Theory of Raman Scattering in Mott-Hubbard Systems

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We present a theory of Raman scattering in the Hubbard model. The scattering of light has two contributions. One gives rise to scattering by spin degrees of freedom in the insulating case where the general form of the scattering Hamiltonian is derived. The fluctuations of the "chiral" spin operator $\sum \mathbf{s}_i \cdot (\mathbf{s}_j \times \mathbf{s}_k)$ are shown to contribute in the B_{2g} scattering geometry. The other contributes in the doped case and is shown to probe the fluctuations of the "*stress tensor.*" This quantity is not conserved, and hence its fluctuations at small $\mathbf{q}$ inherent in optical experiments need not be small, in striking contrast to density fluctuations in usual metals.

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Raman scattering (inelastic light scattering) has provided extremely important quantitative and qualitative information in high-T_c materials, in both the insulating and doped cases.[1-3] Here we will be concerned with the question of describing the nonphonon contribution to Raman scattering, within a unified theoretical framework. A simplifying feature of the Mott-Hubbard system is that a single effective band, or a few bands, are involved, with the result that all couplings to the external world are readily parametrized. For example, the external electromagnetic field couples to electrons through a well-defined phase factor in the hopping matrix element—the Peierls phase. This serves below as the starting point for a unified description of both the "magnetic Raman scattering" in the insulating phase and the "electronic Raman scattering" in the doped, i.e., metallic, phase of the Mott-Hubbard system. In the former case, we reexamine the derivation and regime of validity of the effective Hamiltonian for resonant light scattering. In the doped case we argue that scattering is dominated by *non-free-electron* effects.

We do not intend here to present detailed calculations in either phase in view of their intractability, but rather wish to outline *the framework* in which such calculations should be undertaken. The present theory of Raman scattering would apply to high-T_c systems, if one accepts Anderson's[4] assertion that these are Mott-Hubbard systems. Our conclusions are that this point of view is not only consistent with the bulk of the data for the insulators, but seems to provide a natural and a testable interpretation of the Raman data in the doped case. For simplicity of presentation we confine ourselves to a one-band Hubbard model in two dimensions, leaving the generalizations to a three-band model for a later publication (our U below is the $\epsilon_p - \epsilon_d$ of the latter).

Consider the Hubbard Hamiltonian in the presence of a weak external transverse electromagnetic field $\mathbf{A}$: $H = \tilde{H}_t + H_U$, where $H_U = U\sum n_{r\uparrow} n_{r\downarrow}$ and

$$\tilde{H}_t = \sum_k \epsilon(k) c_\sigma^\dagger(k) c_\sigma(k) - \frac{e}{\hbar c}\sum_k \mathbf{j}(k)\cdot\mathbf{A}(-k) + \frac{e^2}{2\hbar^2 c^2}\sum_{k,k'} A_\alpha(-k)\tau_{\alpha,\beta}(k+k')A_\beta(-k') . \tag{1}$$

The operators $c_\sigma(k)$ destroy electrons with spin σ and momentum k defined for the square lattice in 2D. The energy is $\epsilon(k) = -2t[\cos(k_x)+\cos(k_y)]$, the current operator is

$$j_\alpha(q) = \sum_k v_\alpha(k) c_\sigma^\dagger(k+q/2) c_\sigma(k-q/2) , \tag{2}$$

where $v_\alpha(k) = \partial\epsilon(k)/\partial k_\alpha$, and the stress tensor $\tau_{\alpha,\beta}$ is

$$\tau_{\alpha,\beta}(q) = \sum_k \frac{\partial^2\epsilon(k)}{\partial k_\alpha \partial k_\beta} c_\sigma^\dagger(k+q/2) c_\sigma(k-q/2) . \tag{3}$$

Finally, $\mathbf{A}_\alpha(q) = e_\alpha g(q)(a_q^\dagger + a_{-q})$, with $g(q) = (hc^2/V\omega_q)^{1/2}$ and with a_q destroying photons with polarization $\mathbf{e}$, momentum $\mathbf{q}$, and energy $\omega_q = c|q|$. From this point on we switch to units where $\hbar = c = 1$.

The Raman-scattering cross section is determined by the transition probability rate R obtained from the "golden rule":

$$R(q,\Omega) = 2\pi\sum_{i,f}\exp(-\beta\epsilon_i)|g(k_i)g(k_f)e_\alpha^i e_\beta^f\langle f|M^{\alpha,\beta}(q)|i\rangle|^2\delta(\epsilon_f - \epsilon_i - \Omega) , \tag{4}$$

where $\omega_{i(f)}$, $k_{i(f)}$, and $e_\alpha^{i(f)}$ denote the energy, momentum, and polarization of the initial (final) states of the photon, $\epsilon_{i(f)}$ refer to the Hubbard eigenstates describing the "matter," $\Omega = \omega_i - \omega_f$, and $q = k_i - k_f$.

The scattering operator has both resonant and nonresonant contributions $M(q) = M_R(q) + M_N(q)$:

$$\langle f|M_N^{\alpha,\beta}(q)|i\rangle = \langle f|\tau_{\alpha,\beta}(q)|i\rangle \tag{5}$$

and

$$\langle f|M_R^{\alpha,\beta}(q)|i\rangle = \sum_\nu \left\{ \frac{\langle f|j_\beta(k_f)|\nu\rangle\langle\nu|j_\alpha(-k_i)|i\rangle}{\epsilon_\nu - \epsilon_i - \omega_i} + \frac{\langle f|j_\alpha(-k_i)|\nu\rangle\langle\nu|j_\beta(k_f)|i\rangle}{\epsilon_\nu - \epsilon_i + \omega_f} \right\} . \tag{6}$$

We now proceed to examine the undoped insulating case corresponding to exactly one electron per site for large U/t. It is clear that the low-energy Raman scattering, at least when $\Omega \leq U$, is dominated by the resonant terms [Eq. (6)] since the stress tensor does not have low-energy matrix elements. The Hilbert space splits into bands labeled by n, the number of doubly occupied sites, which we denote by $|n,\alpha\rangle$. The lowest Hubbard band (LHB) $n=0$ can be parametrized by the set of $S^z(r)$ eigenvalues specifying a spin configuration $\alpha=(\{\alpha\})$, while the upper Hubbard band (UHB) with $n=1$ requires the specification of the doubly occupied site r_d and the hole site r_h, in addition to the spins, so that $|1,\alpha\rangle = c_\sigma^\dagger(r_d)c_\sigma(r_h)|\{\sigma\}\rangle$, where $|\{\sigma\}\rangle = \prod_r c_{\sigma_r}^\dagger|0\rangle$. To lowest order in t/U, these are degenerate. The $n=0$ manifold degeneracy is lifted by the superexchange arising from *second-order* hopping "t" matrix elements giving the Heisenberg model $H_{ex}=(4t^2/U)\sum_{r,\mu}[\mathbf{s}_r\cdot\mathbf{s}_{r+\mu}-\frac{1}{4}]$. In contrast, the degeneracy of the $n=1$ manifold (UHB) is lifted already at first order in t by the kinetic energy of the hole and the "double." To leading order in t/U, only $n=1$ UHB states appear as intermediate states, and Eq. (6) (for $k_i\sim k_f\sim 0$) becomes

$$\langle f|M_R|i\rangle \sim \sum_{\alpha,r,r'} \langle 0,\{\sigma_f\}|j_\mu(r)|1,\alpha\rangle\langle 1,\alpha|j_\nu(r')|0,\{\sigma_i\}\rangle \times\left[\frac{e_\mu^f e_\nu^i}{\epsilon_{1,\alpha}-\omega_i}+\frac{e_\mu^i e_\nu^f}{\epsilon_{1,\alpha}+\omega_f}\right], \quad (7)$$

with the bond current operator $j_\nu(r)=it[c_\sigma^\dagger(r+\nu)\times c(r)-\text{H.c.}]$. To the zeroth order in t/U, $\epsilon_{1,\alpha}=U$, and hence can be pulled outside the intermediate-state summation. The sum can be carried out and the resulting answer for the matrix elements of M_R written in terms of effective spin operators as in Fleury and Loudon,[5]

$$H_{LS}=\sum_{r,\mu}(\tfrac{1}{4}-\mathbf{s}_r\cdot\mathbf{s}_{r+\mu})\left[\frac{t^2}{U-\omega_i}\right][\mathbf{A}^f\cdot\boldsymbol{\mu}][\mathbf{A}^i\cdot\boldsymbol{\mu}]. \quad (8)$$

The argument leading to Eq. (8) is the same as the one used in deriving the superexchange, with the modification that $\mathbf{j}\cdot\mathbf{A}$ is replaced by H_t, and the energy denominators do not contain ω_i [we drop the second term of Eq. (6) here and in the rest of the paper[6]]. The latter is a crucial difference since the perturbative expansion relies on the smallness of $t/(U-\omega_i)$ which breaks down in the resonant regime $U\sim\omega_i$. More generally, the calculation of the scattering matrix element [Eq. (7)] involves the UHB propagator

$$G_E(r_h',r_d',\{\sigma'\};r_h,r_d,\{\sigma\}) \equiv \langle\sigma'|c_{r_h'}^\dagger c_{r_d'}[H_t-E]^{-1}c_{r_h}^\dagger c_{r_d}|\sigma\rangle. \quad (9)$$

The intermediate-state summation in Eq. (7) can be written in terms of the off-diagonal elements of G_E where $E=U-\omega$, and since holes and doubles form extended states in the UHB, we expect G_E to have a continuous spectrum with bandwidth of $\sim t$. However, because of the interaction of the carriers with the background spins, the proper calculation is an intricate Brinkman-Rice[7] kind of a problem which will be considered elsewhere.[8] The lowest-order scattering Hamiltonian, as in Eq. (8), is clearly the leading term in a moment expansion $(H_t-E)^{-1}=-E\sum_{m=0}^\infty(E^{-1}H_t)^m$, while the general terms of the expansion contain both nonlocal and multispin exchanges of a rich variety. Here we present the next few terms in the moment expansion, which are obtained by combining Eqs. (7) and (9) and some tedious algebra.

Defining $\Lambda^{\pm}_{\alpha,\beta,\gamma,\delta}\equiv[g(k_i)g(k_f)]^{-1}[A_\alpha^i A_\beta^f \pm A_\gamma^i A_\delta^f]$, $\mathcal{P}_\alpha \equiv\sum_r \mathbf{s}_r\cdot\mathbf{s}_{r+\alpha}$, $\mathcal{Q}_{\alpha,\beta,\gamma}\equiv\sum_r(\mathbf{s}_r\cdot\mathbf{s}_{r+\alpha})(\mathbf{s}_{r+\beta}\cdot\mathbf{s}_{r+\gamma})$, and $\Delta\equiv t/(U-\omega_i)$, we find that

$$\langle f|M_R|i\rangle=\langle f|[O_s\Lambda^+_{x,x,y,y}+O_d\Lambda^-_{x,x,y,y}+O_e\Lambda^+_{x,y,y,x}+O_o\Lambda^-_{x,y,y,x}]|i\rangle, \quad (10)$$

with the operators

$$O_s\equiv t\Delta[N/4-\tfrac{1}{2}(\mathcal{P}_x+\mathcal{P}_y)]+2t\Delta^3[N/4+\mathcal{P}_{2x}+\mathcal{P}_{2y}+\mathcal{P}_{x+y}+\mathcal{P}_{x-y}+2(\mathcal{Q}_{x,y,x+y}+\mathcal{Q}_{y,x,x+y}-\mathcal{Q}_{x+y,x,y})], \quad (11)$$

$$O_d\equiv t\Delta(\tfrac{1}{2}-4\Delta^2)[\mathcal{P}_y-\mathcal{P}_x]-2t\Delta^3[\mathcal{P}_{2x}-\mathcal{P}_{2y}], \quad (12)$$

$$O_e\equiv -t\Delta^3[N+4\mathcal{P}_{x+y}+4\mathcal{P}_{y-x}], \quad (13)$$

and

$$O_o=8t\Delta^3\sum_r i\epsilon_{\mu\mu'}\mathbf{s}_r\cdot(\mathbf{s}_{r+\mu}\times\mathbf{s}_{r+\mu'}), \quad (14)$$

where $\mu=\pm x,\pm y$ and $\epsilon_{\mu,\nu}=-\epsilon_{\nu,\mu}=-\epsilon_{-\mu,\nu}$. The operators in Eqs. (11)–(14) may be regarded as new terms in the effective light-scattering Hamiltonian. Several comments are in order. The terms involving O_s and O_d appear already in the earlier work of Fleury and Loudon;[5] however, one sees that $[O_s,H_{ex}]\neq 0$, and so the A_{1g} scattering does not vanish as it would if O_s were truncated at the lowest order. The previously overlooked terms O_e and O_o contribute, respectively, in the A_{1g},B_{2g} and in the B_{1g},B_{2g} geometries. Their precise contribution is calculable with standard techniques for the antiferromagnet.

One may note that O_o is odd under time reversal, and the corresponding term only contributes inasmuch as $\mathbf{e}^f\neq\mathbf{e}^i$. It is precisely the chiral spin operator that has been suggested to have a nonzero ground-state expectation in some recent theories[9] of high-T_c systems. If that were the case, it would contribute to $\epsilon_{xy}(\Omega=0)$ as can be seen from the elastic limit of Eq. (6). In Raman scattering, we have shown that the fluctuations of this operator contribute in the B_{2g} geometry.

We now turn to the doped case where the nonresonant terms [Eq. (5)] give nontrivial dominant contributions since the kinetic energy, or stress tensor, has matrix element between states in the LHB, i.e., with low-energy transfers.[10] The simplest case corresponds to the geometry $\mathbf{e}_i=\mathbf{e}_f=\mathbf{x}$ for which the scattering intensity is proportional to

$$I_{xx}(q,\Omega)=\sum_{i,f}\exp(-\beta\epsilon_i)|\langle f|\tau_{xx}(q)|i\rangle|^2\delta(\epsilon_f-\epsilon_i-\Omega)\,. \tag{15}$$

The case of parabolic bands[11-14] is obtainable by specializing to $\epsilon(k)\sim k^2$, which by Eq. (3) replaces $\tau_{\alpha,\beta}(q)$ by $\delta_{\alpha,\beta}\rho(q)$. In this case, the scattering vanishes as $q\rightarrow 0$, the integrated intensity vanishing as q^n, with $n=1$ if the long range of the Coulomb interaction is neglected, and with $n=2$ otherwise. This is the inescapable consequence of particle-number conservation, i.e., $[\rho_q,H]\rightarrow 0$ as $q\rightarrow 0$. However, for Mott-Hubbard systems, this suppression does not apply since $\tau_{xx}(q=0)$ does *not* commute with the full Hamiltonian, and *can* cause scattering between the eigenstates of H. The effect of nonparabolicity of the bands [i.e., $\tau_{xx}(q)\neq\rho(q)$] was discussed by Wolff,[15] who found within the random-phase approximation that $I_{xx}(q,\Omega)\neq 0$ for $\Omega\leq qv_f$, the latter constraint being a consequence of the limited phase space available for scattering the quasiparticle. However, for Mott-Hubbard systems, the scattering is actually dominated by the incoherent part of the carrier spectral function which defeats the phase-space limitation, resulting in the nonvanishing of $I_{xx}(q,\Omega)$, over a broad range of $\Omega\approx t$.

We can relate I_{xx} to a "stress susceptibility" by the fluctuation-dissipation theorem and write $I_{xx}(q,\Omega)=\mathrm{Im}\chi^{(\tau)}(q,\Omega)/[1-\exp(-\beta\Omega)]$ and express

$$\chi^{(\tau)}(q,\Omega)=N^{-1}\sum_{k,k'}\gamma^{(\tau)}(k)\gamma^{(\tau)}(k')M_{k,k'}(q,\Omega)\,, \tag{16}$$

where the bare vertex $\gamma^{(\tau)}(k)=\cos(k_x)$. The function M is given as the sum of the "bubble" and the vertex contributions,[8]

$$M_{k,k'}(q,\Omega)=\delta_{k,k'}A_k(q,\Omega)+N^{-1}B_{k,k'}(q,\Omega)\,.$$

The term A_k corresponds to the bubble diagram with fully renormalized propagators and can be formally evaluated in terms of the single-particle spectral functions. The contribution arising from the coherent part of these vanishes as $q\rightarrow 0$ at zero temperature. The incoherent part is, in general, nonzero at all wave vectors but vanishes as $\Omega\rightarrow 0$ as Ω^{γ}, with $\gamma\geq 1$. The same function $M_{k,k'}$ determines the density and current susceptibilities $\chi^{(\rho)}(q,\Omega)$ and $\chi^{(j)}(q,\Omega)$, which are obtainable from Eq. (16), by replacing the stress vertex $\gamma^{(\tau)}$ by unity and the current vertex $v_a(k)$, respectively.

The conservation of particle number leads to the Ward-Takahashi identities relating the vertex correction term $B_{k,k'}$ and the bubble term A_k through $\langle B_{k,k'}(0,\Omega)\rangle_{k,k'}=-\langle A_k(0,\Omega)\rangle_k$; for all Ω, whereby $\mathrm{Im}\chi^{(\rho)}(0,\Omega)\equiv 0\ \forall\ \Omega$. The implication is that the Raman cross section in the doped case, if due to density fluctuations, should vanish with q, the photon momentum transfer which is usually very small compared to, say, the Fermi momentum. In actual experiments on high-T_c materials[1-3] the typical q is roughly the inverse skin depth $\lambda_s\approx 1000$ Å. We then expect that for nonparabolic Mott-Hubbard systems, the resulting integrated intensity should be larger than that in a typical free-electron metal by factors of order $(\lambda_s/a_0)^m$, where a_0 (≈ 5 Å) is a lattice constant, or typical interparticle spacing, and m is either 1 or 2.

We are able at present to give only a very crude estimate of the Raman intensity. It is readily seen that the intensity integrated over frequencies reduces to

$$\int_0^{\Omega_c}d\Omega\, I_{xx}(0,\Omega)=\langle P_G\tau_{xx}P_G\tau_{xx}P_G\rangle-\langle P_G\tau_{xx}P_G\rangle^2\,,$$

where P_G projects out doubly occupied sites, and the cutoff frequency Ω_c is assumed to be in the range $t\leq\Omega\leq U$, so that only LHB states can contribute (crudely $\Omega_c\sim$insulating optical gap), and should scale like t^2 times the hole density δ. This intensity should be seen in the B_{1g} and A_{1g} geometries with $\frac{1}{2}(\tau_{x,x}\mp\tau_{y,y})$ replacing τ_{xx} in Eq. (15). The various moments can be calculated as higher commutators,[8] and expressed as correlation functions.[16] In contrast to the density-fluctuation picture of Raman scattering, which leads to well-known features including a particle-hole continuum with a well-defined cutoff qv_f and a sharp plasma mode arising from collective density fluctuations, in Mott-Hubbard systems the intermediate states probed need not necessarily have any sharp structure since the stress tensor is not expected to create well-defined elementary excitations. Thus we expect a broad continuum with a bandwidth of order t, with an "anomalously" large intensity compared to free-electron metals, scaling like the hole density near half filling. This description is qualitatively consistent with the experiments[1-3] where the largest energy transfer is ~ 1 eV.

An interesting cross-check is provided by considering the optical conductivity $\mathrm{Re}\sigma_{xx}(0,\Omega)=\mathrm{Im}\chi^{(j)}(0,\Omega)/\Omega$. In the approximation where only the bubble contributions to Reσ are retained, these are essentially identical since $\langle\cos^2(k_x)\rangle=1-\langle\sin^2(k_x)\rangle$. The optical experiments[17] on $YBa_2Cu_3O_7$ do seem to bear out, albeit crudely, this pseudoidentity,

$$I_{x,x}(0,\Omega)\sim\Omega/[1-\exp(-\beta\Omega)]\mathrm{Re}\sigma_{xx}(0,\Omega)\,,$$

for small enough Ω, although at higher frequencies there seem to be significant departures.[18] In the limit $\Omega\leq kT$, the above reduces to $I_{xx}\sim kT\,\mathrm{Re}\sigma_{xx}$; together with a temperature-independent $I_{xx}(0,0)$, it implies a linear resistivity, which is a ubiquitous feature in the high-T_c materials.

For this pseudoidentity and also the Ward-Takahashi identity to hold, we would have to argue that the vertex corrections are small in the non-s-wave channels of the function $B_{k,k'}$. Recent work on the Hubbard model[19] shows that this scenario is realized in the limit of high dimensions. We should also stress that these considera-

tions apply only to small momentum transfers in the singlet particle-hole channel; the triplet-channel spin susceptibility for $q \sim [\pi,\pi]$ is expected to have substantial structure in view of the oxygen and copper planar NMR relaxation rates.[20]

We summarize by returning to recent experiments[21] in the insulating case where it is seen that the integrated intensity in the B_{1g}, B_{2g}, and A_{1g} geometries are all of the same order of magnitude, and, in fact, as the laser frequency ω_L is changed, the A_{1g} and B_{2g} intensities can be *larger* than that in B_{1g}. Within B_{1g} geometry, a reasonable understanding of the scattering has been reached by Singh *et al.*[22] The fact that the scattering intensity in A_{1g} and B_{2g} geometries exceeds that in B_{1g} is not easy to reconcile with the fact that the second-neighbor exchange J_2 is smaller than J_1 by at least an order of magnitude. Within our framework, however, it is clear that the ratio of the prefactors of the scattering operators that contribute to A_{1g} and B_{1g} [see Eqs. (11) and (14)] is different from J_2/J_1, and, in fact, it involves a different power of $t/(U-\omega_L)$ when such an expansion is possible. In general, when $U \sim \omega_L$, the forms of the spin operator that can contribute involve arbitrarily long strings of spins; the upper-Hubbard-band propagator is transmuted into the scattering operator. Experiments have been done with laser frequencies between 2.4 and 2.8 eV, which corresponds fairly closely to the effective U in the 2D cuprates.[23] Hence we believe that the experiments are probing the most interesting and difficult region of Raman scattering in the Mott-Hubbard system.

The most striking prediction of this theory concerns the **q** dependence of the inelastic Raman scattering: We are predicting that the integrated intensity *does not* vanish as $q \rightarrow 0$, simply because the conservation laws do not force this vanishing. This prediction should be amenable to experimental test, and is in our opinion a crucial test of the applicability of the standard dielectric function theory of Raman scattering[11,12] to the high-T_c systems. It also follows that the semiconductor n-type InSb and others having strong nonparabolicity could also display a nonvanishing intensity as $q \rightarrow 0$, the effects being proportional to $1-z_k$, and largest when the electron density is low (i.e., the effective $r_s \geq 3$). The other important result shown here is the fact that the B_{2g} geometry is particularly favorable for studying quasielastic time-reversal violation, both in the insulating phase [Eq. (14)] and in the doped phase.[24] It is outside the scope of this work to give an estimate of the relevant correlation functions, and we hope that our result stimulates concrete calculations of these that can be tested against experiment.[24]

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