Band structure and questioned dimensionality of α'-NaV$_2$O$_5$

V. A. Ivanov[1], Z. V. Popovic[2], O. P. Khuong[2] and V. V. Moshchalkov[2]

[1] *Departement Natuurkunde, Universiteit Antwerpen (UIA)*
Universiteitsplein 1. B-2610 Antwerpen, Belgium
[2] *Departement Natuurkunde, Katholieke Universiteit Leuven*
Celestijnenlaan 200D, B-3001 Leuven, Belgium

Abstract. – α'-NaV$_2$O$_5$ is classified as a dimerized layered system with strongly interacting d-electrons of vanadium ions. The derived band gaps, energy dispersion relations and density of electronic states are in good agreement with available experimental and theoretical data. The correlated band gap provides the insulating state of the high-temperature phase whereas the state, earlier misinterpreted as the spin-Peierls state, is governed, in fact, by the opening of the Coulomb gap.

The spin-Peierls phase transition was first observed in one-dimensional organic salts [1]. Since its discovery in CuGeO$_3$ [2,3], there were a lot of efforts to find out a spin-Peierls behavior in other inorganic materials. In the best studied α'-phase of NaV$_2$O$_5$ from the AV$_n$O$_{2n+1}$ family (A stands for alkali or alkali earth element) the opening of a spin gap $\Delta_0 \sim 80$–110 K was observed at $T_c \approx 34$–36 K [4–6]. At present it has become evident that the spin-Peierls scenario cannot adequately describe the properties of the α'-NaV$_2$O$_5$ oxide. A summary of controversies is given in ref. [7].

The α'-NaV$_2$O$_5$ crystal with a $P_{mmn}(D_{2h}^{13})$ symmetry contains the quarter-filled dimers V$^{4+}(3d^1)$–V$^{5+}(3d^0)$ ($T < T_c$) or V$^{4,5+}$–V$^{4,5+}$ ($T > T_c$), forming rungs of two-leg ladders in the (a,b)-plane (fig. 1). Pyramides VO$_5$–VO$_5$ are arranged in layers made by vanadium ions and basal oxygens separated by layers formed by Na and apical oxygens. The crystal field lifts the t_{2g}-degeneracy of the vanadium $3d$-levels with the lowest energy of the d_{xy}-orbital. Theoretical models usually used for α'-NaV$_2$O$_5$ [8–11] are based on a spin-ladder picture. Local-density analysis [12,13] has shown that bands near the Fermi energy are constructed mainly from the d_{xy}-orbitals and revealed the pronounced peaks in the density of electronic states (DOS). But the computations could not establish the character of the band structure: does it have 1D, 2D or 3D features? According to refs. [12,13] the Fermi level lies inside the conducting band thus providing the metallic phase which is in disagreement with experimental observations. The one-electron character of the "first principles" methods cannot take properly into account the influence of many-body effects.

Our approach is based on the hypothesis that the α'-NaV$_2$O$_5$ properties are governed by the electron correlations U, t_a (intrarung electron hopping integral), t_b (hopping along legs in crystallographic b-direction), t_d (diagonal hopping), t_{xy} (interrung hopping between vanadium ions on nearest ladders) (fig. 1). The radial part of the d_{xy}-electron wave function allows to calculate hopping integrals as power series of $(r_B/a)^2$ [14]. The small parameter r_B/a (a is a lattice constant and r_B is an ionic radius) enables us to evaluate roughly the V-V hopping integral $t_{xy} \sim 0.06\,\text{eV}$ as an average value between 0.1 eV ($r_B = 0.98\,\text{Å}$ [8]) and 0.01 eV ($r_B = 0.83\,\text{Å}$). The infrared reflectance studies of α'-NaV$_2$O$_5$ [15, 16] are suitable to extract the intradimer hopping amplitude $t_a \simeq 0.35\,\text{eV}$. The enhanced values of the $t_{a,b,d}$ hopping integrals are influenced by intermediate oxygens. As for on-site interaction U, it is taken to be infinite and somewhat weaker intersite Coulomb interaction simply shifts the on-site electron energies in the charge-ordered phase.

In such a way, we classify the α'-NaV$_2$O$_5$ as a strongly correlated system and the present approach is based on the energy scale $U \gg t_a > t_b > t_d > t_{xy}$, included in the Hubbard-like Hamiltonian for vanadium d_{xy}-electrons:

$$H = U \sum_i N_i^\uparrow N_i^\downarrow + \sum_{\langle i,j \rangle} \Psi_i^+ \, \widehat{t_{ij}} \, \Psi_j, \tag{1}$$

where summation is over all unit cells. $\Psi^+ = (a^+, b^+, c^+, d^+, ...)$ and $N^\uparrow = (n_a, n_b, n_c, n_d, ...)$ are rows (just as Nambu spinors) with the k electron operators from a unit cell comprising k-sites, columns Ψ and $N^\downarrow$ include k operators also, $\widehat{t}$ is a k-by-k fold matrix of electron tunneling with elements $t_{a,b,d,xy}$ (see fig. 1). Then, one can carry out the fermion mapping to X-operators [17, 18] describing intravanadium transitions between the one-particle ground and an empty polar states. Applying the X-operator machinery [14, 19], one can derive *the tight-binding energy bands for strongly correlated electrons*. The applied technique considers the tunnelling part of any correlated Hamiltonian as perturbation with respect to electron correlations included in eigen values of the unperturbed part. The perturbation theory is based on the generalized Wick's theorem as an iteration procedure reducing the time-ordered product of n X-operators to the product of $n-1$ ones thereof. We consider first-order effects aiming at comparing the derived electron spectra with the conventional tight-binding results for non-interacting electrons which are done in the first order of the transfer energy. We concentrate on the influence of band structure, which is of significance for multicomponent compounds such as α'-NaV$_2$O$_5$.

The correlated energies $\xi(p)$ have been extracted from zeros of the inverse Green's function $D_{0\sigma}^{-1}(i\omega, p) = D_{0\sigma}^{(0)-1}(i\omega) + \Sigma(p, i\omega)$ (Dyson equation) for spin σ, where the first-order self-energy is the self-matrix of electron hopping: $\Sigma(p, i\omega) = \widehat{t}(p)$. The zero-th Green's functions are such as $D_{0\sigma,k}^{(0)}(i\omega) = f_k^\sigma \left(-i\omega_n + \varepsilon_k\right)^{-1}$, where ε_k labels electron energies on sites k of a unit cell. The expectation value $f_k^\sigma = \langle X_k^{\sigma\sigma} + X_k^{00} \rangle = 1 - n_k^{\overline{\sigma}}$ is said to be the correlation factor. It is governed by an electron density on site k and provides the band narrowing. The arrangement of dimers is closer to a triangular lattice and the calculations are based on the assumption that the V$_2$-rungs form an ideal triangular lattice. *Below (above) T_c the α'-NaV$_2$O$_5$ is in an ordered (mixed) valence phase.*

At $T < T_c$, vanadiums are packed in sublattices a, b, c, d, m, n, p, q (fig. 1). The unit cell Hamiltonian and $D_{0\sigma}^{(0)}(i\omega)$ include zigzag-ordered d_{xy}-electrons with on-site energy shifts $\varepsilon_{a,d,q,m} = -\varepsilon_{b,c,n,p} \equiv -\varepsilon$, influenced by neighboring Coulomb repulsion V: $\varepsilon = V\Delta n$ (Δn is the charge disproportionation on a rung V$^{4+\Delta n/5-\Delta n}$). At positions a, b, m, n (p, q, c, d) the d_{xy}-electrons have spin projections *down (up)*, respectively. *At any charge disproportionation,*
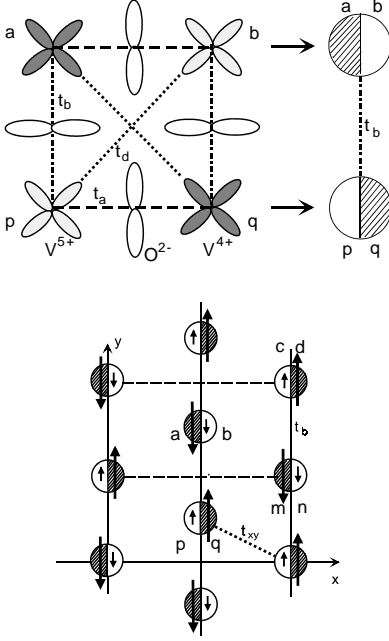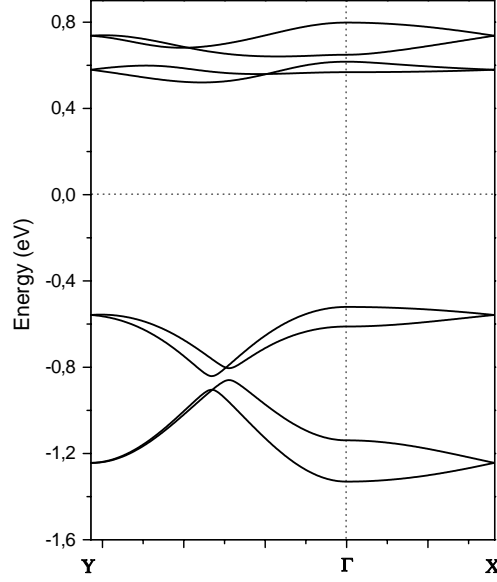
Fig. 1

Fig. 2

Fig. 1 – View of α'-NaV$_2$O$_5$. Each dimer/rung is replaced by a circle. The inter(intra)dimer hopping $t_b(t_a)$ in the b (a)-direction is set along the y (x)-axis. The distances at room temperature between the nearest V-ions on neighboring dimers/rungs are 3.04 Å and the leg constant is 3.61 Å. The dimer size is 3.44 Å. Oxygen p-wave functions (opened) enhance the hopping t_d along ladder diagonals. For $T > T_c$, the orthorhombic unit cell with two dimers is shown in the lower panel. For $T < T_c$, the size of spin arrows (lower panel) reflects the charge disproportionation $\Delta n = n_{a,d,m,q} - n_{b,c,n,p}$ in the monoclinic unit cell; the shaded portions have a zigzag order.

Fig. 2 – The tight-binding energy dispersions for correlated d_{xy}-electrons in α'-NaV$_2$O$_5$ *below* T_c for parameters $t_a = 0.35$ eV, $t_b = 0.15$ eV, $t_d = 0.1$ eV, $t_{xy} = 0.06$ eV and $\varepsilon = V\Delta n$ ($\Delta n = 0.8$, $V = 0.8$ eV [20]. Momenta are given in units $\left|p_x\sqrt{3}\right| = |p_y| = \pi$ of the Brillouine zone boundaries, the Fermi energy, $E_F = 0$, is inside the Coulomb gap $\Delta_C = 1$ eV. Four electrons from a monoclinic unit cell occupy bonding branches completely.

the correlation factors are $f_{a,b,m,n}^\downarrow = 1 - n_{a,b,m,n}^\uparrow = 1$ ($f_{p,q,c,d}^\uparrow = 1 - n_{p,q,c,d}^\downarrow = 1$) inasmuch as sublattices a, b, m, n (p, q, c, d) do not have electrons with spin projection *up*, $n_{a,b,m,n}^\uparrow = 0$ (*down*, $n_{p,q,c,d}^\downarrow = 0$). The perturbative tight-binding tunneling matrix is $\widehat{t}(\overrightarrow{p}) =$

$$
\begin{array}{c}
a^+ \\
b^+ \\
p^+ \\
q^+ \\
c^+ \\
d^+ \\
m^+ \\
n^+
\end{array}
\left(
\begin{array}{cccccccc}
0 & -t_a & B & D & 0 & A_1 & 0 & A \\
-t_a & 0 & D & B & -t_{xy} & 0 & -t_{xy} & 0 \\
B^* & D^* & 0 & -t_a & 0 & A_1 & 0 & A_1 \\
D^* & B^* & -t_a & 0 & C^* & 0 & -t_{xy} & 0 \\
0 & -t_{xy} & 0 & C & 0 & -t_a & B & D \\
A_1^* & 0 & A_1^* & 0 & -t_a & 0 & D & B \\
0 & -t_{xy} & 0 & -t_{xy} & B^* & D^* & 0 & -t_a \\
A^* & 0 & A_1^* & 0 & D^* & B^* & -t_a & 0
\end{array}
\right),
$$

with elements

$$A = -t_{xy} \exp\left[-i\left(\sqrt{3}p_x - p_y\right)\right], \qquad A_1 = -t_{xy} \exp\left[-i\left(\sqrt{3}p_x + p_y\right)\right],$$
$$B = -t_b\left(1 + \exp\left[2ip_y\right]\right), \qquad D = -t_d\left(1 + \exp\left[2ip_y\right]\right), \qquad C = -t_{xy}\exp\left[2ip_y\right].$$

At chosen values $t_a = 0.35\,\text{eV}$, $t_b = 0.15\,\text{eV}$, $t_d = 0.1\,\text{eV}$ we have established that for $t_{xy} \geq 48.8\,\text{meV}$ the electronic spectrum is gapless at $\varepsilon = 0$ and a repulsion V opens the insulating Coulomb gap Δ_C. The resulting energy dispersions are plotted in fig. 2. The flatness of an antibonding band is caused by the diagonal intraladder hopping t_d. The Δ_C is provided by the zigzag ordered energies, $\mp\varepsilon$, parameters $t_{a,xy}$ and competing interdimer hoppings $t_{b,d}$ (fig. 2). The Coulomb magnitude $V_c \sim 0.2\,\text{eV}$ [11] at which the zigzag charge order starts to develop, corresponds to our critical on-site energy shift $\varepsilon_c = 28.56\,\text{meV}$ (disproportionation $\Delta n = 0.14$) for $t_{xy} = 0.06\,\text{eV}$. Note that for these parameters the Coulomb gap value coincides with the critical temperature of the so-called "spin-Peierls" transition: $\Delta_C(\varepsilon_c) = 35\,\text{K}$. For such incomplete disproportionation the charge order is expected to dominate in an energy scale over a spin order. We have also established that a chain-type order for α'-NaV$_2$O$_5$, $\varepsilon_{a,p,c,m} = -\varepsilon_{b,q,n,d} \equiv -\varepsilon$ (cf. fig. 1), does not cause the Δ_C formation, to trigger the phase transition of interest.

For a single ladder ($t_{xy} = 0$) the energy dispersions have an analytical form:

$$\omega_\alpha = 2\alpha t_d \cos p_y \pm \sqrt{\varepsilon^2 + (t_a - 2\alpha t_b \cos p_y)^2} \quad (\alpha = +, -), \tag{2}$$

and at $\varepsilon > (t_a + 2t_b)\sqrt{t_b^2 - t_d^2}/t_d$ the Coulomb gap is

$$\Delta_C = \sqrt{\varepsilon^2 + (t_a + 2t_b)^2} + \sqrt{\varepsilon^2 + (t_a - 2t_b)^2} - 4t_d. \tag{3}$$

Equation (3) is the ladder extension of the "charged-magnon" scenario used for a single V^{4+}–V^{5+} rung ($t_{b,d} = 0$) in refs. [10,16].

At $T > T_c$ the α'-NaV$_2^{4.5+}$O$_5$ can be described by a half-filled Hubbard-like model for bonding electrons with two dimers/sites in an orthorhombic unit cell (fig. 1). For $U = \infty$ an effective Anderson-Hubbard parameter of this model is simply the gain of the intradimer kinetic energy $2t_a$. The electrons are distributed homogeneously with an electron density $n = 1$ per vanadium dimer and the correlation factors are $f_k^\sigma = 1 - n/2 = 1/2$ ($k = a - b, c - d$). Then the correlated-energy bands are formed by branches

$$\frac{\xi_p^\alpha}{t_b + t_d} = \varepsilon_p^\pm + \alpha\sqrt{\tau^2 + \left(\varepsilon_p^\pm\right)^2} \quad \left(\alpha = +, -; \ \tau = \frac{t_a}{t_b + t_d}\right), \tag{4}$$

with the calculated tight-binding non-correlated energies

$$\varepsilon_p^\pm = -\cos p_y \pm 2t \cos\frac{p_y}{2} \cos\frac{p_x\sqrt{3}}{2} \quad \left(t = \frac{t_{xy}}{2(t_b + t_d)}\right). \tag{5}$$

The lower, ξ_p^-, and the upper, ξ_p^+, bands are split due to the presence of two V$^{4.5+}$–V$^{4.5+}$ dimers in a unit cell (Davydov-like splitting). The energy dispersions, eq. (5), with the ranges $-1 - 2t \leq \varepsilon_p^- \leq 1$ and $-1 \leq \varepsilon_p^+ \leq 1/2 + t$ reflect the main peculiarities of the reported "spaghetti" pictures [12,13] rather well.
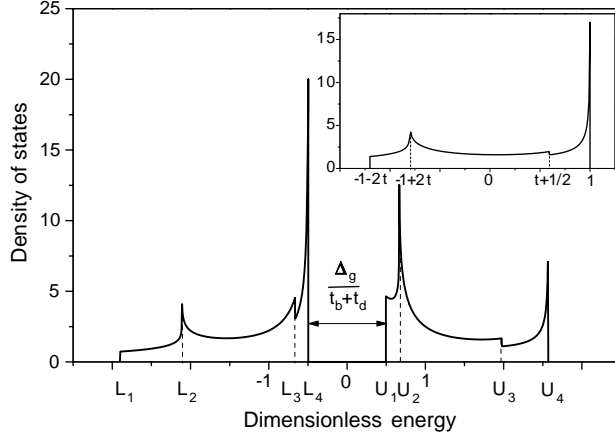
Fig. 3 – High-temperature $(T > T_c)$ DOS as a function of dimensionless energy $\xi/(t_b + t_d)$ (eq. (3)), $E_F = 0$ (main panel). The inset shows DOS for non-interacting bonding electrons as a function of dimensionless energy from eq. (4) (cf. [12]). For energies $L_1 = -1 - 2t - S + T_-$, $L_2 = -1 + 2t - S + P_-$, $L_3 = 1/2 + t - S + R_-$, $L_4 = 1 - S + Q_-$, $U_1 = -1 - 2t - S + T_+$, $U_2 = -1 + 2t - S + P_+$, $U_3 = 1/2 + t - S + R_+$, $U_4 = 1 - S + Q_+$ parameters S, $T_\pm$, $P_\pm$, $Q_\pm$, $R_\pm$ are given in eq. (6).

For dimensionless energies, $\omega_\alpha = \xi_p^\alpha/(t_b + t_d)$, the DOS is given analytically by

$$\rho^\alpha\left(-1 - 2t - S + T_\alpha \le \omega_\alpha \le -1 + 2t - S + P_\alpha\right) =$$

$$= \frac{1}{\pi^2 \sqrt{k_\alpha t}} \left[1 + \frac{q_\alpha^2}{4\left(\alpha\left|\omega\right| + S\right)^2}\right] K(q_\alpha), \tag{6}$$

$$\rho^\alpha\left(-1 + 2t - S + P_\alpha \le \omega_\alpha \le 1 + S + Q_\alpha\right) =$$

$$= \frac{1}{\pi^2 q_\alpha \sqrt{k_\alpha t}} \left[1 + \frac{q_\alpha^2}{4\left(\alpha\left|\omega\right| + S\right)^2}\right] \left[K\left(\frac{1}{q_\alpha}\right) \vartheta(1 - S + Q_\alpha - \omega_\alpha) +\right.$$

$$\left. + F\left(\arcsin\frac{1}{a_\alpha}; \frac{1}{q_\alpha}\right) \vartheta\left(\frac{1}{2} + t - S + R_\alpha\right)\right]. \tag{7}$$

Here ϑ is a Heaviside step function, $S = \sqrt{\tau^2 + (1 + 2t)^2} - \sqrt{\tau^2 + 1} - t$, $T_\alpha = \alpha\sqrt{\tau^2 + (1 + 2t)^2}$, $P_\alpha = \alpha\sqrt{\tau^2 + (1 - 2t)^2}$, $Q_\alpha = \alpha\sqrt{\tau^2 + 1}$, $R_\alpha = \alpha\sqrt{\tau^2 + (1/2 + t)^2}$ and the elliptic integrals F and K have modulus $q_\alpha = \sqrt{[2t(t + k_\alpha) + 1 - (\omega_0^\alpha)^2]/k_\alpha t}/2$ and argument $a_\alpha = \sqrt{k_\alpha(1 + \omega_0^\alpha)(t + k_\alpha)/[2t(t + k_\alpha) + 1 - (\omega_0^\alpha)^2]}$, with $k_\alpha = \sqrt{2(1 - \omega_0^\alpha) + t^2}$, $\omega_0^\alpha = [(\alpha\left|\omega\right| + S)^2 - \tau^2]/[2(\alpha\left|\omega\right| + S)]$. Equations are valid if the ε_p^+-band, eq. (5), is inside the ε_p^--band, i.e. at the realistic for α'-NaV$_2$O$_5$ constraint $t_{xy} < t_b + t_d$. The overlap of the energy bands, eqs. (4), (5), leads in fig. 3 to peculiarities in DOS at $L_3 = 1/2 + t + R_- - S$, $U_3 = 1/2 + t + R_+ - S$ in the main panel and at $1/2 + t$ in inset. *Logarithmic divergencies* at $\varepsilon = -1 + 2t$ and $\varepsilon = 1$ (inset), at $L_2 = -1 + 2t + P_- - S$ and $U_2 = -1 + 2t + P_+ - S$ (main panel) and *the peaks inside* the bands *are manifestations of 2D characteristics* in the electron structure of α'-NaV$_2$O$_5$ compound. In the limiting case of non-correlated electrons the total bandwidth

$4(t_b + t_{xy} + t_d) = 0.8\,\text{eV}$ coincides with the one in ref. [12] for the estimated hopping integrals $t_b = 0.15\,\text{eV}$, $t_{xy} = 0.06\,\text{eV}$ (note, in ref. [12] $t_d = 0$). The lower branches ξ_p^- (eq. (4)) are completely occupied by the two electrons from an orthorhombic unit cell (fig. 1) and for $t_a = 0.35\,\text{eV}$, $t_d = 0.1\,\text{eV}$ the correlated band gap (fig. 3)

$$\Delta_g = \min \xi_p^+ \left(\varepsilon_p^-\right) - \max \xi_p^- \left(\varepsilon_p^+\right)$$

$$= \sqrt{t_a^2 + (t_b + t_d)^2} + \sqrt{t_a^2 + (t_b + t_d + t_{xy})^2} - 2\left(t_b + t_d\right) - t_{xy} \tag{8}$$

acquires magnitude $0.34\,\text{eV}$.

In summary, the α'-NaV$_2$O$_5$ has been analyzed in the framework of the Hubbard-like model. The analysis of the derived dispersions (4)-(5) and DOS (6)-(7) leads to the conclusion about the presence of pronounced 2D features. At $T < T_c$ the zigzag order redistributes V$^{4+/5+}$ ions and it is accompanied by the opening of the Coulomb gap Δ_C (see fig. 2 and eq. (3)). Its estimated magnitude $\Delta_C \simeq 1\,\text{eV}$ corresponds to the observed strong absorption of light [16]. At $T > T_c$ the correlated band gap (8) provides an insulating state of α'-NaV$_2$O$_5$. It is worth noting that electron correlations are responsible for properties of other spin-ladder compounds: Sr$_{14}$Cu$_{24}$O$_{41}$ [21] and SrCuO$_2$ [22]. The studies, reported in ref. [23], give an experimental evidence of the clear semiconducting behaviour of α'-NaV$_2$O$_5$ below and above T_c with the increased dimensionality of an electron transport.

The electron energy dispersions (4) have two periodicities along the *b*-axis, in agreement with ARPES data in NaV$_2$O$_5$ [24]. The hopping parameters t_d, t_{xy} cause exchange *J*-terms, responsible for the splitting of magnon modes observed in inelastic neutron scattering [25–27]. If *J* constants are much smaller than electron hopping, the perturbative spin-dependent terms provide a spin-charge separation for 1D Hubbard or *t-J* models [28]. From that point of view this is an interesting interpretation of a temperature-induced modification of the ARPES spectral intensity as an evidence of availability of spinon and holon Fermi surfaces [24, 29]. However the subsequent spectral analysis [30] rules out the possibility of spin-charge separation in 2D *t-J* model (small *J*). Our study has revealed the importance of interladder couplings in α'-NaV$_2$O$_5$ leading to 2D characteristics in the band structure.

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