Temperature Dependence of Optical Spectral Weights in α'–NaV_2O_5

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Introduction

The low dimensional transition metal oxide α'–NaV_2O_5 has attracted considerable attentation during the past few years because of its very fascinating properties. One key feature of this compound is the 3D charge ordering transition at $T_c = 34\,\mathrm{K}$ which is accompanied by the opening of a spin gap. Our work is motivated by a recent study of the optical properties of α'–NaV_2O_5 by Presura *et al.*[2], who observed a reduction of the integrated optical conductivity (IOC) by 12-14% between 34 K and room temperature. In addition they found a small reduction of the IOC below the phase transition temperature for a polarisation, whereas this reduction is not observed for b polarisation.

Presura *et al.* claimed that the decline of the IOC between T_c and room temperature could be due to charge excitations appearing at $q_b = \pi$ in the Ising model in a transverse field (IMTF), which the charge fluctuations of a single ladder can be mapped onto. In this work we investigate this assumption as well as the several possible scenarios for the low–temperature phase.

Extended Hubbard Model (t-U-V)

$$H = -\sum_{\langle ij\rangle,\sigma} t_{ij}(c^\dagger_{i\sigma}c_{j\sigma}+\mathrm{h.c.})+\sum_{\langle ij\rangle} V_{ij}n_in_j+U\sum_i n_{i\downarrow}n_{i\uparrow} \quad (1)$$

As the t_{ij} are small compared to U ($\approx 4.0\,\mathrm{eV}$) one can eliminate local double occupancies:

Extended t-J Model (t-J-V)

$$H = -\sum_{\langle ij\rangle,\sigma} t_{ij}(\tilde{c}^\dagger_{i\sigma}\tilde{c}_{j\sigma}+\mathrm{h.c.}) + \sum_{\langle ij\rangle} V_{ij}n_in_j + \sum_{\langle ij\rangle} J_{ij}(\mathbf{S}_i\cdot\mathbf{S}_j - \frac{1}{4}n_in_j) \quad (2)$$

with $J_{ij} = 4t_{ij}/U$ and $\tilde{c}^\dagger_{i\sigma} = c^\dagger_{i\sigma}(1-n_{i,-\sigma})$. The sums in equations (1) and (2) run over spin $\sigma = \downarrow,\uparrow$ and all bonds $\langle ij\rangle$ between next neighbours on a trellis lattice shown in fig. 1.

Optical Conductivity

Finite frequency response given by the Kubo formula

$$\sigma_\alpha(\omega) = \frac{1-e^{-\beta\omega}}{\omega}\mathrm{Re}\int_0^\infty d\tau e^{i\omega\tau}\langle j_\alpha(\tau)j_\alpha\rangle \quad (3)$$

with the current operator $j_\alpha = i\sum_{\langle ij\rangle,\sigma} t_{ij}R^{ij}_\alpha(c^\dagger_{i\sigma}c_{j\sigma} - \mathrm{h.c.})$ and R^{ij}_α the vector connecting sites i and j. The connection between the optical conductivity and the kinetic energy is given by the f-sum rule:

$$I_\alpha(T) = \int_0^\infty d\omega\sigma_\alpha(\omega) = \frac{\pi}{2N}\sum_{\boldsymbol{\delta}}\delta^2_\alpha\,\langle -H^{\boldsymbol{\delta}}_{\mathrm{kin}}\rangle. \quad (4)$$

with hopping-vector $\boldsymbol{\delta}$ and polarization axis α.

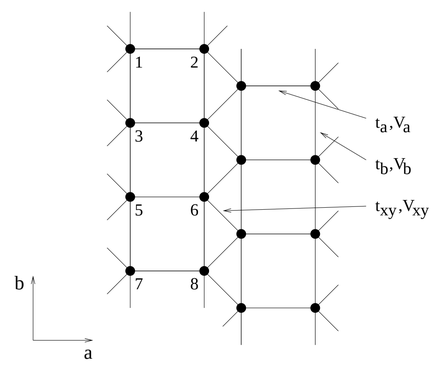

Fig. 1: Trellis lattice of vanadium ions in the (a,b)-plane.

$T > T_c$, Spinless case

Large $U/|t_{ij}|$ and $V_a/|t_{ij}|$: Relevant subspace represented by pseudospin operators $\mathbf{T}_r$. Simplification to single ladder gives the IMTF:

$$H_{ladder} = -2t_a\sum_r T^x_r + 2V_b\sum_r T^z_rT^z_{r+1} \quad (5)$$

Kinetic energy in terms of $h = 2t_a/V_b$:

$$\frac{E_{kin}}{V_b} = \frac{h}{2\pi}\int_0^\pi dk\frac{\cos k + h}{\epsilon(k)}\tanh\left(\frac{\beta V_b\epsilon(k)}{2}\right) \quad (6)$$

with $\epsilon(k) = \sqrt{h^2 + 2h\cos k + 1}$.

Fig. 2 does not show any decrease of E_{kin} at low temperatures
$\longrightarrow$ Charge-only model cannot explain the decline of the IOC
$\longrightarrow$ Spins have to be included.

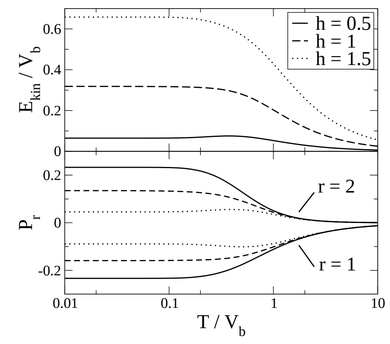

Fig. 2: Temperature dependence of the kinetic energy and pseudospin correlation function $P_r = \frac{1}{N}\sum_{r'}\langle T^z_rT^z_{r+r'}\rangle$ of the IMTF.

$T > T_c$ including Spins

Calculation of $\sigma_\alpha(\omega)$ for the t-J-V–model by finite temperature Lanczos technique on a 4×4 site system. Fig 3. shows strong decrease at low temperature, especially for b polarisation. From Fig 4. one can see that the decline of E_{kin} is due to the loss of short range AF spin correlations.

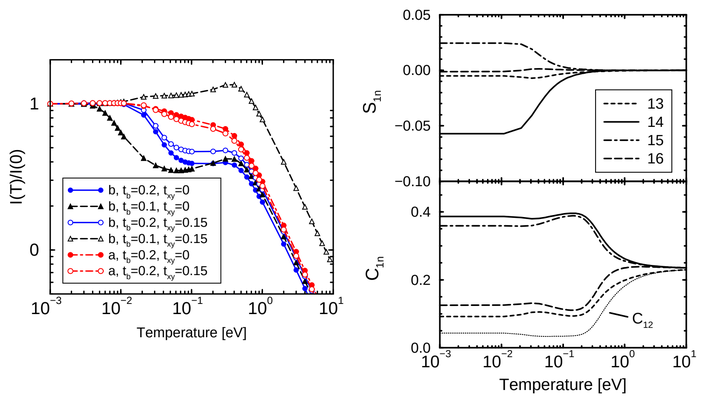

Fig. 3: Left panel: IOC for a- (dot-dashed) and b- polarisation for the t-J-V-model. Parameters are $t_a = 0.4$, $V_a = V_b = 0.8$, $V_{xy} = 0.9$ eV. Right panel:: Spin $S_{1n} = \langle S^z_1S^z_n\rangle$ (top) and charge correlations $C_{1n} = \langle n_1n_n\rangle$ (bottom), see fig. 1. Parameters as above with $t_b = 0.2$, $t_{xy} = 0.15$.

$T > T_c$, Two Rung System

Analytic evaluation of $I(T)$ on a 4 site systems gives formulas for the low temperature behaviour:

$$I_a(T) \propto \frac{1}{1+3e^{-\beta E^0_{st}}} + 3\kappa\frac{1}{3+e^{\beta E^0_{st}}} \quad (7)$$

$$I_b(T) \propto \frac{1}{1+3e^{-\beta E^0_{st}}} \quad (8)$$

For b-polarisation only the singlet channel contributes, whereas for a-polarisation both singlet and triplet channel have non-vanishing matrix elements.

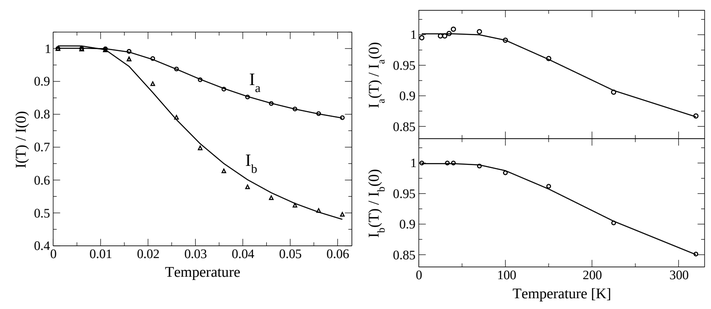

Fig. 4: Left panel: IOC for a and b direction for the numerical data. Fit gives $E^0_{st} = J = 76(61)\,\mathrm{meV}$ for $a(b)$ direction. Right panel: IOC for a (top) and b direction for the experimental data. Fit gives $E^0_{st} = J = 39(35)\,\mathrm{meV}$ for $a(b)$ direction consistent with other experiments.

Low Temperature Phase

Below T_c a dimerisation of the lattice takes place, accompanied by zig-zag charge ordering and slightly larger hopping amplitudes along the ladders, because bonding oxygen atoms are shifted[3]. We studied four possible scenarios:

- All ladders distorted $\leftrightarrow$ every second ladder distorted
- Without diagonal hopping t_D $\leftrightarrow$ With diagonal hopping t_D (t_D connects sites 1 and 4 and so on, see fig. 1). We induced the charge ordering by adding onsite energies favouring zig-zag order to the hamiltonian (1). In all cases we get a rather small charge transfer of $\Delta_c = 0.03\,\mathrm{eV}$ and slightly larger t_b (and t_D) in order to reproduce the experimental spectra. The change of the hopping amplitudes at T_c are in our calculations:
- Without t_D: $t_b = 0.2 \to 0.207$
- With t_D: $t_b = 0.08 \to 0.085, t_D = 0.095 \to 0.098$
- In both cases: $t_{xy} = 0.07 \to 0.08$

A larger charge transfer proposed in recent studies[4] does not lead to satisfying results.

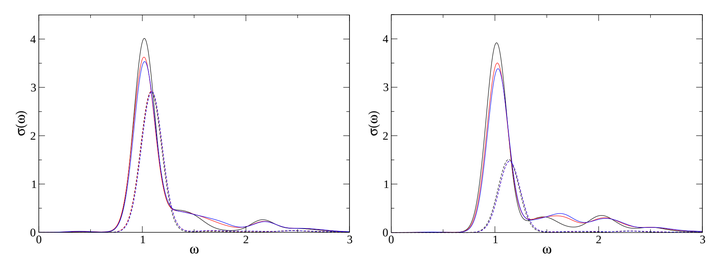

Fig. 5: Optical conductivity for the t-U-V model for a- (solid) and b direction (dashed lines). Black curve: $T \approx T_c$. Red curve: $T < T_c$ and all ladders distorted. Blue curve: $T < T_c$ and every second ladder distorted. Left panel: without diagonal hopping. Right panel: Including diagonal hopping t_D.

Conclusion

We have shown, based on the correlation functions of the IMTF, that the low energy charge excitations do not lead to a substantial T-dependence of the IOC's. However, for the quarter filled t-J-V model we find a large decrease of IOC's in the temperature range $0.2J < T < J$. This change in kinetic energy is magnetic in origin and can be explained by the destruction of short-range AF spin correlations.

In addition we found that the t-U-V model can describe the distorted low-temperature phase, too, provided charge transfer is small. From our calculations it is not possible to destinct between the two proposed charge order patterns, i.e. all ladders distorted (2 different V-sites)[4] or only every second ladder distorted (3 different V-sites)[3].

References

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