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Raman Scattering in Mott-Hubbard Systems

B. Sriram Shastry and Boris I. Shraiman

AT&T Bell Laboratories
Murray Hill, New Jersey 07974

ABSTRACT

We present a theory of Raman Scattering in the Hubbard model. The scattering of light has resonant and non-resonant contributions. The resonant term gives rise to scattering by spin degrees of freedom in the insulating case, for which we derive a general form of the effective scattering Hamiltonian. The later involves four terms with distinct symmetry properties, one of them being the "chiral" spin operator $\sum S_i \cdot (S_j \times S_k)$. The corresponding four distinct correlation function can be measured directly in the experiments with different scattering geometries. The non-resonant term contributes to the scattering 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 with the density fluctuation in usual metals.

1. INTRODUCTION

Raman scattering has provided extremely important quantitative and qualitative information on high-T_c materials, both in the insulating and doped cases.[1-3] Here we will be concerned with the question of describing the non-phonon contribution to Raman scattering within a unified theoretical framework. The commonly made classification of "magnetic-Raman", and "electronic-Raman" scattering to describe the undoped and doped cases is, from a fundamental point of view, artificial, especially in a Mott-Hubbard system with its complex many-body behavior. Yet the Mott-Hubbard system is simple, as it involves effectively just one or few bands with the result that the coupling to the external electromagnetic field is readily parametrized. Specifically, when the photon frequency is smaller than an appropriate band gap, the external field couples to the electron through the phase of the hopping matrix element, i.e. via the Peierls coupling. This will serve below as a starting point for the unified description of Raman scattering in both insulating and doped cases. We shall reexamine the derivation of the effective Hamiltonian for the "magnetic" Raman scattering in the insulating case, paying particular attention to the regime of validity and emphasizing the symmetry aspects. We will show that for the incident light frequency comparable with Mott gap (i.e. under the resonance conditions), the effective Hamiltonian involves a hierarchy of long range and multiple spin terms, which however split naturally into four groups with distinct symmetry properties. In particular we shall indicate a method to isolate scattering with A_2 symmetry (odd under 2D square lattice reflections) which measures the fluctuations of the topological charge, a quantity that is odd under time reversal. More generally, we will show that the symmetry analysis allows us to disentangle the four correlation functions from five measurements. In the doped case we shall argue that scattering is dominated by the *non-free* electron effects and indicate the connection with optical conductivity. We do not intend here to present detailed calculations in either phase, but rather wish to outline the framework in which such calculations should be undertaken. Finally the present theory of Raman scattering would apply to Hi$-T_c$ systems, if one accepts Anderson's[4] assertion that these are Mott-Hubbard systems. Our conclusions, anticipating somewhat the rest of the paper, are that this point of view is not only consistent with the bulk of the data, but seems to be a natural, almost unique and in any case a testable explanation of the Raman data.

In Section 2 we shall derive the general expression for the Raman scattering intensity starting from the Peierls coupling, then relate the scattering intensity to specific correlation functions while emphasizing the symmetry aspects.

Section 3 deals with the case of the insulating phase. First the derivation of super-exchange effective Hamiltonian governing the dynamics of the lower Hubbard band (LHB) is reviewed. Next, the light scattering effective Hamiltonian is constructed using the moment expansion, making explicit the distinction between the exchange and light scattering effective Hamiltonians under the conditions of resonant scattering.

Section 4 addresses the doped case: the inapplicability of density fluctuation theory to light scattering for the Hubbard model is discussed and the importance of the non-free electron effects is emphasized.

Section 5 outlines the experimental implication of the theoretical analysis presented below. Finally, Appendix A deals with the relation between the quasielastic limit of the resonant Raman scattering metric element and the dielectric function. A brief account of this work has appeared in Ref. 10.

2. GENERAL FORMALISM

For simplicity of presentation we confine ourselves to a one band Hubbard model; the generalizations to a three band model, that reduces to the above under appropriate projections, will be presented in a later publication. Consider Hubbard Hamiltonian $H = H'_T + H_U$ in the presence of an external transverse e-m-field, described by a vector potential $\vec{A}$:

$$H'_T = -t \sum_{<ij>} \left[e^{i\frac{e}{\hbar c}\int_i^j \vec{A}\cdot d\vec{\ell}} c^+_{j\sigma} c_{i\sigma} + c^+_{i\sigma} c_{j\sigma} e^{+i\frac{e}{\hbar c}\int_j^i \vec{A}\cdot d\vec{\ell}} \right] \tag{1}$$

and $H_U \equiv U \sum_r n_{r\uparrow} n_{r\downarrow}$. Here i and j refer to appropriate Wannier states in this "one band". Assuming that the vector potential has a spatial variation that is much slower than the lattice length scale ($a_o = 1$), we approximate

$$\int_{\vec{r}}^{\vec{r}+\hat{x}} \vec{A}\cdot d\vec{\ell} \to \vec{A}\left(\vec{r}+\frac{\hat{x}}{2}\right)\cdot\hat{x}$$

Introducing usual fourier components $c_{k\sigma} = \frac{1}{\sqrt{N}} \sum_k \exp(i\mathbf{k}\cdot\mathbf{r})\ c_{r\sigma}$, and $A^\alpha_q = \sum_{\vec{q}} A^\alpha(\vec{r}+x/2)\ \exp\ (i\,\vec{q}\cdot r+\hat{x}/2)$, we expand Eq. (1) out to second order in A to find $H'_T = H_T + H_{coupl.}$, where H_T is the zero field hopping $T_\mu = \sum_r [c^+_{r+\mu} c_r + h.c.]$ and $H_T = -t \sum_\mu T_\mu$,

$$H_{coupl} = -\left[\frac{e}{\hbar c}\right] \Sigma\, j^\alpha_q A^\alpha_{-q} + \frac{1}{2}\left[\frac{e}{\hbar c}\right]^2 \Sigma\, \tau^{\alpha\beta}_{q_1+q_2}\, A^\alpha_{-q_1} A^\beta_{-q_2} \tag{2}$$

with the current operator

$$j^\alpha_q = \sum_k \frac{\partial \varepsilon_k}{\partial k^\alpha}\ c^+_{k+\frac{q}{2},\sigma} c_{k-\frac{q}{2},\sigma}$$

and the second rank stress tensor

$$\tau^{\alpha,\beta}_q = \sum_k \frac{\partial^2 \varepsilon_k}{\partial k^\alpha \partial k^\beta}\ c^+_{k+\frac{q}{2},\sigma} c_{k-\frac{q}{2},\sigma} \tag{3}$$

where $\varepsilon_k = -2t \sum_\alpha \cos k_\alpha$ is the usual band energy. Note that replacing $\varepsilon_k \sim k^2$ yields the familiar coupling with $\tau_{\alpha\beta}$ replaced by the density operator. In the second quantized form the electromagnetic field operator A^α becomes

$$A^\alpha_q = g_q(e_\alpha a_{-q} + \bar{e}_\alpha a^+_q) \tag{4}$$

where $\hat{e}$, is the complex unit vector labelling photon polarization, overbar denotes complex conjugation, $g_q \equiv \sqrt{\frac{hc^2}{\omega_q \Omega}}$, $\omega_q = c|q|$ is the photon energy and a^+_q is the photon creation operator with energy momentum $\omega,\vec{q}$ and appropriate polarization. From here onwards we shall switch to the units where $\hbar = c = 1$.

In order to study the scattering we will write Eq. (2) as two terms $H_{coupl} = V^{(1)} + V^{(2)}$; where $V^{(1)} = -e\,\Sigma\, j^\alpha_q A^\alpha_{-q}$ and $V^{(2)} = \frac{1}{2} e^2\, \Sigma\, \tau^{\alpha\beta}_{q_1+q_2}$ $A^\alpha_{-q_1} A^\beta_{-q_2}$. The basic Raman scattering cross section is proportional to the transition rate R, which is obtained from the Golden rule

$$R = \sum_{i,f} |\langle f| M |i\rangle|^2 \, \delta (E_f - E_i) \, , \tag{5}$$

where M is the effective scattering operator causing a scattering between the initial state i and some final state f. These states are direct products of the 1-photon states and appropriate matter states (i.e. states of the Hubbard model) with energies $E_i = \omega_i + \varepsilon_i$, $E_f = \omega_f + \varepsilon_f$, where ε's denote the eigenvalues of the Hubbard H. The initial and final energy, momentum and polarization of the photon field are $(\omega_i, \vec{k}_i, \hat{e}_i)$ and $(\omega_f, \vec{k}_f, \hat{e}_f)$ respectively with an energy momentum transfer $\Omega = \omega_f - \omega_i$, and $\vec{q} = \vec{k}_f - \vec{k}_i$. The two terms in H_{coupl} have different character: $V^{(1)}$ can cause scattering only with the help of a virtual intermediate state and hence is called the resonant term, whereas the term $V^{(2)}$ can cause transitions between these states in lowest order and is non-resonant. After elementary manipulations we write the "matter part", of the inelastic scattering, with an energy momentum transfer Ω,q summed over final states and averaged over initial states with an appropriate Boltzman weight, as

$$R(q,\Omega) = \sum_{f,i} e^{-\beta\varepsilon_i} \, |g_{ki} \, g_{kf} \langle f| M |i\rangle|^2 \delta (\varepsilon_f - \varepsilon_i - \Omega) \tag{6}$$

where $M = M_R + M_N$,

$$\langle f|M_N|i\rangle = e_i^\alpha \bar{e}_f^\beta \, \langle f|\tau_q^{\alpha\beta}|i\rangle \tag{7}$$

and

$$\langle f|M_R|i\rangle = \sum_\nu \left[\frac{\langle f|(\vec{j}_{k_f} \cdot \bar{\hat{e}}_f)|\nu\rangle\langle\nu|(\vec{j}_{-k_i} \cdot \hat{e}_i)|i\rangle}{\varepsilon_\nu - \varepsilon_i - \omega_i} \right.$$

$$\left. + \frac{\langle f|(\vec{j}_{-k_i} \cdot \hat{e}_i)|\nu\rangle\langle\nu|(\vec{j}_{k_f} \cdot \bar{\hat{e}}_f)|i\rangle}{\varepsilon_\nu - \varepsilon_i + \omega_f} \right] \tag{8}$$

The transition matrix element given in Eqn. 8 defines the resonant scattering operator $M_R \equiv e_i^\alpha \bar{e}_f^\beta \, M_R^{\alpha\beta}$

$$M_R^{\alpha\beta}(t) = \int_0^\infty dt\, e^{-i(\omega_{in} - i0^+)t'}\, i\,[j_{k_i}^{\alpha}(t+t'),\, j_{k_f}^{\beta}(t)] \tag{9}$$

Combining resonant and non-resonant contribution one defines the effective scattering operator $M = e_i^{\alpha}\bar{e}_f^{\beta} M^{\alpha\beta}$:

$$M^{\alpha\beta} = \tau_q^{\alpha\beta} + M_R^{\alpha\beta}\ .$$

The scattering rate as a function of energy and momentum transfer probes the correlation function of M:

$$R = \sum_i e^{-\beta\epsilon_i} \int dt\, e^{i\Omega t} < i\,|M_q^{\dagger}(0)\, M_q(t)|\,i> \tag{10}$$

The scattering operator $M = M^{\alpha\beta}\, e_i^{\alpha}\bar{e}_f^{\beta}$ can be decomposed into four terms corresponding to one dimensional irreducible representation of the 2-d square lattice symmetry groups C_{4v}

$$M = \frac{1}{2} O_{A_1}(e_i^x\bar{e}_f^x + e_i^y\bar{e}_f^y) \tag{11}$$

$$+ \frac{1}{2} O_{B_1}(e_i^x\bar{e}_f^x - e_i^y\bar{e}_f^y)$$

$$+ \frac{1}{2} O_{B_2}(e_i^x\bar{e}_f^y + e_i^y\bar{e}_f^x)$$

$$+ \frac{i}{2} O_{A_2}(e_i^x\bar{e}_f^y - e_i^y\bar{e}_f^x)$$

where we introduced operators O_ν labeled by representation $\nu \in \{A_1, A_2, B_1, B_2\}$ (in the notation of Hammermesh[5]) with A_1 transforming like $x^2 + y^2$, B_1-like $x^2 - y^2$, A_2-like $x^3y - y^3x$ and B_2-like xy. The lattice symmetry implies that there are only four independent correlation functions $<O_\nu^{\dagger} O_{\nu'}> = C_\nu \delta_{\nu\nu'}$ contributing to the scattering rate in Eqn. 10.

The above symmetry consideration are of crucial importance in interpreting the polarization dependence of Raman scattering. Indeed, the scattering observed in experiment with different scattering geometries can be expressed in terms of correlation functions C_ν.

In Table I below we summarize the result for the three commonly used experimental configuration with linearly polarized light (A'_{1g}, B_{1g} and B_{2g} in the notation of Ref. 1-3) as well as for scattering of left circularly polarized light into left or right polarization (denoted LL and LR respectively).

Table I

		$\hat{e}_i$	$\hat{e}_f$	R
I	A'_{1g}	$(\hat{x} + \hat{y})/\sqrt{2}$	$(\hat{x} + \hat{y})/\sqrt{2}$	$C_{A_1} + C_{B_2}$
II	B_{1g}	$(\hat{x} + \hat{y})/\sqrt{2}$	$(\hat{x} - \hat{y})/\sqrt{2}$	$C_{B_1} + C_{A_2}$
III	B_{2g}	$\hat{x}$	$\hat{y}$	$C_{A_2} + C_{B_2}$
IV	LR	$(\hat{x} + i\hat{y})/\sqrt{2}$	$(\hat{x} - i\hat{y})/\sqrt{2}$	$C_{B_1} + C_{B_2}$
V	LL	$(\hat{x} + i\hat{y})/\sqrt{2}$	$(\hat{x} + i\hat{y})/\sqrt{2}$	$C_{A_1} + C_{A_2}$

One may note that in contrast with the linear response phenomena (e.g. dielectric tensor) the information obtained from experiment with linearly polarized light is insufficient to determine the scattering of circularly polarized light. Intuitively this can be understood as the result of the interference between incoming and outgoing waves which becomes observable in the Raman experiment because of the bilinear dependence of the scattering matrix element on the e-m fields.

Thus we observe that there are four independent scattering geometries which allow us to measure the correlation functions

$$C_{A_1} = \frac{1}{2}\left[2R_I + R_{II} - R_{III} - R_{IV}\right] \tag{12a}$$

$$C_{A_2} = \frac{1}{2}\left[R_{II} + R_{III} - R_{IV}\right] \tag{12b}$$

$$C_{B_1} = \frac{1}{2}\left[R_{II} + R_{IV} - R_{III}\right] \tag{12c}$$

$$C_{B_2} = \frac{1}{2}\left[R_{III} + R_{IV} - R_{II}\right] \tag{12d}$$

while the 5th configuration provides a useful consistency check which can be used to filter out background contributions:

$$R_V = R_I + R_{II} - R_{IV} \tag{13}$$

In the next section we shall consider the effective scattering operator in more detail deriving explicit expressions for O_v in terms of spin operators (for the undoped case). Specifically we shall draw attention to the A_2 scattering channel which, as is already evident from Eqn. 11, involves the operator O_{A_2} which is odd under time reversal (note that $e_i^x \bar{e}_f^y - e_i^y \bar{e}_f^x$ field configuration that it is coupled to is odd under time reversal). In general one expects that all scattering channels allowed by symmetry will contribute.

3. MAGNETIC RAMAN SCATTERING IN THE INSULATING STATE

Let us now specialize to the case where the Hubbard model has precisely N_a electrons, where N_a is the number of sites. This ground state for this model, in the limit of large U is an antiferromagnetic insulator with the low energy excitation described by the Heisenberg Hamiltonian.[6] The eigenstates fall into two categories, one at or near zero energy and another near U and these are the familiar lower and upper bands of Hubbard. It is clear, that the low energy Raman scattering is dominated by the resonant term M_R in Eq. (8) since the matrix elements of stress tensor in Eq. (7) vanish. The current operators in Eq. (8) necessarily take us from the lower to the upper Hubbard band, and hence the energy differences in the denominators of Eq. (8) are of O(U). The scattering caused by M_R can, to the lowest order in t/U, be expressed as a simple effective spin operator, but to higher orders there are severe difficulties in doing so.

In order to establish notation, let us review briefly the derivation of superexchange.[6] We denote by $\{|n;\alpha>\}$, the manifold of states with "n" doubly occupied sites. The $n = 0$ manifold can be parametrized by the set of $s^z(r)$ eigenvalues specifying the spin configuration $\alpha = (\{\sigma\})$, while the states with double occupancy require in addition the specification of the coordinates of the "hole" and the "double" sites, r_h, r_d so that for $n = 1, \alpha = (\{\sigma\}, r_h, r_d)$ and

$$|1,\alpha> = c^+_\sigma(r_d)c_\sigma(r_h)\ |\{\sigma\}> \tag{14}$$

where

$$|\{\sigma\}> = \prod_r c^+_{\sigma_r}(r)\ |0>$$

To the zeroth order in tU^{-1} these states are degenerate: $H_U|n,\alpha> = nU\ |n,\alpha>$. The $n = 0$ manifold the degeneracy is lifted by the second order process, the superexchange while for $n \neq 0$ case it is lifted in the 1st order by the formation of *extended* hole-double states. To the 1st order in tU^{-1} the lower Hubbard band acquires an admixture of $n = 1$ states:

$$|\sigma> = |0,\{\sigma\}> - \frac{1}{U}\sum_\alpha |1,\alpha><1,\alpha|\ H_T\ |0,\{\sigma\}> . \tag{15}$$

The calculation of the ground state energy to the 2nd order is non-trivial since the Hamiltonian is *not* diagonal in the $|\sigma>$ basis, with the matrix elements

$$<\sigma'|H|\sigma> = \frac{1}{U}\sum_\alpha <0,\{\sigma'\}|\ H_T\ |1,\alpha><1,\alpha|\ H_T\ |0,\{\sigma\}> \tag{16}$$

$$= -\frac{t^2}{U}\sum_{\{\tilde\sigma\}}\sum_r\sum_\mu <0,\{\sigma'\}|(c^+_{r+\mu}c_r)(c^+_r c_{r+\mu})|0,\{\tilde\sigma\}>$$

$$<0,\{\tilde\sigma\}|(c^+_{r+\mu}c_r)(c^+_r c_{r+\mu})|0,\{\sigma\}>$$

$$= -\frac{4t^2}{U}\sum_{\{\tilde\sigma\}}\sum_{r,\mu} <0,\{\sigma'\}|\ P_{r+\mu,r}\ |0,\{\tilde\sigma\}><0,\{\tilde\sigma\}|\ P_{r+\mu,r}|0\{\sigma\}>$$

where $(c^+_{r+\mu}c_r) \equiv c^+_{r+\mu,\sigma}\ c_{r,\sigma}$ and we defined the singlet projection operator

$P_{ij} = \frac{1}{4} - \vec{S}_i \cdot \vec{S}_j = \frac{1}{2}(c^+_{r+\mu} c_r)(c^+_r c_{r+\mu})$, the last equality holding on the manifold of singly occupied states. Upon trivial summation over intermediate state Eqn. 16 leads to the superexchange effective Hamiltonian

$$H_{ex} = J \sum_{r,\mu} [\vec{S}_r \cdot \vec{S}_{r+\mu} - \frac{1}{4}] \tag{17}$$

with Heisenberg exchange constant $J \equiv \frac{4t^2}{U}$.

Note that H_{ex} is only valid to the (t^2U^{-2}) and the next order correction would require diagonalization of the $|1,\alpha>$ manifold with respect to H_T so that the intermediate state energy entering the denominator in Eqn. 15 can be determined to (tU^{-1}). The latter is non-trivial and would lead to higher order (and longer range) exchange term in H_{ex}, as will be discussed in some more detail later.

Let us now return to the light scattering and observe that in the case of the insulator it is entirely due to the second order (resonant) process described by Eqn. 8. To the leading order in t/U only n = 1 intermediate states contribute and Eqn. 8 becomes (under the assumption $k_i \simeq k_f \simeq 0$),

$$g_f^{-1} g_i^{-1} <f|M_R|i> \simeq \sum_\alpha <0,\{\sigma_0\}| \sum_r j_r(r)|1,\alpha> <1,\alpha| \sum_{r'} j_v(r')| 0,\{\sigma_i\}> \times \left[\frac{e^i_r \bar{e}^f_r}{E_{1,\alpha} - \omega_i} + \frac{e^i_r \bar{e}^f_v}{E_{1,\alpha} + \omega_f}\right] \tag{18}$$

with the current operator

$$j_v(r) \equiv it[c^+_\sigma(r+v)c_\sigma(r) - c^+_\sigma(r)c_\sigma(r+v)] \tag{19}$$

To the "zeroth" order in t/U, $E_{1,\alpha} = U$ and hence can be pulled outside the intermediate state summation allowing Eqn. 18 to be rewritten in terms of the spin operators in the same fashion as Eqn. 16, thus leading to the identification of the M_R matrix elements with those of an effective scattering Hamiltonian

$$H_{LS} = \sum_{r,\mu} \left[\frac{1}{4} - \vec{S}_r \cdot \vec{S}_{r+\mu}\right] (\hat{e}^*_f \cdot \hat{\mu})(\hat{e}_i \cdot \hat{\mu}) \left[\frac{1}{U - \omega_i} + \frac{1}{U + \omega_f}\right] \tag{20}$$

This scattering Hamiltonian is that of Elliot, Fleury and Loudon.[7]

The difficulty arises when one attempts to evaluate Eqn. 18 to higher order and is again related to the lifting of the degeneracy of $|1,\alpha>$ states by the hopping part of the Hamiltonian, $<1,\alpha'|H_T|\ 1,\alpha> \neq 0$. The sum over intermediate states has the form

$$\sum_{\alpha} \frac{1}{U - \omega_i + H_T} |1,\alpha><1,\alpha| = \sum_{r_d,r_h} \sum_{\{\sigma\}} \sum_{r'_d,r'_h} \sum_{\{\sigma'\}} G_{U-\omega_i}(r'_h,r'_d,\{\sigma'\}; r_h,r_d,\{\sigma\}) \tag{21}$$
$$|r'_h,r'_d,\{\sigma'\}><r_h,r_d,\{\sigma\}|$$

where we defined the upper Hubbard band propagator:

$$G_E(r'_h,r'_d,\{\sigma'\}; r_h,r_d,\{\sigma\}) = \tag{22}$$

$$= <\sigma'|(c^+_{r_h} c_{r'_d})(H_T - E)^{-1}(c^+_{r_d} c_{r_h})|\sigma>$$

We observe that the expansion

$$(H_T - E)^{-1} = - E^{-1} \sum_{m=0}^{\infty} (E^{-1}H_T)^m \tag{23}$$

which was implicit in passing from Eqn. 18 to Eqn. 20 is no longer an expansion in $t/U << 1$ as it was in the derivation of superexchange but is rather an expansion in $t/(U - \omega_i)$ which under the resonance conditions $\omega_i \approx U$ is quite dangerous indeed! We expect that for $|E| \leq$ const $\sim O(t)$, G_E has a cut corresponding to the continuous spectrum of hole-double states.

Thus while for $|E| = |U - \omega_i| >> t$ the correction to H_{LS} can be computed perturbatively, the opposite limit is quite intricate and involves the study of propagation of holes (or doubles) in the arbitrary spin background.[8,9]

Let us formally express G_E using Eqn. 23

$$G_E = - E^{-1} \sum_{\Gamma_1,\Gamma_2} \left(\frac{t}{E}\right)^{\ell_{r_1}+\ell_{r_2}} <\sigma'|T_r\{\chi(r'_d)\left[\prod_{r\in\Gamma_1}\chi(r)\right]\chi(r_d)\tilde{\chi}(r_h)$$

$$\left[\prod_{\tilde{r}\in\Gamma_2}(-\tilde{\chi}(\tilde{r}))\right]\tilde{\chi}(r'_h)\}|\sigma> \tag{24}$$

where $\chi^{(r)}_{\sigma\sigma'} \equiv c_{r,\sigma}c^+_{r,\sigma'} = \frac{1}{2}\delta_{\sigma\sigma'} - \vec{S}\cdot\hat{\tau}_{\sigma\sigma'}$ is the spin density matrix ($\tilde{\chi}_{\sigma\sigma'} = c^+_{\sigma'}c_{\sigma} = 1 - \chi_{\sigma\sigma'}$) and $\Gamma_{1(2)}$ denote walks connecting $r'_{d(h)}$ and $r_{d(h)}$ (ℓ_Γ

being the length of the walk). Equation 24 clearly contains an off diagonal contribution in both $r_{h,d}$ and $\{\sigma\}$ spaces.

Since j_μ implies $|r_h - r_d| = 1$ we need $G_E(r',r' + \mu', \{\sigma'\}; r,r + \mu,\{\sigma\})$ which perturbatively has the form

$$G_E = + E^{-1}\, \delta_{r,r'}\, \delta_{\mu,\mu'}\, \delta_{\{\sigma\},\{\sigma\}} \tag{25}$$

$$+ t^2 E^{-3} \sum_{\nu,\nu'} \delta_{r',r+\nu}\, \delta_{\mu',\nu'-\nu+\mu}$$

$$<\{\sigma'\}T_r\{\tilde{\chi}(r)\tilde{\chi}(r+\nu)\ \chi(r+\mu+\nu')\ \chi(r+\mu)\}|\{\sigma\}>$$

This combined with Eqn. 18 and 21 leads, after somewhat tedious spin algebra, to a corrected scattering matrix element in the form

$$<f|M_R|i> = g_f g_i <\sigma_f| \Big\{ O_{A_1}(e_x^i \bar{e}_x^f + e_y^i \bar{e}_y^f) \tag{26}$$

$$+ O_{B_1}(e_x^i \bar{e}_x^f - e_y^i \bar{e}_y^f) + O_{B_2}(e_x^i \bar{e}_y^f + e_y^i \bar{e}_x^f)$$

$$+ i\, O_{A_2}(e_x^i \bar{e}_y^f - e_y^i \bar{e}_x^f) \Big\} |\sigma_i>$$

with operators:

$$O_{A_1} \equiv \frac{t^2}{U-\omega} \sum_r \left\{ \left[\frac{1}{4} - \frac{1}{2} S_r \cdot S_{r+x} - \frac{1}{2} S_r \cdot S_{r+y} \right] \right.$$

$$+ \frac{t^2}{(U-\omega)^2} \left[\left[\frac{1}{2} + 2S_r \cdot S_{r+2x} + 2S_r \cdot S_{r+2y} \right. \right. \qquad (27a)$$

$$+ 2S_r \cdot S_{r+x+y} + 2S_{r+x} \cdot S_{r+y} + 4(S_r \cdot S_{r+x})(S_{r+y} \cdot S_{r+x+y})$$

$$\left. \left. + 4(S_r \cdot S_{r+y})(S_{r+x} \cdot S_{r+x+y}) - 4(S_r \cdot S_{r+x+y})(S_{r+x} \cdot S_{r+y}) \right] \right\}$$

$$O_{B_1} \equiv \frac{t^2}{U-\omega} \sum_r \left\{ \frac{1}{2} [S_r \cdot S_{r+y} - S_r \cdot S_{r+x}] - \right.$$

$$\left. \frac{t^2}{(U-\omega)^2} \left[2S_r \cdot S_{r+2x} - 2S_r \cdot S_{r-2y} + 4S_r \cdot S_{r+x} + 4S_r \cdot S_{r+y} \right] \right\} \qquad (27b)$$

$$O_{B_2} = - \frac{4\,t^4}{(U-\omega)^3} \sum_r [S_r \cdot S_{r+x+y} - S_{r+x} \cdot S_{r+y}] \qquad (27c)$$

$$O_{A_2} = \frac{8t^4}{(U-\omega)^3} \sum_r \varepsilon_{\mu\mu'} \vec{S}_r \cdot (\vec{S}_{r+\mu} \times \vec{S}_{r+\mu'}) \qquad (27d)$$

where $\mu = \pm x, \pm y$ and $\varepsilon_{\mu\nu} = - \varepsilon_{\nu\mu} = - \varepsilon_{-\mu\nu}$. The operator in curly brackets on the RHS of Eqn. 26 can be interpreted as a new effective light scattering Hamiltonian.

Several comments are in order. Let us first discuss the physical implication of the new terms appearing in the $t/|U-\omega| << 1$ expansion and then deal with it's validity and possible generalization.

The terms involving O_{A_1} and O_{B_1} appear already in Fleury-Loudon-Elliott (FLE) effective Hamiltonian,[7] however one observes that in general $[O_{A_1}, H_{ex}] \neq 0$ which

has a consequence that A_1 scattering does not vanish as it would if O_{A_1} were truncated at the lowest order. Furthermore in the resonant regime $\omega \sim U$ the "correction" terms are no longer small (note, that the magnitude of the next nearest neighbor exchange terms in H_{LS} is governed by $t/|U-\omega|$ and is *not* related to the strength of corresponding exchange terms in the effective Heisenberg model governing the lowest Hubbard band). Hence the scattering in A_1 configuration need not be small compared to B_1. The terms O_{B_2} and O_{A_2} are new and contribute in various scattering geometries as shown in Table 1. Their exact contribution to the 2-magnon peaks can be obtained by standard techniques.

One may note that operator O_{A_2} is odd under time reversal and contributes to the scattering only in as much as $\mathbf{e}_i \neq \mathbf{e}_f$. It is precisely the chiral spin operator that plays a central role in a number of recent theories[11,12] of high-T_c systems.

Finally, we emphasize that Eqn. 27a-d are based on the moment expansion of $G_{U-\omega}$ and is strictly valid only for $t/|U-\omega| \ll 1$. It is therefore insufficient for predicting the dependence of Raman cross-sections on ω_i in the resonant regime. It is however plausible that the different terms of Eqn. 26 may peak at different frequencies lying within upper Hubbard band, $|U-\omega_i| \sim O(t)$. A more complete treatment of the problem requires a detailed analysis of Eqn. 24 which can be carried out within the Brinkman-Rice (and spin-wave) approximation.[9] This problem will be addressed in the forthcoming publication. As evident from Eqn. 26 in general the scattering will involve matrix elements of products of spin operator which are both non-local in space and form arbitrary long chains of sites. While it would still be possible in principle to write down the corresponding scattering Hamiltonian it would hardly be useful without further approximation.

4. RAMAN SCATTERING IN THE DOPED CASE

Once the number of electron deviates from half filling, the nonresonant terms (Eq. 7) give nontrivial contributions since the kinetic energy, or stress tensor, has matrix elements between states in the *lower* Hubbard band, i.e. with low energy transfers. The simplest case corresponds to the geometry $\hat{e}_i = \hat{e}_f = \hat{x}$ for which the scattering intensity can be written as

$$I_{xx}(q,\omega) \propto \sum_{i,f} e^{-\beta\varepsilon_i} |\langle f| \tau_q^{xx} |i\rangle|^2 \, \delta(\varepsilon_f - \varepsilon_i - \Omega) \tag{28}$$

Note that this matrix element, for the free electron gas[13-16] would necessarily vanish as $q \to 0$, since in that case τ_q^{xx} reduces to the density fluctuation operator ρ_q, and since $\lim_{q\to 0} [\rho_q, H] = 0$, the scattering intensity $I_{xx}(q,\omega)$ must vanish as $q \to 0$. This is the inescapable consequence of particle number conservation. However for the Mott-Hubbard systems, the above argument does not apply since $\tau_{q\to 0}^{xx}$ does *not* commute with the full Hamiltonian and *can* cause scattering between the exact eigenstates of H.

The effect of non-parabolicity of the quasiparticle bands (i.e. $\tau_q^{xx} \neq \rho_q$) was discussed by P. Wolff[17] who found that $I_{xx}(q,\omega) \neq 0$ for $\omega < qv_F$ the latter constraint being the consequence of limited phase-space available for scattering the quasiparticle. Here we point out that for the Mott-Hubbard system the scattering is actually dominated by the incoherent part of the carrier spectral function which defeats the phase-space limitation and results in nonvanishing scattering intensity, $I_{xx}(q,\omega)$, over a broad range of $\omega = O(t)$. The lack of q dependence (for $q\to 0$) of the integrated intensity $= \int d\omega \, I_{xx}(q,\omega)$ will be one of the key consequences of our argument.

To proceed, let us rewrite Eqn. 28 in terms of the stress-stress correlation

$$I_{xx}(q,\omega) = \pi \, \mathrm{Im} \sum_{\nu,\mu} e^{-\beta\varepsilon_\mu} \frac{\langle \mu|\tau_{-q}^{xx}|\nu\rangle \langle \nu|\tau_q^{xx}|\mu\rangle}{(\varepsilon_\nu - \varepsilon_\mu - \omega) - i0^+} \tag{29}$$

The evaluation of this function can be done within perturbation theory using the expression

$$I_{xx}(q,\omega) = \mathrm{Im}\, \chi^{(3)}(q,\omega)/[1 - \exp(-\beta\omega)] \tag{30}$$

in terms of the generalized susceptibility which we write as

$$\chi^{(\alpha)}(q,\omega) = \frac{1}{N} \sum_{k,k'} \gamma_k^{(\alpha)} \gamma_k^{(\alpha)} L_{k,k'}(q,\omega) \, .$$

Here $\gamma_k^{(3)} = \partial^2 \varepsilon_k / \partial k_x^2$ is the bare vertex, and we define two more bare vertices $\gamma_k^{(1)} = 1$ and $\gamma_k^{(2)} = \partial\varepsilon_k/\partial kx$. Thus defined, $\chi^{(1)}$ and $\chi^{(2)}$ are measures of density-correlations and current correlations. The important function L is given by ($k \equiv (\vec{k}, k_o)$)

$$L_{k,k'}(\vec{q},\omega) \equiv \delta_{k,k'} A_k(q,\omega) + \frac{1}{N}(1-\delta_{k,k'}) B_{k,k'}(\vec{q},\omega)$$

with $A_k(q,\omega) = \int_{-\infty}^{+\infty} \frac{dk_o}{2\pi i} G(k+q/2) G(k-q/2)$ and $B_{k,k'} = \int_{-\infty}^{+\infty}\int_{-\infty}^{+\infty} \frac{dk_o}{(2\pi i)} \frac{dk'_o}{(2\pi i)}$ $\Gamma(k,k';q) G(k'+q/2)G(k'-q/2)$. Here G is the fully dressed one particle Green's function, and Γ is the two particle scattering amplitude. (The vertex functions can be defined as $\Lambda^{(\alpha)}(k,q)=\gamma_k^{(\alpha)} + tr_{k'} \Gamma(k,k';q) G(k'+q/2) G(k'-q/2)\gamma_k^{(\alpha)}$. These obey the usual particle hole singlet channel Bethe Salpeter equations for which Γ is the resolvent kernel and $tr_k \equiv \int \frac{dk_o}{(2\pi i)} \int \frac{d^dk}{(2\pi)^d}$.)

As stressed above, density fluctuations are necessarily suppressed at finite frequencies in the long wavelength limit and hence $\chi^{(1)}(0,\omega)$ must vanish for all nonzero ω. This is ensured by a Ward Takahashi identity, (with $q \equiv (0,\omega)$)

$$1 + tr_{k'} \Gamma(k,k';0,\omega) = G^{-1}(k+q/2) - G^{-1}(k-q/2) .$$

This identity is usually used as a bench test for conserving approximations and requires us to renormalize vertices appropriately with propagators. This implies that A and B as defined are *not* independent. We can expand B in terms of the invariants of the square lattice

$$B_{k,k'}(0,\omega) = \alpha(0,\omega) + \sum_j \sin k_j \sin k'_j \beta(0,\omega) + \sum_j \cos k_j \cos k'_j \gamma(0,\omega) + \cdots$$

Thus the Ward identity yields $\alpha(0,\omega) = -\frac{1}{N}\sum_k A_k(0,\omega)$. The other two susceptibilities are found as $\vec{q} \to 0$

$$\chi^{(3)}(0,\omega) = \left[\frac{4t^2}{N}\right] \sum_k (\cos^2 k_x) A_k(0,\omega) + t^2 \gamma(0,\omega) \qquad (31a)$$

and

$$\chi^{(2)}(0,\omega) = \frac{4t^2}{N} \sum_k (\sin^2 k_x) A_k(0,\omega) + t^2 \beta(0,\omega) \qquad (31b)$$

$\mathrm{Im}\frac{1}{\omega}\chi^{(2)}(0,\omega)$ is recognizable as the real part of the conductivity at frequency ω, (see Appendix) and $\mathrm{Im}\chi^{(3)}(0,\omega)$ is our result for the Raman intensity in the doped

material. It is clear that the two terms get contribution from a common term A which are related, but also contain independent contributions from β and γ. The energy scales of the function γ and β must, however, be similar. We are unable to compute these functions at this stage, but can provide a calculation of the common A term in order to give a crude estimate of the Raman intensity. Let us evaluate the "bubble diagram" with fully dressed Green's functions

$$A_k(q,\omega) = \mathrm{Im} \int \frac{d\omega'}{2\pi} G(\frac{q}{2} + k,\omega') \, G(\frac{q}{2} - k, \omega + \omega') \tag{32}$$

where

$$G(k,\omega) \equiv i \int dt e^{i\omega t} <T\{c_k^+(t)\, c_k(0)\}> = \int d\omega' \frac{a_k(\omega')}{\omega - \omega' + i(\omega' - \mu)\delta} \tag{33}$$

the propagator for the carriers is expressed in term of its spectral representation

$$a_k(\omega) = z_k \, \delta[\omega - \varepsilon_k] + \sigma_k(\omega) \tag{34}$$

which we broke up here into a coherent, z_k, and incoherent $\sigma_k(\omega)$, pieces.

Combining Eqn. 29, 30 one finds quite generally:

$$A_k(q,\Omega) = \int_{\mu-\Omega}^{\mu} d\omega \, a_{k-\frac{q}{2}}(\omega) \, a_{k+\frac{q}{2}}(\Omega+\omega) \tag{35}$$

The coherent contribution has the form

$$A_k^{coh}(q,\Omega) = z_{k+\frac{q}{2}} \, z_{k-\frac{q}{2}} \, n_{k-\frac{q}{2}}(1-n_{k+\frac{q}{2}}) \, \delta(\Omega + \varepsilon_{k-\frac{q}{2}} - \varepsilon_{k+\frac{q}{2}})$$

which for the free electron band $\varepsilon_k = k^2/2m$ becomes

$$I_{xx}^{coh}(q,\Omega) = 4m \, \Omega \left[(v_F q)^2 - \Omega^2\right]^{-1/2} \theta(\Omega + v_F q)\theta(v_F q - \Omega) \tag{36}$$

implying that the integrated intensity $\int d\Omega \, I_{xx}^{coh}(q,\Omega) \sim k_F q$ and hence vanishes as $q \to 0$. This is the consequence of the phase space constraint as mentioned above. Including the incoherent part of the spectral function one finds for the $q \to 0$ limit

$$A_k^{inc}(q=0,\Omega) = \sum_k \tau_k^2 z_k[\theta(\mu - \varepsilon_k)\,\theta(\Omega + \varepsilon_k - \mu)\sigma_k(\Omega + \varepsilon_k)$$

$$+ \theta(\varepsilon_k - \mu)\theta(\Omega - \varepsilon_k + \mu)\sigma_k(\varepsilon_k - \Omega)]$$

$$+ \int_{\mu-\Omega}^{\mu} d\omega\, \sigma_k(\omega)\, \sigma_k(\Omega+\omega)$$

While the precise form of $I^{inc}(q=0,\Omega)$ depends on the details at the incoherent density of states for $\Omega << \mu$ one has:

$$A_k^{inc}(0,\Omega) \approx \Omega[\delta(\varepsilon_\mu - \mu)\, z_k\sigma_\mu(\mu) + \sigma_k^2(\mu)] \tag{37}$$

At this point one observes that if the incoherent spectral density vanishes at the Fermi energy, i.e. $\sigma_\mu(\mu) = 0$, as it must for the Landau Fermi liquid in order for the quasiparticle to have an infinite lifetime on the Fermi surface, $\Omega^{-1}A_k^{inc}(0,\Omega)$ also vanishes at $\Omega = 0$ at least for $T = 0$). In other words, the Landau liquid picture implies at $T = 0$ that the bubble contribution to the scattering vanishes for zero energy transfers as super linearly with Ω!

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 $t \le \Omega \le U$, 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. (19). The various moments can be calculated as higher commutators, and expressed as correlation functions.[18]

So far we have only dealt with the nonresonant contribution to scattering. In the doped case it is clear that the physics of the resonant term in Eq. (8) consist of two terms corresponding to whether the intermediate states $|\nu\rangle$ belong to the lower or upper Hubbard bands. (We will assume that ε_i and ε_f are in the LHB, and that $\omega_i \sim \omega_f \sim U$.) The reason of course is that the current operator now has matrix elements both within the

LHB and across the two bands. These new matrix elements are mirrored in the optical conductivity (see Appendix Eq. (A.2)) where the Re σ ($q = 0, \omega$), upon doping, would display a reduction in magnitude for $\omega \sim U$ (by a factor λ say) and an increase in weight at lower frequencies ω of $O(t)$, also at $\omega = 0$, so as to satisfy the optical f-sum rule.

We thus expect that $M_R^{\alpha\beta}$ in the doped case is decomposable into two pieces

$$M_R^{\alpha\beta} = A_R^{\alpha\beta} + B_R^{\alpha\beta}$$

where A and B correspond to restricting the intermediate states in the lower and upper Hubbard bands. The terms $B_R^{\alpha\beta}$ can be treated in a similar fashion as in the undoped case, at least for ω_i such that $U - \omega_i << t$, and the formal expressions are as in Eqs. (26), (27). We should, however, express a renormalization of the absolute values of the prefactors by an amount of the order of λ (referred to above) corresponding to a loss of optical spectral weight in the UHB. (The overall effect on Raman intensity would then be a reduction of $O(\lambda^2)$).

The terms $A_R^{\alpha\beta}$ are obtained by constraining $|\nu>$ to the LHB, and we show in the Appendix A, that these are of $O((t/\omega_i)^2)$ for diagonal elements A_R^{xx}, and of $O(t/\omega_i)$ for off diagonal elements, which measure inelastic time reversal breaking.

5. COMPARISON WITH EXPERIMENTS

Turning now to experiments, we first discuss the insulating case. Recent experiments[1-3,19] show 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_2, intensities can be *larger* than B_{1g}. Within B_{1g} geometry, a reasonable understanding of the scattering has been reached by Singh *et al.*,[20] who calculated within the Heisenberg antiferromagnet, various frequency moments of the appropriate scattering operators and find that these match the experiments, providing an estimate of J. The fact that the scattering intensity in A_{1g} and B_{2g} geometries exceeds the 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 scattering operator that contributes to A_{1g} is different from J_2/J_1, it in fact, involves a different power of $t/(U-\omega_L)$ when such an expansion is possible. In general, when $U \sim \omega_L$, we have emphasized that the expansion fails, and in fact the scattering in A_{1g} or B_{2g} geometries is not limited by the B_{1g}. The forms of the spin operator that can

contribute to A_{1g}, in the case of $U \sim \omega_L$, involve arbitrarily long strings of spins — the upper Hubbard band propagator is turned into the scattering operator. Experiments have been done with laser frequencies of around 5000 Å corresponding to ~ 2.48 eV — this in fact corresponds fairly closely to the effective U in the 2-d cuprates.[2] Hence the range probed so far is also, interestingly enough, very close to the Coulomb energy, and hence we believe that the experiments are probing the most interesting and *difficult* region of Raman scattering (where an expansion in t/U is most unlikely to converge).

We have demonstrated explicitly that the scattering receives contribution from 4 distinct correlation functions corresponding to different representation of the 2D square lattice group. These correlation function can be determined by performing measurements with different scattering geometries as explained in Section II. Of particular interest is the A_2 channel which at finite frequency receives contribution from the fluctuations of the topological charge, $\vec{S}_i \cdot (\vec{S}_i \times S_k)$, which play a crucial role in a number of recently proposed theories of high-T_c materials.[11,12] Also, as shown in the Appendix the $\Omega \to 0$ limit of the A_2 scattering intensity is related to the off diagonal part of the dielectric tensor and could remain finite only if the time reversal symmetry were violated by the ground state. For the doped case our conclusions are drastically different from those of 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 $\mathrm{Re}\sigma_{xx}(0, \Omega) = \mathrm{Im}\chi^{(2)}(0, \Omega)/\Omega$. In the approximation where only the bubble contributions to $\mathrm{Re}\sigma$ are retained, these are essentially identical since $(\cos^2(k_x)) = 1 - (\sin^2(k_x))$. The optical experiments[22] on $YBa_2Cu_3O_7$ do seem to bear out, albeit crudely, this pseudoidentity,

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

for small enough Ω, although at higher frequencies there seem to be significant departures.[23] In the limit $\Omega \leq kT$, the above reduces to $I_{xx} \sim kT\, Re\sigma_{xx}$; together with a temperature-independent $I_{xx}(0,0)$, it implies a linear resistivity, which is 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[24] shows that this scenario is realized in the limit of high dimensions.

In conclusion we have presented a theory of Raman scattering in Mott Hubbard systems, examining the consequences of the Peierls form of coupling. With this point of view both the insulating and doped cases are described by effective scattering operator. We have shown in this paper that this point of view leads to radically different predictions for integrated scattering intensity for the doped cases that should be taken as hallmark of Mott Hubbard systems distinguishing these sharply from the nearly free electron gas.

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Appendix A: Quasielastic Limit of the Resonant Terms in Raman Scattering and Relationship to the Dielectric Function.

The contribution of the resonant terms in the doped case arising from intermediate states in the lower Hubbard bond has an interesting behavior depending sensitively on the polarization. Consider Eq. (8) with all momenta set at zero,

$$\langle f|\, A_R^{\alpha\beta}\,|i\rangle = \sum_{\nu\in LHB} \left[\frac{\langle f|j_\beta|\nu\rangle\,\langle\nu|j_\alpha|i\rangle}{\varepsilon_\nu - \varepsilon_i - \omega_i} - \frac{\langle f|j_\alpha|\nu\rangle\,\langle\nu|j_\beta|i\rangle}{\varepsilon_i - \varepsilon_\nu - \omega_f} \right] \quad (A.1)$$

For completeness and comparison we note that the real part of the optical conductivity ($q = 0$) tensor is given by

$$\mathrm{Re}\,\sigma^{xx}(\omega) = \left[\frac{\pi e^2}{L^d}\right] \sum_{\nu\neq\mu} (e^{-\beta\varepsilon_\mu} - e^{-\beta\varepsilon_\nu})\,\delta\,(\omega^2 - (\varepsilon_\nu - \varepsilon_\mu)^2)\;|\langle\mu|\,j_x\,|\nu\rangle|^2 \quad (A.2)$$

and

$$\mathrm{Re}\,\sigma^{xy}(\omega) = \frac{1}{\sigma} \sum_{\nu\neq\mu} (e^{-\beta\varepsilon_\mu} - e^{-\beta\varepsilon_\nu})\,P\,\frac{1}{(\varepsilon_\nu - \varepsilon_\mu)^2 - \hbar^2\omega^2}$$

$$\times\,[\langle\mu|j^x|\nu\rangle\,\langle\nu|j^y\,|\mu\rangle - \langle\mu|\,j^y|\,\nu\rangle\,\langle\nu|\,j^x|\,\mu\rangle] \quad (A.3)$$

In the insulating case the intermediate states in (A.1) ε_ν (UHB) are at an energy of $O(U)$ above ε_i (LHB), and hence for $\omega_i \sim \omega_f \sim U$, the second term is negligible.

In the doped case the contribution from these LHB intermediate states makes an intensity distinction between M_R^{xx} and M_R^{yy}. The term M_R^{xx} is crudely $\sim \sum_\nu (j_x|\nu\rangle\,\langle\nu|j_x)\,\frac{t}{\omega_i^2}$ since $\varepsilon_\nu - \varepsilon_i \sim O(t) \ll \omega_i \sim O(U)$. On the other hand the off diagonal term

$$\langle f|\, M_R^{xy}\,|i\rangle \simeq \frac{1}{\omega_i} \sum_\nu (\langle f|j_y\,|\nu\rangle\,\langle\nu|j_x\,|i\rangle - \langle f|\,j_x\,|\nu\rangle\,\langle\nu|j_y|i\rangle)$$

is larger. In fact at $T = 0$ in the quasielastic limit $|f\rangle \to |i\rangle$, this reduces to approximately $\sim \int_0^U \mathrm{Re}\,[\sigma^{xy}(\omega) - \sigma^{yx}(\omega)]\;d\omega$ (see Eq. (A.3)).

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