Raman Response in Doped Antiferromagnets

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Abstract

The resonant part of the $B_{1g}$ electronic Raman scattering response is calculated within the $t-J$ model on a planar lattice as a function of temperature and hole doping, using a finite-temperature diagonalization method for small systems. Results, directly applicable to experiments on cuprates, reveal on doping a very pronounced increase of the width of the two-magnon Raman peak, accompanied by a decrease of the total intensity. At the same time the peak position does not shift substantially in the underdoped regime.

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Proper understanding of electronic properties of cuprates, representing a remarkable example of strongly correlated systems, remains a challenge for theoreticians and experimentalists. One of very useful probes for the investigation of electronic excitations in these materials has been the Raman scattering. Using the latter method, it has been clearly established that in the reference insulating substance, e.g. in La$_2$CuO$_4$, the most pronounced electronic Raman processes at low temperatures correspond to the short-wavelength magnetic (two-magnon) excitations [1], which can be well described within the Heisenberg model for the planar antiferromagnet (AFM) [2].

A more general framework for the theoretical explanation of the Raman scattering in strongly correlated systems, and specifically in cuprates, has been first given within the Hubbard model, where the effective Raman operator for resonant and off-resonant conditions has been derived [3]. Nevertheless, due to the difficulties in performing the theoretical analysis and due to the possibility of other relevant processes, an agreement on the appropriate interpretation is still lacking for a number of Raman scattering results in cuprates.

One aspect concerns the pronounced temperature dependence of the linewidth in the undoped AFM [4]. The latter has been attributed to the phonon-induced broadening [5], but an important role could be as well played by higher-order resonant processes [6], becoming relevant in the resonant-Raman conditions.

Another problem is the doping dependence on the Raman scattering [7,8]. Recent experiments, performed on YBaCuO materials in the resonant regime [8], show a dramatic increase of the broadening of the two-magnon peak with doping, so that spectra appear essentially flat in the normal phase \( T > T_c \) when approaching the ‘optimum’ doping cases with highest \( T_c \). At the same time, the peak position does not move appreciably.

The aim of the present paper is to analyse the influence of doping and finite temperatures on the Raman spectra within the framework of the one-band \( t-J \) model, assuming that the dominant resonant contribution remains of the Loudon-Fleury type due to the spin exchange [9]. Assuming the Raman operator we calculate the spectra at various hole concentrations \( c_h \) and temperatures \( T \) by employing the recently introduced finite-T diagonalization method.
for small correlated systems [10]. $T > 0$ results are of interest by themselves. However, in
the case of Raman spectra, where quite broad features are expected particularly in doped
systems, using finite $T > 0$ mainly represents a technical advantage to obtain macroscopic-
like spectra even for small systems, as already established in previous applications of the
method to the evaluation of optical conductivity and dynamical spin correlations [11]. The
improvement over the usual $T = 0$ Lanczos method can be judged by comparing our results
with previous Raman spectra obtained within the $t - J$ model containing few mobile holes
[12]. While some aspects appear similar (e.g. lowest frequency moments), actual spectra
calculated at $T = 0$ from the ground state are dominated by few peaks, which are clearly
size-dependent.

One of the most studied prototype models of correlated systems, and specifically of the
low-energy properties of cuprates, is the $t - J$ model [13]

$$H = -t \sum_{\langle ij \rangle} (c_{js}^\dagger c_{is} + \text{H.c.}) + J \sum_{\langle ij \rangle} (\vec{S}_i \cdot \vec{S}_j - \frac{1}{4} n_i n_j),$$

(1)

where $c_{is}^\dagger (c_{is})$ are projected fermionic operators, prohibiting double occupancy of sites. The
ground state of the model has been studied both by analytical [13] and numerical methods
[14]. Recent investigations of the finite-$T$ dynamical spin and charge response [11] established
also qualitative (as well as quantitative) contact with anomalous normal-state properties of
cuprates, confirming the $t - J$ model as the proper framework for studies of the low-energy
physics for such doped AFM.

The operator, relevant for the resonant Raman scattering in cuprates, cannot be deter-
mined uniquely within the $t - J$ model (in contrast to operators for some other charge and
spin response functions), since necessarily Raman processes involve higher resonant levels.
Here we adopt a view that more complete models for cuprates, e.g. the three-band model,
can be for processes of interest mapped onto an effective $t - U$ Hubbard model [15],[13], where
the upper Hubbard-band states now correspond to charge transfer excitations in cuprates.
Within the Hubbard model near half-filling the Raman-scattering operator has been derived
in the limit $t/U \ll 1$ [3], yielding the well known form for the Heisenberg AFM [3].
\[ R_1 = A \sum_{(ij)} (\mathbf{\epsilon}_{\text{inc}} \cdot \mathbf{r}_{ij}) (\mathbf{\epsilon}_{\text{sc}} \cdot \mathbf{r}_{ij}) (\mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4} n_in_j), \]

where \( \mathbf{\epsilon}_{\text{inc}}, \mathbf{\epsilon}_{\text{sc}} \) are the incident and the scattered electric-field vector directions, respectively, \( \mathbf{r}_{ij} \) is the vector connecting sites \( i \) and \( j \), and the amplitude factor \( A = 4t^2/(U - \omega_{\text{inc}}) \propto J \) incorporates the resonance at the incident-light frequency \( \omega_{\text{inc}} \sim U \).

For the undoped (insulating) case the operator \( R_1 \) is the only resonant term of the order \( t^2/U \), hence the dominant one, at least outside the resonance \( |U - \omega_{\text{inc}}| > t \). In the doped AFM additional processes of the same order of magnitude are possible. The latter correspond to the hopping term involving three neighboring sites, leading partly also to the next-neighbor hopping correction, i.e. to the \( t' \) term, within the \( t - t' - J \) model [13,15],

\[ R_2 = \frac{A}{4} \sum_{(i,j,k),ss'} (\mathbf{\epsilon}_{\text{inc}} \cdot \mathbf{r}_{ij}) (\mathbf{\epsilon}_{\text{sc}} \cdot \mathbf{r}_{jk}) c^\dagger_{ks'} c_{js'} c^\dagger_{js} c_{is}. \]

The contribution of such term clearly scales with the hole concentration (it vanishes at half filling), so it is a quantitative question whether it can become important in the relevant parameter regime of cuprates.

The Raman spectral function is given by

\[
I(\omega) = \frac{1}{\pi N} \text{Re} \int_0^\infty dt \ e^{i\omega t} \langle R(t)R(0) \rangle = \frac{1}{NZ} \sum_{n,m} e^{-\beta E_n} |\langle m|R|n \rangle|^2 \delta(\omega - E_m + E_n),
\]

where \( \omega = \omega_{\text{sc}} - \omega_{\text{inc}}, \beta = 1/T \) (we use furtheron \( k_B = \hbar = 1 \)), and \( Z \) is the partition function. Sums run over all eigenstates \( n, m \) with corresponding energies \( E_n, E_m \) and are size-normalized with \( 1/N \).

To calculate \( I(\omega) \) within the \( t - J \) model on a square lattice we study finite clusters with \( N = 16 - 20 \) sites. Via Eq.(4) this would be impossible with the method of full diagonalization [13], used previously to investigate the Raman response for the Heisenberg model with maximum lattice size \( N = 16 \). Instead, we employ the recently developed method for finite-\( T \) dynamical (and statical) correlation functions [10], based on the Lanczos iteration procedure combined with random sampling. The method is quite convenient for the analysis.
of the Raman response, in particular for doped systems, since quite broad spectral features and a weak \( T \)-dependence require a modest number of Lanczos steps \( M < 100 \) \cite{10,11}. In results presented below we thus reach systems with \( N_{st} \sim 400,000 \) states in the largest \( q,S_z \)-basis sector, while the sampling involves typically \( N_0 \sim 200 - 500 \) initial configurations. As discussed quite extensively in connection with previous applications, the spectra reveal a macroscopic-like behavior only at finite \( T > T^* \), where \( T^* \) is related to the low-energy level spacing and thus dependent on the system size and hole doping. In all calculations we will fix \( J/t = 0.3 \), as relevant for cuprates \cite{13}, where \( J \sim 950 \text{ cm}^{-1} \). For such parameters we typically reach \( T^* \sim 0.1 \) \( t = J/3 \) in the doped system with \( 0.1 < c_h < 0.3 \).

We restrict our analysis to the dominant \( B_{1g} \) scattering geometry with \( \vec{\epsilon}_{inc} = (\vec{\epsilon}_x + \vec{\epsilon}_y)/\sqrt{2} \) and \( \vec{\epsilon}_{sc} = (\vec{\epsilon}_x - \vec{\epsilon}_y)/\sqrt{2} \). Let us first discuss results for the undoped Heisenberg model. Here the low-energy levels are quite sparse and hence we reach only \( T^* \sim 0.5 \) \( J \) in the largest \( N = 20 \) system. In Fig. 1 we present the \( I(\omega) \) spectra for several \( T > T^* \). Consistent with \( T \sim 0 \) results \cite{2,4} and with the low-\( T \) experiments \cite{4} we observe a two-magnon peak at \( \omega \sim 3.3 \) \( J \), being quite narrow at low \( T < J \), and having the width limited by quantum fluctuations \cite{2}. From Fig. 1 it follows that the peak width starts to increase substantially only at higher \( T \sim J \), where a gradual transition to a broad featureless spectrum occurs. It should be also noted that in spite of large broadening the peak does not move with \( T \) up to \( T \sim 2 \) \( J \). Nevertheless, when considering undoped cuprates, such as \( \text{La}_2\text{CuO}_4 \) with \( J \sim 1400 \) \( \text{K} \), clearly other mechanisms have to be invoked to account for the observed pronounced \( T \)-dependent width at lower \( T \ll J \) \cite{3,8}.

We proceed to the doped systems, where we first consider only the exchange part \( R = R_1 \), Eq.(2). Here \( T \) seems to play a less essential role, provided that \( T^* < T < J \). Typically we observe for doped systems only a steady decrease of the intensity with \( T \), e.g. a \( \sim 20\% \) reduction in the interval \( T/J = 0.3 - 1.0 \) for \( c_h \sim 0.2 \), and \( \sim 30\% \) for \( c_h \sim 0.05 \). On the other hand, the dependence on doping is essential, as evident from Fig. 2, where we present spectra for various hole concentrations \( c_h = N_h/N \leq 0.25 \) at lowest \( T = 0.15 t > T^* \). Already the smallest possible (for available sizes \( N \)) finite doping \( c_h = 0.05 \) increases dramatically the
width of the two-magnon peak, and spectral features become overdamped on approaching
the ‘optimum’ doping $c_h \sim 0.2$. It is however remarkable, that the peak position does not
shift appreciably in the underdoped regime $c_h \lesssim 0.1$. Only for $c_h > 0.15$ the spectra change
to a broad central-peak form with a maximum at $\omega = 0$.

Another experimentally relevant quantity is the total spectral intensity $I_0 = \int_0^\infty I(\omega)d\omega$
and its variation with doping. At the same time, we evaluate the frequency moments $\langle \omega \rangle$,
$\langle \omega^2 \rangle$, and the spectral width $\sigma = \sqrt{\langle \omega^2 \rangle - \langle \omega \rangle^2}$, where the averages are calculated with
respect to the $I(\omega > 0)$ part of the spectra. Quantities on Fig. 3 are calculated within
various-size systems for $T = 0.5J$, where values should be quite close to the $T = 0$ results.
Results for finite $c_h > 0$ are presented for $N = 16 - 20$. For undoped system $N_h = 0$ we
include values for $N = 20$, as well as for $N = 26$ at $T = 0$. In the latter case, we get e.g. the
width $\sigma \sim 0.8J$, very close to analytical predictions $[2]$. The scattering of values in Fig. 3
for similar $c_h$ could be partly attributed to the error due to the restricted random sampling
$[10]$ for larger systems, while smaller systems at chosen $T \sim T^*$ could still suffer from finite
size effects.

As evident from Fig. 3, the total intensity $I_0$ decreases steadily but substantially with
doping, i.e. by a factor $\sim 3$ from $c_h = 0$ to $c_h = 0.25$. The average frequency $\langle \omega \rangle$ starts to
increase slowly only at $c_h > 0.15$. The enhancement can be attributed to the emergence of
high-frequency tails with $\omega > t$ in doped systems, whereas the peak position moves in the
opposite way. On the other hand, the spectral width $\sigma$ shows a more dramatic variation, in
particular at low doping $c_h < 0.1$.

Let us finally discuss the influence of the three-site hopping term $R_2$, Eq.(3). Our results
indicate that it is less important at low $T$ and in the concentration range of interest $c_h \leq 0.25$,
either calculated separately as $R = R_2$ or combined $R = R_1 + R_2$. When evaluated as $R = R_2$
we observe that $\Delta I(\omega)$ is essentially featureless up to $\omega \sim 4t$, and becoming larger only at
$T > J$, while at $T \sim T^*$ it amounts only to e.g. $0.2 I_0$ at $c_h = 2/16$. Taken into account
as $R = R_1 + R_2$, the largest corrections are at $c_h = 0.25$ as expected, i.e. $\delta I_0/I_0 \sim 0.3$.
Nevertheless, the most pronounced changes are not in the regime $\omega < 2t$ (spectra are even
flatter there), but rather in the large-\( \omega \) tails, leading to substantially increased \( \langle \omega \rangle \) (e.g. \( \sim 70\% \) for \( c_h \sim 0.25 \)) and \( \sigma \) at the ‘optimum’ doping. It seems that the second-neighbor-hopping processes are quite restricted at low \( T \), due to remaining short-range magnetic correlations, disappearing only at higher \( T > J \). It should be however noted that we did not introduce an analogous three-site term (or a simpler \( t' \) hopping term) in the \( t - J \) model, Eq.(1) \[13,15\], which possibly could lead to some quantitative modifications.

How can we interpret the above results for the \( B_{1g} \) Raman scattering? Up to the overdoped situation \( c_h \sim 0.3 \) the scattering seems to be dominated by the spin-exchange part, Eq.(2), since the latter appears to determine the low-energy fluctuations even at the ‘optimum’ doping \[11\]. Still, there are evident changes with doping. In an undoped AFM a well developed longer-range order (long-range at \( T = 0 \)) induces a sharp peak in \( I(\omega) \). In the underdoped regime \( c_h < 0.1 \) the reduced AFM correlation length does not shift the peak, but induces a large broadening and a reduction of the intensity. In this respect the doped system behaves quite similarly to an undoped AFM but at elevated \( T \sim J \), as one can conclude from the similarity of spectra on Figs. 1,2. For \( c_h > 0.1 \) the spin system seems to approach a totally incoherent one. Any coherence in the contribution of different bonds in Eq.(2) is eliminated, leading to a reduction of the intensity and a broad featureless spectrum with a central maximum at \( \omega = 0 \).

Finally let us comment on the relation of our results to experiments. A systematic resonant-Raman scattering study has been recently performed on a sequence of \( YBa_2Cu_3O_{6+x} \) materials and \( YBa_2Cu_4O_8 \) \[8\]. Although it is not straightforward to relate in this case the actual doping of \( CuO_2 \) layers with our model doping \( c_h \) (and consider also the possible role of \( CuO \) chains), it seems that experiments for the underdoped materials correspond well to our model results, both regarding the shape of the Raman spectra and their intensity variation with doping. On the other hand, entering the ‘optimum’-doping and the overdoped regime both experiments and the theory show consistently a nearly flat Raman response.

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FIGURES

FIG. 1. Raman intensity $I/A^2$ vs. $\omega/J$ for the undoped Heisenberg model at various $T$, as calculated for $N = 20$. An additional smoothening width $\Delta = 0.2 \, J$ is used at low $T$.

FIG. 2. $I/A^2$ vs. $\omega/J$ for the $t - J$ model at various hole concentrations $c_h$ and $T = 0.5 \, J$. The smoothening is $\Delta = 0.2 \, J$.

FIG. 3. Total Raman intensity $I_0$ (triangles), the average frequency $\langle \omega \rangle / J$ (circles), and the spectral width $\sigma / J$ (squares) vs. doping $c_h$ at $T = 0.15 \, t$, as evaluated for systems with $N = 16 - 20$. For $c_h = 0$ also $N = 26$ results at $T = 0$ are included. Lines are guides to the eye only.
