Temperature dependence of optical spectral weights in quarter-filled ladder systems

Markus Aichhorn¹,², Peter Horsch¹, Wolfgang von der Linden² and Mario Cuoco³

¹Max-Planck-Institut für Festkörperforschung, Heidenbergstr. 1, D-70569 Stuttgart, Germany
²Institut für Theoretische Physik, Technische Universität Graz, Petersgasse 16, A-8010 Graz, Austria
³I.N.F.M. di Salerno, Dip. Scienze Fisiche “E.R. Caianiello”, I-84081 Baronissi Salerno, Italy

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The temperature dependence of the integrated optical conductivity $I(T)$ reflects the changes of the kinetic energy as spin and charge correlations develop. It provides a unique way to explore experimentally the kinetic properties of strongly correlated systems. We calculated $I(T)$ in the frame of a $t$-$J$-$V$ model for ladder systems, like NaV$_2$O$_5$, and show that the measured strong $T$ dependence of $I(T)$ for NaV$_2$O$_5$ can be explained by the destruction of short range antiferromagnetic correlations. Thus $I(T)$ provides detailed information about super-exchange and magnetic energy scales.

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For discrete lattice models, which are usually employed to describe strong correlation physics in transition metal oxides, the integrated optical conductivity (IOC) is directly proportional to the kinetic energy:\footnote{1}

$$I_{\alpha}(T) = \int_{0}^{\infty} d\omega \sigma_{\alpha}(\omega) \propto \langle -H_{\text{kin},\alpha} \rangle,$$  \hspace{1cm} (1)\footnote{2}

which applies for finite temperatures. Eq. (1) is even more powerful, since it holds for different polarizations $\alpha$ individually. This option is particularly useful for anisotropic systems, like the quarter-filled ladder compounds NaV$_2$O$_5$ and LiV$_2$O$_5$ which show highly anisotropic optical spectra\footnote{3},\footnote{4}. Here the $T$-dependence of IOC provides insight into the interplay between kinetic energy and interactions.

Our work is motivated by a recent study of the optical properties of NaV$_2$O$_5$ by Presura et al.\footnote{5}, who observed a reduction of $I(T)$ by 12-14% between 4K and room temperature. They proposed a fitting formula for the optical conductivity (integrated up to 2.25 eV) $I(T)/I(0) = (1 - f \exp(-E_0/T))$ with $f \sim 0.35(0.47)$ and $E_0 \sim 286(370)$K for $a(b)$ polarization, respectively, predicting a reduction of almost 50% for $\sigma$-polarization at several hundred degrees centigrade. As the main absorption is near 1 eV one may wonder about the origin of such a dramatic change of kinetic energy at low temperatures.

A key feature of NaV$_2$O$_5$, closely related to its charge dynamics\footnote{6}, is the 3D charge ordering transition at 34K\footnote{7} which is accompanied by the opening of a spin gap\footnote{8}. The precise structure of the low-$T$ phase is still under dispute\footnote{9}. As discussed below, the charge fluctuations of a single ladder can be mapped onto the Ising model in a transverse field (IMTF)\footnote{10}. It has been pointed out in Refs.\footnote{11},\footnote{12} that the parameters for NaV$_2$O$_5$ are such that the IMTF is close to its quantum critical point. The corresponding soft charge excitations appear at $q_b = \pi$ and therefore do not directly contribute to $\sigma(\omega)$. Yet it was shown in Ref.\footnote{13} that when including spin degrees of freedom the soft charge excitations contribute a small absorption continuum in $\sigma_\alpha(\omega)$ within the charge gap, and may explain the anomalous absorption observed by Damascelli et al.\footnote{14}. Presura et al.\footnote{5} suggested that the low energy excitations at $q_b = \pi$ may also explain the 30 meV activation energy for charge transport\footnote{15}, which is surprisingly low in view of the 0.8 eV optical gap. Furthermore it was conjectured\footnote{16} that these excitations may cause the $T$-dependence of IOC’s and hence $E_0$ in Presura’s fit should measure the charge gap.

The picture which evolves from our calculations is different. The kinetic energy of the IMTF does not show any significant temperature dependence which could be attributed to $E_0$. The temperature scale of the variation of the kinetic energy is set by the bare Coulomb interaction. The $t$-$J$-$V$ model, however, in agreement with the experimental data, displays a dramatic decrease of kinetic energy in the range $0.2 J < T < J$, where $J$ is the exchange integral\footnote{17},\footnote{18} (order 0.1eV) of the effective 1D Heisenberg model which describes the spin dynamics of NaV$_2$O$_5$\footnote{19}. We attribute the large decrease of $I(T)$ to the destruction of short range AF correlations by thermal population of local triplet excitations. Thereby IOC’s allow to measure the super-exchange contribution to $E_{\text{kin}}$.

At quarter filling, NaV$_2$O$_5$ can be described by a $t$-$J$-$V$ model\footnote{20},\footnote{21}, as the hopping matrix elements $t_{ij}$ between vanadium $d_{xy}$ orbitals are small compared with the intra-atomic interaction ($U \sim 4$ eV):

$$H = -\sum_{\langle i,j \rangle,\sigma} t_{ij} \tilde{c}_{i,\sigma}^\dagger \tilde{c}_{j,\sigma} + H.c. + \sum_{\langle i,j \rangle} J_{ij} (\mathbf{S}_i \cdot \mathbf{S}_j - \frac{1}{4} n_i n_j) + \sum_{\langle i,j \rangle} V_{ij} n_i n_j,$$  \hspace{1cm} (2)\footnote{22}

Elimination of local double occupancies\footnote{23} yields the Heisenberg term with $J_{ij} = 4t_{ij}^2/U$ and constrained electron creation operators $\tilde{c}_{i,\sigma}^\dagger = c_{i,\sigma}^\dagger (1-n_{i,-\sigma})$, which enter the densities $n_i = \sum_{\sigma} c_{i,\sigma}^\dagger c_{i,\sigma}$ in the Coulomb repulsion ($V_{ij}$) between neighbors (Fig.1). $\mathbf{S}_i$ is the spin-$\frac{1}{2}$ operator at site $i$. The sums are over spin $\sigma = \uparrow, \downarrow$ and all...
neighbor bonds $\langle i,j \rangle$ on the trellis lattice, depicted in fig.1. Typical parameters values are included in Fig.3.

We begin with the investigation of charge fluctuations in the spinless case. Due to the interactions $V_{ij}$ this is a nontrivial problem, which can be simplified to a single ladder model without changing the essence of the problem, provided $V_{xy}$ and $t_{xy}$ are not too large. For large $U/|t_{ij}|$ and $V_{a}/|t_{ij}|$ the relevant subspace of one electron per rung can be represented by pseudospin operators $T_{r}$, where the eigenvalues $\pm \frac{1}{2}$ of $T_{r}^{z}$ correspond to the left/right position of the electron within the rung $r$, and one obtains the one-dimensional (1D) IMTF [13]:

$$H_{\text{ladder}} = -2t_{a} \sum_{r} T_{r}^{x} + 2V_{b} \sum_{r} T_{r}^{z}T_{r+1}^{z}. \quad (3)$$

Here the Ising interaction, due to Coulomb repulsion, favors zigzag charge correlations on the ladder, while the kinetic energy $E_{\text{kin}}(\propto t_{a})$ appears as a transverse field opposing these correlations.

The 1D IMTF is exactly solvable [13]. The kinetic energy, expressed in terms of the dimensionless parameter $h = 2t_{a}/V_{b}$, is given by

$$\frac{E_{\text{kin}}}{V_{b}} = \frac{h}{2\pi} \int_{0}^{\pi} dk \frac{\cos k + h}{\epsilon(k)} \tanh(\frac{\beta V_{b} \epsilon(k)}{2}), \quad (4)$$

where $\beta = 1/k_{B}T$ and $\epsilon(k) = \sqrt{h^{2} + 2h \cos k + 1}$ is the quasiparticle dispersion which becomes soft at $k = \pi$ as $h \rightarrow 1$ (i.e. $E_{0} = V_{b}\epsilon(\pi) = |2t_{a} - V_{b}|$).

Fig.2 shows the temperature dependence of the exact results for $E_{\text{kin}}$ and the pseudo-spin correlation functions (CF) $P_{r} = \frac{1}{N} \sum_{i} \langle T_{i}^{z}T_{i+r}^{z} \rangle$ for nearest and next-nearest neighbors ($r = 1,2$). Obviously, the temperature dependence of $E_{\text{kin}}$ and the CF's is dictated by the bare interaction $V_{b}$ as only T-scale, provided $t_{a} \gg V_{b}$. The CF's decrease gradually with increasing $h = 2t_{a}/V_{b}$, and vanish only in the limit $h \rightarrow \infty$. We note one peculiarity of the exact solution: for $h < 1$ the magnetization $\langle T_{r}^{z} \rangle \propto E_{\text{kin}}$ is not strictly monotonic in temperature. Here $\langle T_{r}^{z} \rangle$ profits from a decrease of $z$-correlations. In the high temperature limit one finds $E_{\text{kin}} \sim h/T$.

From Fig.2 we conclude that the charge-only model cannot explain the decline of $I(T)$ below room temperature. To study the problem including spin there are two alternatives: (i) the complete spin-pseudospin model [14,1] and (ii) the $t$-$J$-$V$ model which we shall choose.

For the calculation of the optical spectra and IOC’s for the $t$-$J$-$V$ model we employ, as in Ref. [1], the finite temperature Lanczos Technique developed by Jaklic and Prelovsek [20]. The exact diagonalization (ED) was performed for a $4 \times 4$ site system. As we are studying an insulator, we can restrict the calculation of $\sigma_{a}(\omega)$ to the finite frequency response given by the Kubo formula

$$\sigma_{a}(\omega) = \frac{1 - e^{-\beta \omega}}{\omega} \text{Re} \int_{0}^{\infty} d\tau e^{i\omega \tau} \langle j_{a}(\tau)j_{a} \rangle, \quad (5)$$

where $j_{a} = i \sum_{(i,j)\sigma} t_{ij}R_{\alpha}^{ij}(\hat{c}_{i,\sigma}^{\dagger}\hat{c}_{j,\sigma} - h c)$ is the $\alpha(=a, b)$ component of the current operator and $R_{\alpha}^{ij}$ denotes the vector connecting sites $i$ and $j$. In the following, energies and temperature are given in eV.

Fig.3 displays the temperature variation of $I(T)$ for the $t$-$J$-$V$ model for several sets of parameters on a log-log scale. The data show a strong decrease at low temperature, which is particularly pronounced for $b$-polarization, while the decrease of the kinetic energy at high temperatures ($T \gtrsim V_{b}$) is similar to that discussed for the pseudo-spin (charge only) model. The low-T variation is controlled by the magnetic exchange $J \propto t_{b}^{2}/V_{b}$ as can be seen by comparing the data for $t_{b} = 0.1$ and 0.2 eV. Furthermore, the results for $t_{xy} = 0$ and $t_{xy} = 0.15$ are similar (for $t_{b} = 0.2$ [2] which is consistent with the suppression of super-exchange related to $t_{xy}$, as discussed in Ref. [14].

The near-neighbor spin CF's $S_{1n}$ for $t_{b} = 0.2$ and $t_{xy} = 0.15$ are shown in Fig.4. The comparison with the kinetic energy for these parameters (open circles in Fig.3) clearly illustrates that the low temperature decrease in
$E_{\text{kin}}$ is due to the loss of short-range AF spin-correlations, whose $T$-variation is controlled by the magnetic exchange $J$. The largest spin CF’s at low temperatures are $S_{14}$ and $S_{15}$ consistent with zigzag charge correlations, i.e. large $C_{14}$ and $C_{15}$. As discussed along with Fig.2, the short-range charge correlations, however, exist up to higher temperatures, determined by $V_b$.

![FIG. 3. IOC for $a$- (dot-dashed) and $b$-polarization (solid and dashed lines). Results are given for both $t_{xy} = 0.15$ (open) and $t_{xy} = 0$ (filled symbols). Common parameters are $t_a = 0.4$, $V_a = V_b = 0.8$ and $V_{xy} = 0.9$ eV. The set $t_b = 0.2$ and $t_{xy} = 0.15$ corresponds to spectra shown in Ref. Eq.[3].](image)

![FIG. 4. Spin- $S_{1n} = \langle S_1^n S_2^n \rangle$ (top) and charge-correlations $C_{1n} = \langle n_1^n n_2^n \rangle$ (bottom), (see Fig.1), as function of temperature ($t_b = 0.2, t_{xy} = 0.15$).](image)

Finally, the smallness of $C_{12}$ below $T \lesssim 0.4$ supports the validity of the pseudo-spin model, where only one electron per rung is allowed for. We also note that the data for $C_{1n}$ are consistent with the pseudo-spin correlations $P_r$, shown in Fig. 2.

We emphasize that the kinetic energy is essentially determined by short-range spin-correlations. In the insulating state $E_{kin}$ has two major contributions which lead to the two distinct T-scales in Fig.3: (i) valence fluctuations, which have been discussed already in the context of the pseudo-spin model, and (ii) virtual excitations, which give rise to magnetic super-exchange whose $T$-variation is controlled by $J$.

To understand the low-T behavior of IOC’s it is revealing to study a two-rung system including all terms in Eq.6, except $t_{xy}$ and $V_{xy}$, and to employ the spectral representation of $I(T)$ (eq.7):

$$I(T) = \frac{\pi}{Z} \sum_n e^{-\beta E_n} \sum_{m \neq n} \frac{1 - e^{-\beta(E_m - E_n)}}{E_m - E_n} |\langle m| j_\alpha | n \rangle|^2 \tag{6}$$

where the summation is over eigenstates of $H$ and $Z$ is the partition function. The eigenstates are characterized by $(S, p_{\alpha}, p_b)$, i.e. total spin $(S = \text{singlet/triplet})$ and parity $p_{\alpha/b}$ in $a/b$ direction. The level scheme is sketched in Fig.5. The ground state $(s, +, +)$ is a fully symmetric singlet. The lowest excited states are the triplet states $(t, +, -)$ and $(t, -, +)$, where $(t, +, -)$ is lower in energy, as it is composed of bonding and anti-bonding orbitals in $a$-direction, while $(t, -, +)$ consists of orbitals in $b$-direction and lies already $\approx 400$ meV above the ground state. Its thermal population in eq.6 can be neglected at low $T$. The difference between the two polarizations is due to the fact that the current operators $j_\alpha$ conserve the symmetries $(S, p_{\alpha}, p_b)$, except for $p_{\alpha} \rightarrow - p_{\alpha}$. One finds that $j_b$ has non vanishing matrix elements between the ground state and the first excited singlet with $(s, +, -)$, whereas the matrix elements involving $(t, +, -)$ vanish. This has the important consequence that the triplet state $(t, +, -)$ contributes to $Z$ but not to the numerator of the Kubo formula (Eq.(6)), leading to

$$I_b(T) \sim \frac{1}{\Delta E_s} \frac{1}{1 + 3e^{-\beta \Delta E_{st}}} \tag{7}$$

Here the factor 3 reflects the degeneracy of the triplet and $\Delta E_s$ is the energy difference between the lowest singlet states. The situation is different for $a$-polarization, where also $(t, -, +)$ contributes, resulting in an extra term

$$I_a(T) \sim \frac{1}{\Delta E_s(1 + 3e^{-\beta \Delta E_{st}})} + 3\kappa \frac{1}{\Delta E_t(3 + 3e^{-\beta \Delta E_{st}})} \tag{8}$$

where $\kappa$ is a ratio of matrix elements. In both cases the low-T behavior is governed by local singlet-triplet excitations of energy $E_{st}$ and hence by the exchange integral $J(= E_{st})$ of the effective 1d Heisenberg model $H = J \sum S_i S_{i+1}$. The influence is however different for $a$- and $b$-polarization. The $T$-dependence via the optical excitation energies $\Delta E_s$ and $\Delta E_t$ ($\sim 1$ eV) is marginal at low $T$. 

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We infer that also in extended systems, the low-
T behavior is due to local singlet-triplet excitations
with energy, estimated from the two-rung formula,
\[ E^{\infty}_{st} = 4t_0^2/[2(V_a + V_b)\sqrt{1 + 4(t_a/V_b)^2}] \]
for \( V_a, V_b \gg t_a, t_b \). For the standard parameters and \( t_b = 0.2 \), \( E^{\infty}_{st} \approx 70 \text{ meV} \).

As the results of the two-rung system are mainly
determined by symmetry arguments and energy-scale sepa-
ration, we consider the expressions in Eqs.(7,8) as generic
and \( \Delta E \) and \( \kappa \) determined by symmetry arguments and energy-scale separa-
tion, the two-rung model. For \( \kappa \) from Fig.3 (symbols). Inset: sketch of optical excitations .

experiments, e.g. from the T-dependence of the spectra and to explore
the changes of the kinetic (and super-exchange) energy
in correlated systems (particularly in systems with small
\( J \)), which cannot be achieved otherwise.

A similar fit of the experimental data in \[ \frac{I(T)}{I(0)} \]
\( J \approx 39 \text{ meV} \), consistent with values obtained from other
experiments, e.g. \( J \approx 48.2 \text{ meV} \) and 37.9 meV. According to \( E^{\infty}_{st} \), we estimate \( t_b \approx 1.5 \), like in [13,15] instead of 0.2 eV used in this study and in Ref. [11].

Finally one may wonder about the change of \( E_{kin} \) at
the 34 K transition [6]. As this charge order transition indicates
the onset of 3D long-range order, there is not neces-
sarily a large change of short range correlations to be ex-
pected. This view is consistent with the small change
of \( I(T) \) at 34 K in Presura’s data.

In conclusion, we have shown, based on the exact
CF’s of the IMTF, i.e. ignoring spin degrees of free-
dom, that the low energy excitations associated with the (charge)
correlations, do not lead to a substantial T-dependence of IOC’s. However, for the quarter-filled
\( t-J-V \) model we find, in agreement with experimental
data, a large decrease of IOC’s in the temperature range
0.2 \( J < T < J \). This change in kinetic energy is magnetic
in origin, and is explained by the destruction of short-
range spin-correlations as \( T \) is increased. We argue there-
fore, that the observed T-dependence of optical spectral
weights in NaV₂O₅ is explained by the decrease of the
magnetic super-exchange. This underlines the potential
of optical experiments to infer the magnetic exchange
\( J \) from the T-dependence of the spectra and to explore
the changes of the kinetic (and super-exchange) energy
in correlated systems (particularly in systems with small
\( J \)), which cannot be achieved otherwise.

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