over all indices. In the saddle-point approximation in the boson variables, the Fourier transforms of the Green's functions can be written

$$\overset{\pm\mp}{G}_{\substack{Lii' \\ \sigma\sigma_1 \\ \sigma_3\sigma_4}}(\varepsilon) = 2\pi i \rho_{\substack{Lii' \\ \sigma\sigma_4}}(\varepsilon)(Z^+_{Li\sigma\sigma_1}Z_{Li'\sigma_3\sigma_4})\begin{cases} f_l(\varepsilon), \\ f_l(\varepsilon)-1, \end{cases}$$

$$\rho_{\substack{Lii' \\ \sigma\sigma_4}}(\varepsilon) = \frac{1}{\pi}\,\mathrm{Im}\{\overset{R}{G}_{\substack{Lii' \\ \sigma\sigma_4}}(\varepsilon)\}. \tag{14}$$

The density matrix $\hat{\rho}$ and the retarded Green's function $\hat{G}_R$ are found from the solution of the effective one-particle Hamiltonian (6) in which the boson operators are replaced by their expectation values at the saddle point. The retarded Green's function $\hat{G}_R$ contains only new fermion operators. The final expression for the tunneling current is

$$I = \frac{e}{\hbar}\int d\varepsilon \ \mathrm{tr}\{\tilde{T}_{RL}\hat{\rho}_L(\varepsilon)\tilde{T}_{LR}\hat{\rho}_R(\varepsilon)\}[f_L(\varepsilon)(\varepsilon)], \tag{15}$$

where

$$\tilde{T}_{\substack{LRij \\ \sigma\sigma'}} = \sum_{\sigma_1} Z^+_{Li\sigma\sigma_1}T_{LRij}Z_{Rj\sigma_1\sigma'}.$$

Expression (15) is of the same form as in the one-particle problem, (1) (see also Ref. 10), but the original tunneling matrix elements are now replaced by their renormalized values. The densities of states also depend on the boson variables through the spectrum.

Let us consider tunneling between a metal, in which there is no interaction (this is the R system), anda half-filled metal (the L system), in which the electron–electron interaction is strong. In this case we have $Z_R = 1$, and Z_L becomes[4]

$$Z_L = \frac{(ep_\sigma + dp_{-\sigma})}{[(p^2_\sigma + e^2)(p^2_{-\sigma} + d^2)]^{1/2}}, \quad Z_L = 8d^2(1-2d^2).$$

Near the surface the values of Z_L are slightly different from their values in the interior for a homogeneous system, but this circumstance is unimportant to a qualitative derivation.

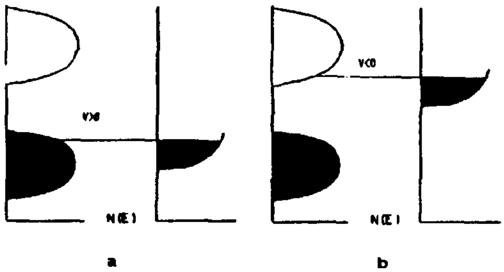

FIG. 1.

At half-filling, at a certain critical value of the interaction[4,11] $U=U_c$ (U_c is on the order of the width of the band in the original metallic phase; for a cubic lattice we would have $U_c=8t$), there is a metal–insulator phase transition. Near this transition, on the side of the metal phase, the following relation holds:[11]

$$d^2=\frac{1}{4}\left(1-\frac{U}{U_c}\right), \quad Z_L=1-\left(\frac{U}{U_c}\right)^2.$$

The tunneling current vanishes at the point of the transition and remains zero in the insulating phase (at half–filling), regardless of the sign of the voltage. In the insulating phase ($U>U_c$) the factors which renormalize the tunneling matrix element become[11]

$$Z_L=2\delta\sqrt{\frac{U/U_c}{U/U_c-1}},$$

where δ is the deviation from half-filling. A tunneling conductivity exists in the insulating phase only to the extent that the filling deviates from one-half. In the case of an infinitely strong repulsion, the probability for a double occupation is zero (as it is for empty sites). When the filling deviates from one-half, however, the width of the band (amd, correspondingly, the magnitude of the tunneling matrix element) is nonzero and has a value on the order of $t\cdot\delta$. The conductivity exists because of either holes ($\delta<0$) or electrons ($\delta>0$).

Tunneling into a Hubbard insulator is fundamentally different from tunneling into an ordinary band insulator. In the latter case, with a positive voltage on the tip (Fig. 1a), there is a tunneling from the filled valence band into vacant states above the Fermi level in the tip. When there is a negative voltage on the tip (Fig. 1b), electrons tunnel from under the Fermi level in the tip into vacant states in the conduction band.

Formally, the tunneling into the Hubbard insulator looks the same as it would if "valence band" and "conduction band" were understood to be a half-filled original band split by the Coulomb repulsion. This simple picture, however, ignores the renormalization of the tunneling matrix element, which essentially reflects the following circumstance. The "new one-particle fermions" (described by the operators $\hat{c}_{i\sigma}$) are good quasiparticles (in the slave-boson method). A real particle (a fermion, described by the operators $\hat{f}_{i\sigma}$), which is a combination of a slave boson and a one-particle fermion, undergoes tunneling. In the course of the tunneling, boson degrees of freedom should tunnel along with the "one-particle fermion." This circumstance is, in fact, taken into account in the renormalized tunneling matrix element.

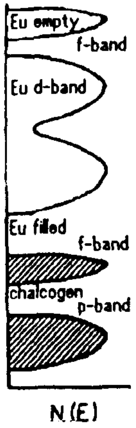

FIG. 2.

At a qualitative level, the reason for the cutoff of the tunneling current is as follows: For tunneling into an empty Hubbard band it is necessary to create a doubly occupied site; the probability for that process is small. In the case of tunneling from a filled band in a Hubbard insulator, it is necessary to create an empty site [since, as can be seen from (12) and (13), the expectation values are calculated in terms of the equilibrium state of the two noninteracting L and R systems]. The creation of an empty site automatically implies the creation of a doubly occupied site somewhere in the system. The tunneling current is accordingly blocked even in the case of tunneling out of a filled band. In the case of a filling other than half-filling, the tunneling conductivity is nonzero only to the extent that there is a deviation from exact half-filling.

If we were to go beyond the saddle-point approximation in the boson degrees of freedom (if we were to take fluctuations into account), we would apparently find a nonzero conductivity commensurate in size with these fluctuations even in the case of exact half-filling. There are some convenient entities for an experimental test: the semiconductors EuX (X=O, S, Se, Te), which have the band structure shown qualitatively in Fig. 2. The valence band is formed from p states of the chalcogen, and the empty conduction band from d states of the Eu. In the band gap there is a half-filled Hubbard band, formed by f states of Eu (Ref. 12). The Eu atoms can be thought of as a Kondo lattice in a medium of chalcogen atoms [in this case we have $Z_L \simeq \exp(-U/t)$; Ref. 13]. If the tip of the tunneling microscope is positioned over chalcogen atoms, a tunneling current and an image of the atoms should be observed when there is a positive voltage on the tip. If the tip is instead positioned over Eu atoms, there will be no tunneling current and no image of the Eu atoms, regardless of whether the voltage is positive or negative. Accordingly, the contrast of the scanning-tunneling-microscope images, which is characteristic of one-particle tunneling [as in the case of, for example, the GaAs(110) surface], should not exist in this case.

This result can be explained in the following way. If there is a strong Hubbard repulsion near half-filling, it is convenient to rewrite the Hamiltonian by projecting the action of the operators onto states without doubly occupied sites. This approach has been used to describe an RVB state (by Baskaran, Zou, and Anderson[14]). The terms in the

Hamiltonian which describe jumps $(t_{ij}\hat{f}^{+}_{i\sigma}\hat{f}_{j\sigma})$ transform to $t_{ij}(1-\hat{n}_{i-\sigma})\hat{f}^{+}_{i\sigma}\hat{f}_{j\sigma}$ $(1-\hat{n}_{j-\sigma})$. At exact half-filling, an averaging yields an effective jump integral $(t^{\mathrm{eff}}_{ij}=t_{ij}\delta)$, in agreement with our result.

Anderson and Zou's calculation[15] of the tunneling conductivity for an RVB state shows that this conductivity is proportional to the applied voltage (the tunneling current is proportional to the square of the voltage) and exists because of an inelastic decay of quasiparticles (holons and spinons) into the allowed phase volume in the course of the tunneling (this volume is proportional to the voltage). Accordingly, the tunneling conductivity in the limit $V\rightarrow 0$, which is usually understood as $\sigma=dI/dV|_{V\rightarrow 0}$, approaches zero; i.e., the current is cut off.

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