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Interaction and doping dependence of optical spectral weight of the two-dimensional Hubbard model

A. J. Millis and S. N. Coppersmith
AT&T Bell Laboratories, Murray Hill, New Jersey 07974
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We calculate the total and low-frequency spectral weights of the optical conductivity of the two-dimensional Hubbard model for various values of the interaction strength and carrier concentration. In some limits we obtain exact results and bounds. For general parameter values we obtain estimates using the variational Monte Carlo technique. We show our results disagree with those obtained by exact diagonalization of small systems, and argue that the latter are less accurate because of finite-size effects.

The optical conductivity of strongly correlated electron systems is of current experimental interest because measurements on high-T_c superconductors[1,2] and on quasi-one-dimensional organic conductors[3] show large deviations from the predictions of band theory. The optical conductivity is also of fundamental theoretical interest because the spectral weight at low frequencies is the natural order parameter for the Mott transition.[4] The optical conductivity of model systems has been studied by approximate mean-field calculations,[5-7] by analysis of integrable one-dimensional (1D) models,[8-10] by exact diagonalization of small systems,[11-15] and by quantum Monte Carlo techniques.[16] The uncertain quantitative applicability of the analytic mean-field calculations, the size limitations of the exact diagonalization and Monte Carlo results and the possibility that the integrable 1D models do not exhibit generic behavior lead us to consider other methods for obtaining information about the optical conductivity.

Here we study the optical conductivity of the Hubbard model by exploiting sum rules,[4,17] which relate the spectral weight in various frequency regimes of the conductivity to ground-state properties. We obtain some exact results and variational bounds, most of which apply in any dimension. For a range of parameters we obtain estimates of the total spectral weight of the $d=2$ Hubbard model from variational wave functions. Information concerning the division of spectral weight between high and low frequencies is more difficult to obtain, but we derive some results from the mapping, believed valid at large U, of the Hubbard model onto the t-J model.[4,18,19]

We study the Hubbard model at zero temperature using simple modifications of arguments introduced by Kohn.[4] Kohn considered electrons interacting via the Coulomb force and moving in a periodic potential. We consider fermions hopping with matrix element t between nearest-neighbor sites of a d-dimensional hypercubic lattice with unit lattice constant and subject to a repulsive interaction U when two fermions occupy the same site. We assume periodic boundary conditions in the x direction and that the system is subject to a flux $\Phi(t)$, which we represent by a vector potential $\mathbf{A}(t)=[\Phi(t)/N_x]\hat{\mathbf{x}}$, where N_x is the number of sites in the x direction. The Hamiltonian may be written

$$H=-\sum_{\mathbf{n},\boldsymbol{\tau},\sigma} t(e^{i\mathbf{A}(t)\cdot\boldsymbol{\tau}}c^{\dagger}_{\mathbf{n}\sigma}c_{\mathbf{n}+\boldsymbol{\tau}\sigma}+\text{H.c.})+U\hat{D}\,. \tag{1}$$

Here $\mathbf{n}$, a vector of integers, labels a lattice site, $\boldsymbol{\tau}$ is a vector connecting site $\mathbf{n}$ to a nearest neighbor, and H.c. means Hermitian conjugate. The density operator at site $\mathbf{n}$ for fermions of spin σ, $\hat{\rho}_{\mathbf{n}\sigma}=c^{\dagger}_{\mathbf{n}\sigma}c_{\mathbf{n}\sigma}$. The doubly-occupied-sites operator $\hat{D}=\sum_{\mathbf{n}}\hat{\rho}_{\mathbf{n}\uparrow}\hat{\rho}_{\mathbf{n}\downarrow}$. When no explicit time argument is given, A and Φ are assumed to be static. The carrier concentration $\rho=\sum_{\sigma}\langle\rho_{\mathbf{n}\sigma}\rangle$, and the doping $\delta=1-\rho$.

The complex optical conductivity $\tilde{\sigma}(\omega)$ gives the linear response of the system to a uniform time-dependent electric field generated by a time-dependent flux via Eq. (1). Standard linear-response arguments applied to Eq. (1) yield a Kubo formula for $\tilde{\sigma}(\omega)$:

$$\tilde{\sigma}(\omega)=-\frac{\langle\hat{T}_x\rangle}{i\omega}-\frac{1}{i\omega}\sum_{n\neq 0}\frac{|\langle n|\hat{J}_{px}|0\rangle|^2}{\omega-(E_n-E_0)+i0^+}\,. \tag{2}$$

Here

$$\hat{T}_x=-t\sum_{\mathbf{n},\sigma}c^{\dagger}_{\mathbf{n}\sigma}c_{\mathbf{n}+\hat{\mathbf{x}}\sigma}+(\text{H.c.})\,,$$

the paramagnetic current operator $\hat{J}_{px}=\delta\hat{H}/\delta\mathbf{A}|_{\mathbf{A}=0}$ is given by

$$\hat{J}_{px}=-it\sum_{\mathbf{n},\sigma}(c^{\dagger}_{\mathbf{n}\sigma}c_{\mathbf{n}+\hat{\mathbf{x}}\sigma}-\text{H.c.})\,,$$

and the eigenfunction and energy of the nth eigenstate are $|n\rangle$ and E_n. In deriving Eq. (2) we have first let $\Phi(t)\to 0$ and then taken $N_x\to\infty$, and we have assumed zero temperature.

We write the real part of $\tilde{\sigma}(\omega)$ as $\sigma(\omega)$. For a system with at least a discrete translation invariance (such as the Hubbard model) at $T=0$ one expects on general grounds[4] $\sigma(\omega)=(\omega_p^{*2}/4)\delta(\omega)+\sigma_{\text{reg}}(\omega)$, where $\lim_{\omega\to 0}\omega\sigma_{\text{reg}}(\omega)=0$. The system is metallic if $\omega_p^{*2}>0$ and is insulating if $\omega_p^{*2}=0$.[20] In the latter case, $\lim_{\omega\to 0}\sigma_{\text{reg}}(\omega)=0$. These definitions must be modified for disordered systems or nonzero temperatures. ω_p^{*2} is thus an order parameter[4] for the Mott metal-insulator transition which may occur in a nondisordered system as interaction strength U and carrier concentration are varied.

Another useful way to characterize the optical conductivity is the total oscillator strength or spectral weight $\omega_p^2/8 \equiv \int_0^\infty \sigma(\omega)d\omega$. The spectral weight is conventionally written as a plasma frequency because in a free-electron gas the quantity ω_p inferred from the oscillator strength is identical to the frequency of the long-wavelength plasmon oscillation. In a strongly correlated system the two quantities may be different;[21] in this paper we consider only spectra weights, not the frequency of any plasmon mode, but we retain the convention of writing oscillator strength as $\omega_p^2/8$.

Both ω_p^2 and ω_p^{*2} are nontrivial functions of U and the doping δ, and are also experimentally measurable. They are related to ground-state properties by two identities due to Kohn.[4] To derive an identity for ω_p^2 we note from Eq. (2) that $\lim_{\omega\to\infty}\tilde{\sigma}(\omega) = -\langle\hat{T}_x\rangle/i\omega$ and we use the Kramers-Kronig relation linking the real and imaginary parts of $\tilde{\sigma}$. The result is

$$\int_0^\infty d\omega\sigma(\omega) \equiv \frac{\omega_p^2}{8} = -\frac{\pi}{2}\langle\hat{T}_x\rangle . \tag{3}$$

Equation (3) is the f-sum rule for the Hubbard model with periodic boundary conditions. We believe that Eqs. (1)–(3) provide the most natural resolution of previously discussed ambiguities[11,12,14,15] in the applicability of the f-sum rule to finite systems with periodic boundary conditions. We have explicitly verified (3) by exact diagonalization of three and four site rings.

To derive an identity for ω_p^{*2} we use Eq. (3) to obtain an expression for $\lim_{\omega\to 0}\tilde{\sigma}(\omega)$ and again exploit the Kramers-Kronig relation. To obtain a second identity for ω_p^{*2} we use standard perturbation theory to obtain the second derivative of the ground-state energy with respect to a change in Φ. The result is

$$\frac{\omega_p^{*2}}{4} = \pi N_x^2 \frac{d^2E_0}{d\Phi^2} = -\pi\langle\hat{T}_x\rangle - 2\pi\sum_{n\neq 0}\frac{|\langle n|\hat{J}_{px}|0\rangle|^2}{(E_n - E_0)} . \tag{4}$$

The plasma frequency ω_{p0} of the noninteracting model [Eq. (1) with $U=0$] is easily found to be $\omega_{p0}^2 = -4\pi E_0/d$ ($U=0$). Because $[\hat{J}_{px},H]=0$ if $U=0$, and because in the thermodynamic limit the ground state has no current, $\omega_{p0}^{*2} = \omega_{p0}^2 - O(1/N_x)$. As U is increased at fixed δ, $|\langle\hat{T}_x\rangle|$ must decrease, and for $U\neq 0, [\hat{J}_{px},H]\neq 0$, so that the sum in Eq. (3) becomes nonzero. Thus[21]

$$O \leq \omega_p^{*2}[U] \leq \omega_p^2[U] \leq \omega_{p0}^2 . \tag{5}$$

Further exact results may be obtained in several limits. The ground-state energy is known[22] to order U^2, and the Hellman-Feynman theorem $\langle T\rangle = E_0 - U\partial E_0/\partial U$. For the 2D square lattice and $\delta < 0.2$ one obtains

$$\frac{\omega_p^2}{\omega_{p0}^2} = 1 - 7.5\times 10^{-3}(U/t)^2 + O(U/t)^4 . \tag{6}$$

A calculation of ω_p^{*2} to $O(U^2)$ is in progress; results will be presented elsewhere.[23]

For a half-filled band ($\delta=0$) and $U\to\infty$, the Hubbard model maps onto the Heisenberg model. This is an insulator, so $\omega_p^{*2}=0$. From the Hellman-Feynman theorem, $\langle\hat{T}\rangle = t(dE_0/dt)$, where E_0 is the ground-state energy. For the 2D square lattice, the known result for E_0 (Ref. 24) yields

$$\lim_{U\to\infty}\frac{\omega_p^2}{\omega_{p0}^2} = \frac{5.8t}{U} . \tag{7}$$

Another simple limit is $U=\infty$, $\delta\to 0$. At $\delta=0$ and $U=\infty$ the material is an insulator, with $\omega_p^* = \omega_p = 0$. If one hole is added, the ground state is a fully polarized ferromagnet.[25] This ground state optimizes the kinetic-energy operator; thus, $\omega_p^{*2} = \omega_p^2 = 8\pi t/dN$ (N is the number of sites). The ground state is not known for nonzero δ; however, (i) additional holes may only impede each others motion, so that the kinetic energy of δ holes is less than or equal in magnitude to δ times the kinetic energy of one hole and (ii) the energy calculated by assuming that the ground state remains a fully polarized ferromagnet in an upper bound on the true ground-state energy. Thus, at $U=\infty$ as $\delta\to 0$

$$\frac{8\pi t\delta}{d} > \omega_p^2 \geq \frac{8\pi t\delta}{d}[1 - c_d\delta] , \tag{8}$$

where d is the dimension and c_d is a dimensionality-dependent constant. For example, when $d=2$, $c_d = 2\pi$. Therefore ω_p^2 (and so ω_p^{*2}) vanishes at least as fast as δ at $U=\infty$.

For other values of U and δ, in $d>1$, little is known about ω_p^2 and $\omega^{p^{*2}}$. Numerical studies of small (10 site or—for small U—16 site lattices) have been performed.[11–16] However, the small system sizes and the lack of theoretical analysis of finite-size corrections make interpretation of the results uncertain. To obtain further information we have used two approximate methods, which we discuss in turn. In each method, we must evaluate expectation values of operators using a given wave function; we do this by the variational Monte Carlo (VMC) method.[26]

In one approach we use the Hubbard Hamiltonian, Eq. (1), and the Gutzwiller wave function $|\psi_g\rangle$. This is obtained by taking $|\psi_M\rangle$, the M-particle wave function which minimizes the kinetic-energy term in Eq. (1), and explicitly projecting out some fraction of doubly occupied sites:

$$|\psi_g\rangle = g^{\hat{D}}|\psi_M\rangle . \tag{9}$$

$0 \leq g \leq 1$ is a variational parameter, chosen by minimizing the expectation value of H in the class of states $|\psi_g\rangle$. One obtains for the ground-state energy E a variational estimate $E_{\rm var}$ of the form $E \leq E_{\rm var} \equiv T(g) + UD(g)$, with g chosen so that $dE_{\rm var}/dg = dT(g)/dg + UdD(g)/dg = 0$. Here $T(g)$ and $D(g)$ are the expectation values of the kinetic-energy operator and of $\hat{D}$ in the state $|\psi_g\rangle$. Because $T(g)<0$ and $UD(g)>0$, the $T(g)$ so obtained gives, in general, an estimate for, rather than a bound on, the total oscillator strength ω_p^2. However, at $U=\infty$, $g=D(g)=0$, so $\omega_p^2 > (4\pi/d)T(g=0)$. The bound given in Eq. (8) is, however, better. One might naively attempt to estimate ω_p^{*2} from Eq. (4) by using the variational wave function to compute $E(\Phi)$; however, we have shown elsewhere[27] that for the Gutzwiller wave function, $\omega_p^{*2} = \omega_p^2$.

We have also computed ω_p^2 and ω_p^{*2} by using a canonical transformation due originally to Kohn[4] to define a new

Hamiltonian $\tilde{H}=e^{iS}He^{-iS}$ with $[\tilde{H},\hat{D}]=0$. An expression for $\tilde{H}$, valid to leading order in t/U and δ, has been given for $\Phi=0$.[18,19] This leading order expression for $\tilde{H}$ is often referred to as the t-J model. The generalization to the case of a nonzero static flux is straightforward: we rewrite Eq. (1) in terms of operators $\tilde{c}^{\dagger}_{\mathbf{n}\sigma}=e^{-i\mathbf{A}\cdot\mathbf{n}}c^{\dagger}_{\mathbf{n}\sigma}$, apply the procedure of Refs. 18 and 19 obtaining a result written in terms of the $\tilde{c}^{\dagger}$, and then undo the transformation. The result is

$$\tilde{H}=-\sum_{\mathbf{n},\tau,\sigma} te^{i\mathbf{A}\cdot\tau}P_d c^{\dagger}_{\mathbf{n}\sigma}c_{\mathbf{n}+\tau\sigma}P_d+\frac{2t^2N}{U}+\frac{4t^2}{U}\sum_{\mathbf{n},\tau}\mathbf{S}_{\mathbf{n}}\cdot\mathbf{S}_{\mathbf{n}+\tau}+\cdots . \tag{10}$$

The projection operators P_d annihilate states for which $\hat{D}|\psi\rangle\neq 0$, and $\mathbf{S}_{\mathbf{n}}=\sum_{\alpha,\beta}c^{\dagger}_{\mathbf{n}\alpha}\boldsymbol{\sigma}_{\alpha\beta}c_{\mathbf{n}\beta}$.

Because the mapping from H to $\tilde{H}$ is a canonical transformation, the ground-state energies E_0 of the two models are the same. Thus using Eqs. (3) and (10) and the relation $\langle T\rangle=t\,dE_0/dt$, we obtain for dimension $d=2$, to leading order in t/U and δ,

$$\omega_p^2=4\pi\left[-\langle\tilde{T}_x\rangle+\frac{4t^2}{U}-\frac{16t^2}{U}\langle\mathbf{S}_{\mathbf{n}}\cdot\mathbf{S}_{\mathbf{n}+\hat{\mathbf{x}}}\rangle\right]. \tag{11}$$

Here

$$\tilde{T}_x=-t\sum_{\mathbf{n},\sigma}P_d c^{\dagger}_{\mathbf{n}\sigma}c_{\mathbf{n}+\hat{\mathbf{x}}\sigma}P_d$$

and the expectation value is to be taken in the ground state of Eq. (10).

Similarly, beginning from Eq. (10) and following the steps that led to Eqs. (4) and (5) yields

$$\omega_p^{*2}<-4\pi\langle\tilde{T}_x\rangle<\omega_p^2 . \tag{12}$$

The right-hand side inequality follows from Eq. (11) and the result $\langle\mathbf{S}_{\mathbf{n}}\cdot\mathbf{S}_{\mathbf{n}+\hat{\mathbf{x}}}\rangle<0$. Note that the left-hand side inequality in Eq. (12) is valid only if (i) higher-order terms in t/U and δ can be neglected, and (ii) if the exact ground-state wave function is used.

In previous work[19] it has been argued that the ground state of $\tilde{H}$ [Eq. (10)] is well approximated by the particular Gutzwiller wave function $|\psi_{g=0}\rangle$. We have used this wave function to evaluate the expectation values in Eqs. (11) and (12).

In Fig. 1 we have presented results obtained for $\omega_p^2(U,\delta)$ from VMC calculations described above, from Eq. (6), and from exact diagonalization of small systems.[11,14] For $U=4$ (not shown) VMC, exact diagonalization, quantum Monte Carlo, and perturbation theory to $O(U^2)$ agree that $\omega_p^2\approx 0.9\omega_{p0}^2$ approximately independent of doping for $\delta\lesssim 0.2$. For larger U, the agreement between the various methods is not impressive. In general the VMC results show lower magnitude and more doping dependence of the total oscillator strength than do the diagonalizations. Comparison with the result of Eq. (6) leads us to suspect that the calculation based on Eqs. (11) and (12) and $|\psi_{g=0}\rangle$ is more reliable than the calculation based on Eq. (1) and $|\psi_g\rangle$ calculation at large U and small δ, in agreement with previous results.[19] The disagreement between the different diagonalizations and the disagreement of both with the prediction of Eq. (6) (at $U=20$) suggests that the diagonalizations have not been done for large enough systems. However, on a qualitative level, all calculations agree that to have a substantial ($\gtrsim 50\%$) reduction of total oscillator strength from the band theory value requires a large U ($U\gtrsim 10$) and that a noticeable doping dependence of ω_p requires a very large U, say $U\gtrsim 20$. Although the applicability of the one-band Hubbard model to high-T_c superconductors is not clear, it is

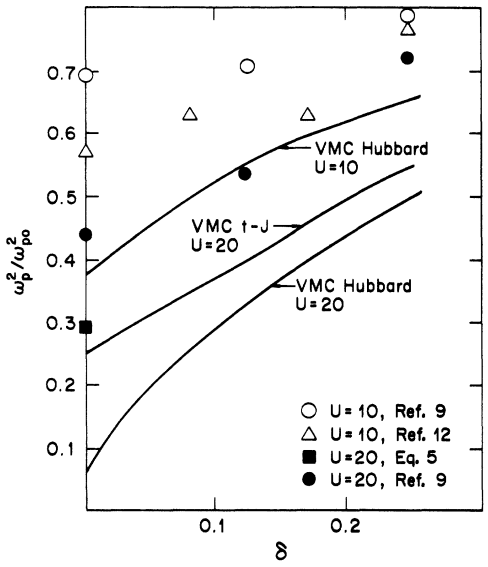

FIG. 1. Doping (δ) dependence of total oscillator strength ω_p^2 normalized to noninteracting, zero doping value ω_{p0}^2. Solid lines: results of VMC calculations. Other symbols: results of cluster calculations and mapping to Heisenberg model [Eq. (5)], as indicated.

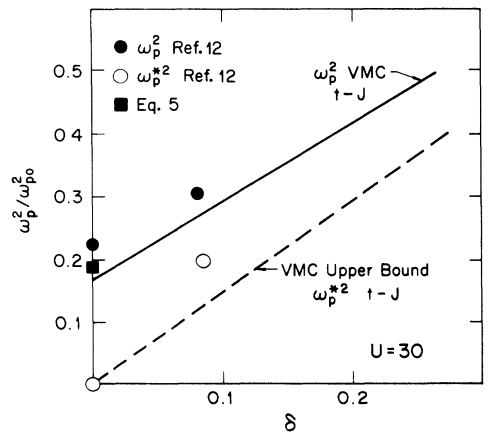

FIG. 2. Doping (δ) dependence of low frequency (ω_p^{*2}) and total (ω_p^2) oscillator strength, both normalized to noninteracting value ω_{p0}^2. Dashed line: VMC estimate for t-J model upper bound on ω_p^{*2} from Eq. (11). This estimate is about 15% larger than the bound obtained from the Nagaoka state [Eq. (6)] which is $\omega_p^{*2}/\omega_{p0}^2=1.23\delta$. Solid line: VMC estimate for ω_p^2 for t-J model, $U=30$. Other symbols: results of cluster calculations and mapping to Heinsenberg model [Eq. (5)], as indicated.

interesting that both a low value and a strong doping dependence of optical spectral weight may have been observed in reflectivity experiments on high-T_c superconductors.[1,2]

The VMC estimate for $-\langle \tilde{T}_x \rangle$ is plotted against doping in Fig. 2, and is compared with the estimate from Eq. (11) for the total oscillator strength for $U=20$ and 30 and with results from exact diagonalization. The doping dependence of the total oscillator strength [obtained from Eq. (11)] is very similar to that of the bound on the low-frequency oscillator strength [from Eq. (12)] suggesting that the principal effect of doping is to add oscillator strength, presumably primarily at low frequencies. A different result, the transfer of a large amount of spectral weight from high to low frequencies with doping, was found in a calculation[14] of $\sigma(\omega)$ for a 10-site cluster and $U=10$ or 30. The VMC estimate for $-\langle \tilde{T}_x \rangle$ is about 15% larger than the value of ω_p^{*2} obtained in the Nagaoka state.

In this paper we have used exact arguments and variational wave functions to obtain information about the magnitude and doping dependence of the total and low-frequency spectral weight in the optical conductivity of the Hubbard model, especially for strong correlation and low dopings. Some of our results may be useful as benchmarks for future numerical calculations. We suggest that presently available diagonalization studies overestimate the spectral weight and underestimate its doping dependence, perhaps because the presently accessible system sizes are too small.

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