Simultaneous Charge Ordering and Spin Dimerization in Quasi-Two-Dimensional Quarter-Filled Ladders

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(Dated: March 22, 2022)

We study the spin-pseudospin Hamiltonian of the Ising Model in Transverse Field (IMTF) for pseudospins, coupled to the XY-spins on a triangular lattice. This model appears from analyses of the quarter-filled ladder compound NaV$_2$O$_5$, and pseudospins represent its charge degrees of freedom. In the molecular-field approximation we find that the model possesses two phases: charge-disordered without spin gap; and a low-temperature phase containing both the anti-ferroelectric (zigzag) charge order and spin dimerization (spin gap). The phase transition is of the second kind, and the calculated physical quantities are as those one expects from the Landau theory. One of particular features of the phase diagram is that the inter-ladder spin-pseudospin coupling, responsible for the spin gap generation, also destroys the IMTF quantum critical point, resulting in the exponential behavior of $T_c$ in the region of Ising’s coupling where the IMTF is always disordered. We conclude that our mean-field results give a qualitatively correct description of the phase transition in NaV$_2$O$_5$, while a more sophisticated analysis is warranted in order to take into account the thermal fluctuations and, probably, the proximity of the IMTF quantum critical point.

PACS numbers: 71.10.Fd, 71.10.Hf, 75.30.Et, 64.60.-i

I. INTRODUCTION

Properties of the models with coupled spin and orbital degrees of freedom have been actively studied from early 70-th, mostly in the context of the Jahn-Teller transition metal compounds. In recent years it has been a growing theoretical effort in order to understand the ground-state properties and excitation spectra of a particular (high-symmetric) class of one-dimensional spin-orbital models. The latter can also be viewed (up to some slight modifications at most) as two spin-Peierls class of one-dimensional spin-orbital models. Th e latter can also be viewed (up to some slight theoretical effort 70-th, mostly in the context of the Jahn-Teller transition metal compounds. As was re-emphasized recently, one of particular features of the phase diagram is that the inter-ladder spin-pseudospin coupling, responsible for the spin gap generation, also destroys the IMTF quantum critical point, resulting in the exponential behavior of $T_c$ in the region of Ising’s coupling where the IMTF is always disordered. We conclude that our mean-field results give a qualitatively correct description of the phase transition in NaV$_2$O$_5$, while a more sophisticated analysis is warranted in order to take into account the thermal fluctuations and, probably, the proximity of the IMTF quantum critical point.

It has been argued that the spin-gapped compounds Na$_2$Ti$_2$Pn$_2$O$_6$ and NaV$_2$O$_5$ can be modeled by a two-band quarter-filled Hubbard Hamiltonian. In the regime of strong on-site Coulomb repulsion such Hamiltonian can be mapped onto a spin-orbital model so the appearance of a spin gap is due to interplay between the charge and orbital degrees of freedom. Instead, one can work directly with the electronic Hamiltonian. Phase diagram of the 1D-spin-orbital models are known from mean-field analyses, renormalization group, bosonization, conformal field theory, numerics, and/or their combinations. As was re-emphasized recently from the view point of application to real materials we are mostly interested in, high symmetry of a spin-orbital Hamiltonian is rather artificial. Even if the total spin is conserved from physical grounds, i.e., the symmetry of the Hamiltonian’s spin sector is indeed SU(2), the symmetry of the orbital sector is usually lower (e.g., Ising or XY-type). So, applicability of the results obtained from high-symmetric Hamiltonians (e.g., SU(2)⊗SU(2)) to the cases of low-symmetry Hamiltonians following from microscopic considerations of a given problem, is not so straightforward.

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In this paper we will explore a particular scenario for the spin gap mechanism in a spin-orbit model, where the orbital (“pseudospin”) degree of freedom corresponds physically to the ordering charge displacement (disproportionation). Such a model appears in the applications to NaV$_2$O$_5$ and Na$_2$Ti$_2$Pn$_2$O$_6$. The quarter-filled ladder-system NaV$_2$O$_5$ is up-to-date the only well-established transition-metal compound with a localized spin-$\frac{1}{2}$ moment distributed, in the high-temperature phase, equally over two transition-metal ions in a mixed valence state. This unique situation of two V$^{4.5}$ ions sharing one spin $s = \frac{1}{2}$ leaves room for the charge disproportionation $2V^{4.5} \rightarrow V^4 + V^5$ occurring below the critical temperature of $T_c = 34$ K, together with the opening of a spin-gap (without a long-range order for spins). At $T_c$ the unit cell doubles both along the ladders (b-direction) and along the rungs (a-direction), see Ref. [21] for a review.
There have been several mechanisms proposed for the driving force of this phase transition: the spin-phonon coupling like in a spin-Peierls system\textsuperscript{22}, the electron-phonon coupling, the intra-ladder Coulomb repulsion between \( V^4 \) and \( V^5 \)-ions on the neighboring rungs\textsuperscript{22,23,24,25}, or a combination of those interaction terms\textsuperscript{26}. The experiments seem however to rule out\textsuperscript{21,22,25} the original proposal\textsuperscript{13} for the spin-Peierls scenario, while the zigzag charge ordering pattern predicted by the Coulomb-driven scenarios of this phase transition\textsuperscript{22,24,25} is now established crystallographically\textsuperscript{29}. Another argument in favor of the key role played by the charge ordering at this phase transition, comes from the fact that the coupling between spins and zigzag-ordered charge leads to\textsuperscript{27} the observed dispersion\textsuperscript{29} along the \( a \)-direction (perpendicular to the ladders) of the gapped magnon mode in the low-temperature phase.

Since \( \text{NaV}_2\text{O}_5 \) is an insulator, one can model the charge degrees of freedom in \( \text{NaV}_2\text{O}_5 \) on a given rung by a pseudospin \( \mathcal{S} \). The system of quarter-filled coupled ladders can be mapped onto a triangular lattice where one spin and one pseudospin reside on the same site\textsuperscript{22,25,26}, compare Fig. 1. Mostovoy and Khomskii in their study of \( \text{NaV}_2\text{O}_5 \) proposed the following mechanism for the spin dimerization driven by charge ordering (i.e., spin gap generation in the charge ordered phase)\textsuperscript{22,25} the antiferroelectric (AFE) in-ladder ordering (i.e., zigzag charge ordering) is due to the inter-rung Coulomb repulsion. The ordering pattern is shifted by a half of the inter-rung distance from ladder to ladder (see Fig. 1), and it results in alternation of the spin-exchange coupling along the ladder induced by its left and right neighbors. Then, as a consequence of the spin-exchange dimerization, the systems acquires a spin gap. Mostovoy and Khomskii\textsuperscript{22} also pointed out that for this effect to occur, it suffices for the dimerization term of the spin-exchange coupling (i.e, the inter-ladder spin-pseudospin coupling) to be linear over the charge order parameter. This mechanism resembles so the familiar spin-Peierls scenario, with the proviso that phonons are replaced by the charge order parameter.

Going deeper into this suggestion for the coupling which could be a key to the problem of \( \text{NaV}_2\text{O}_5 \), we perform a detailed analysis of such a phase transition starting from a simplest possible Hamiltonian compatible with symmetry restrictions. The effective Hamiltonian is that of the Ising Model in Transverse Field (IMTF) for pseudospins \( \mathcal{S} \), coupled to the spin-$\frac{1}{2}$ \( XY \)-chains. Since the phase transition in \( \text{NaV}_2\text{O}_5 \) is \textit{thermal} in nature, we treat the charge (pseudospin) sector of the Hamiltonian in the molecular-field approximation, while retaining in our analysis the experimentally observed 1D nature of the spin excitations. Recent x-ray\textsuperscript{31,32} and NMR\textsuperscript{33} experiments showed large fluctuation regions on the both sides of \( T_c \) in \( \text{NaV}_2\text{O}_5 \), and the order parameter critical index is found to be \( \beta \approx 0.17 \pm 0.19 \). It is close to \( \beta = 1/8 \) of the 2D Ising model, indicating along with the correlation lengths measurements\textsuperscript{31} on the 2D-Ising universality class of this phase transition. According to Ref.\textsuperscript{31} the 1D character of structural fluctuations manifests itself only at \( T \gtrsim 60K \). This allows us to expect the mean-field treatment to be a qualitatively reasonable approximation for the problem we are dealing with.

We find that our spin-pseudospin model has a phase diagram with two phases: the disordered one without charge order or spin gap; and the ordered low-temperature phase with both the anti-ferroelectric (zigzag) charge order and spin gap. One of our most important findings is that the model always orders (continuously, via a second-order transition) into that phase at low temperatures. The critical temperature of this phase transition shows various regimes of dependence on model’s parameters, but it never vanishes as far as the inter-ladder spin-pseudospin coupling is non-zero. Physically, it means that the role of the inter-ladder coupling is twofold: this coupling not only creates the spin gap when the phase transition is driven by the charge (pseudospin) ordering in the IMTF, but it also destroys the quantum critical point of the IMTF and generates the charge-ordered and spin-gapped phase in the case when charge ordering in the pure IMTF is impossible.

The rest of the paper is organized as follows. In Section II we define the model Hamiltonian and describe the methods employed to handle it. In Section III we present the mean-field equations which govern the critical behavior of the system. Section IV presents the analysis of the phase diagram, i.e., the critical temperature of the anti-ferroelectric phase transition as a function of the Hamiltonian’s couplings. The results for the order parameter, spin and pseudospin susceptibilities, specific heat capacity calculated in the vicinity of the anti-ferroelectric phase transition are given in Section V. The summary of our results and the discussion on their application for \( \text{NaV}_2\text{O}_5 \) are presented in the final Section VI.
II. MODEL AND FORMALISM

A. Model

We choose the following effective Hamiltonian:

$$H = -\Omega \sum_{m,n} T_{mn}^x + \frac{1}{2} J_1 \sum_{m,n} T_{mn}^x T_{m,n+1}^x + J_{II} \sum_{m,n} S_{mn} S_{m,n+1}$$  \hspace{1cm} (1)

$$+ \sum_{m,n} S_{mn} S_{m,n+1} \left[ J_{ST} T_{mn}^z T_{m,n+1}^z + \varepsilon \left( T_{m+1,n+1}^z - T_{m-1,n}^z \right) \right]$$

where the spin operators for a given site are defined as $S^i = \frac{1}{2} \sigma^i$ ($i = x, y, z$) via the Pauli matrices $\sigma^i$. The pseudospin operators are related in the same way to the Pauli matrices $\tau^i$: $T^i = \frac{1}{2} \tau^i$. We use the distinct notations for the Pauli matrices $\sigma, \tau$ in order to emphasize the fact that they operate in different spaces (spin, pseudospin). So the components, e.g., $\sigma^x$ and $\tau^x$ do not have the same quantization axis. By labelling the two sets of Pauli’s matrices $\sigma, \tau$ with $x, y, z$ we mean only their conventional representation and commutation relations. The subscript $1 \leq m \leq M$ stands for the number of ladder, while $1 \leq n \leq N$ numbers the rung. We assume the Hamiltonian coupling constants to be positive. Note in making comparison with the microscopic Hamiltonians derived for NaV$_2$O$_5$, we retain only linear term over the rung charge displacement operator $T^x$. This inter-ladder coupling term is allowed by the point symmetry group $D_{2h}$ (the NaV$_2$O$_5$ space group is $D_{2h}^{13} - Pmnnm$). Note that a similar intra-ladder spin-pseudospin coupling $\sum_{m,n} S_{mn} S_{m,n+1}(T_{m,n+1}^z - T_{m,n}^z)$ would be odd with respect to the mirror-plane through the bridging-oxygen (the $bc$-plane), therefore it is forbidden by symmetry.

In the following we will work with the dimensionless Hamiltonian $\mathcal{H} = H/\Omega$

$$\mathcal{H} = -\sum_{m,n} T_{mn}^x + \frac{1}{2} g \sum_{m,n} T_{mn}^x T_{m,n+1}^x$$  \hspace{1cm} (2)

$$+ \sum_{m,n} D_{mn} \left[ J + \lambda T_{mn}^z T_{m,n+1}^z + \varepsilon \left( T_{m+1,n+1}^z - T_{m-1,n}^z \right) \right] ,$$

dimensionless temperature $T \rightarrow T/\Omega$, and dimensionless couplings

$$g = J_1/\Omega \ , \ J = J_{II}/\Omega ,$$

$$\lambda = J_{ST}/\Omega \ , \ \varepsilon = \varepsilon/\Omega ,$$

where the dimerization operator $D_{mn} \equiv S_{mn} \cdot S_{m,n+1}$. For the applications we have in mind, the range of the model’s parameters under consideration will be restricted to the cases

$$(J, \lambda) \lesssim g \ , \ \varepsilon \ll (g, \max\{J, \lambda\})$$  \hspace{1cm} (4)

In the present study we consider the Hamiltonian with the $XY$ spin-spin interaction, i.e.,

$$D_{mn} = D_{mn}^{XY} = S_{mn}^x S_{m,n+1}^x + S_{mn}^y S_{m,n+1}^y$$  \hspace{1cm} (5)

$$= \frac{1}{2} \left( S_{mn}^+ S_{m,n+1}^- + S_{mn}^- S_{m,n+1}^+ \right)$$

where we used the conventional definition $S^{\pm} \equiv S^x \pm iS^y$.

B. Molecular Field Approximation

We proceed with the approximate treatment of our model in the following way: The model Hamiltonian (1) can be viewed as $\mathcal{H} = \mathcal{H}_{\text{IMTF}} + \mathcal{H}_{\text{XY}}$. $\mathcal{H}_{\text{IMTF}}$ is the Hamiltonian of the IMTF in terms of the pseudospin operators $T$ which is exactly solvable by its own. Dependence of the effective coupling $J_{\text{eff}}$ on $T$ in the $XY$ spin Hamiltonian $\mathcal{H}_{\text{XY}} = \sum_{mn} J_{mn}^{\text{eff}} D_{mn}$ precludes the total Hamiltonian from being exactly solvable.
We apply a version of the Mean Field Approximation (MFA) which is rather known as the Molecular Field method (Approximation) in the theory of phase transitions\textsuperscript{34}. This approximation assumes separability of the total Hamiltonian density matrix $\rho$ over $S$ and $T$, i.e.,

$$\rho = \rho^S \otimes \rho^T$$

and the following single particle (operator) ansatz for the pseudospin density matrix

$$\rho^T = \prod_{m,n} \rho_{mn}^T, \quad \rho_{mn}^T = \frac{1}{Z_{mn}^T} \exp(-\beta h_{mn} T_{mn})$$

Here $h_{mn}$ is the on-site vector of the molecular field to be defined self-consistently, $\beta$ is inverse temperature (we set the Boltzmann constant $k_B = 1$), and

$$Z_{mn}^T = 2 \cosh \frac{\beta |h_{mn}|}{2}$$

in the on-site partition function.

Since the independent spin-pseudospin averaging\textsuperscript{8} with the single-particle density matrix\textsuperscript{8} makes $J_{\text{eff}}$ a function of the average values of $T$, no further approximation is needed for the density matrix $\rho^S$. The statistical mechanical problem with

$$\rho^S = \frac{1}{Z_S} e^{-\beta \mathcal{H}_S}, \quad Z_S = \text{Tr} e^{-\beta \mathcal{H}_S},$$

$\mathcal{H}_S = \mathcal{H}_{XY}$ (here Tr includes only spin-operator states) is exactly solvable\textsuperscript{35} and will be dealt with in the next subsection. To summarize more qualitatively, we solve exactly the problem of independent spin $XY$ chains with their exchange constants determined by the molecular fields of quasi-two-dimensional pseudospins.

C. Spin Hamiltonian

The standard Jordan-Wigner transformation\textsuperscript{35,36} (JWT) when applied independently to one-dimensional spin operators on a given ladder, results in fermionic operators which commute on different ladders (see, e.g., Ref.\textsuperscript{37}). Strictly speaking this does not cause problems for our choice of Hamiltonian \textsuperscript{2}, since its spin part does not couple different ladders. However we find this situation with the 1D JWT somehow unpleasant, especially keeping in mind possible generalizations of the present Hamiltonian for future work. There are generalizations of the JWT for $d > 1$ available in the literature (see, e.g., Ref.\textsuperscript{38,39}).

We will employ a version of the 2D JWT proposed by Azzouz\textsuperscript{39}, particularly convenient for our model. We also include to the spin Hamiltonian a term generated by the uniform external magnetic field

$$\mathcal{H}_\text{ext} = \sum_{m,n} h_{\text{ext}} S_{mn}^z$$

The transformation is given by the following equations\textsuperscript{39}

$$S_{mn}^- = \exp(-i\pi \sum_{k,l \in \mathcal{A}_{mn}^+} n_{kl}) c_{mn} \quad (12a)$$

$$S_{mn}^+ = c_{mn}^\dagger \exp(i\pi \sum_{k,l \in \mathcal{A}_{mn}^-} n_{kl}) \quad (12b)$$

where $n_{mn} = c_{mn}^\dagger c_{mn}$ is the fermionic number operator, and the above summations include all rungs and ladders lying on the left from the $m$-th ladder plus rungs lying below $n$-th rung at the $m$-th ladder, i.e.,

$$k,l \in \mathcal{A}_{mn} \leftrightarrow \{1 \leq k \leq m-1, 1 \leq l \leq N\} \cup \{k = m, 1 \leq l \leq n-1\}$$

The fermionic operators satisfy the canonical anticommutation relations

$$\{c_{mn}^\dagger, c_{kl}\} = \delta_{mk} \delta_{nl} \quad (14a)$$

$$\{c_{mn}, c_{kl}\} = 0 \quad (14b)$$
and
\[
D_{mn}^{XY} = \frac{1}{2} (c_{mn}^\dagger c_{m,n+1} + c_{m,n+1}^\dagger c_{mn})
\]
\[
S_{mn}^z = c_{mn}^\dagger c_{mn} - \frac{1}{2}
\]  
(15)  
(16)  
For the effective coupling $J_{mn}^{\text{eff}}$ determined by the mean-field values of the pseudospin operators we take a dimerized ansatz, so
\[
\mathcal{H}_{XY}^{\text{MF}} = \sum_{m,n} J_{mn}^{\text{eff}} D_{mn}^{XY}, \quad J_{mn}^{\text{eff}} = A_m + (-1)^n B_m
\]
(17)  
Applying then the JWT defined above and a Bogolyubov transformation, we obtain
\[
\mathcal{H}_S \equiv \mathcal{H}_{XY}^{\text{MF}} + \mathcal{H}_{\text{ext}} = -\frac{1}{2} \mathcal{M} \mathcal{N} h_M + \sum_{m,q,\nu} E_{m\nu}(q) d_{mq\nu}^\dagger d_{mq\nu}
\]
(18)  
where the new fermionic operators $d_{mq\nu}$ also satisfy the canonical anticommutation relations, and their spectrum is
\[
E_{m\nu}(q) = h_M + \nu \sqrt{A_m^2 \cos^2 qa + B_m^2 \sin^2 qa}
\]
(19)  
The extra index $\nu = \pm 1$ is due to dimerization, i.e., doubling of fermion species, summation over $q$ includes the reduced Brillouin zone $[-\pi/2, \pi/2]$, and $a$ is the distance between rungs (we will set $a = 1$ in the following). For more details, see, e.g., Refs. [40,41]. The operator of the total $z$-component of spin per rung reads
\[
s_z \equiv \frac{1}{\mathcal{M} \mathcal{N}} \sum_{m,n} S_{mn}^z = -\frac{1}{2} + \frac{1}{\mathcal{M} \mathcal{N}} \sum_{m,q,\nu} d_{mq\nu}^\dagger d_{mq\nu}
\]
(20)  
The spin contribution to the free energy per rung from the Hamiltonian $\mathcal{H}_S$ (cf. Eqs. (10)) is
\[
f_S = -\frac{1}{\beta \mathcal{M} \mathcal{N}} \ln Z_S
\]
\[
= -\frac{\ln 2}{\beta} - \frac{1}{\pi \beta \mathcal{M}} \sum_{m,\nu} \int_0^{\pi} dq \ln \cosh \left( \frac{\beta E_{m\nu}(q)}{2} \right)
\]
(21)  

III. MEAN-FIELD EQUATIONS

The molecular fields are defined from the condition that they minimize the mean-field free energy $\mathcal{F} = \langle \mathcal{H} \rangle + T \langle \ln \rho \rangle$. The angular brackets stand for averaging with the mean-field density matrix (10), and the Hamiltonian $\mathcal{H}$ is given by Eq. (4). We consider the case when the external magnetic field $h_M$ is absent. (Note that contrary to $f_S$ (21), the quantity $f_T$ defined in the same fashion via $Z^T$ is not the pseudospin free energy).

We take the following ansatz for the Ising pseudospin magnetizations (i.e., the charge ordering parameters in terms of the real physical quantities)
\[
\langle T_{m,n}^z \rangle = m_z
\]
\[
\langle T_{m,n}^x \rangle = (-1)^{m+n} m_x
\]
(22)  
(23)  
So, similar to the case of the IMTF ($g > 0$) we assume the possibility of the anti-ferroelectric (AFE) in-ladder charge ordering (i.e., the zigzag ordering), alternating however from ladder to ladder. It is easy to see from the Hamiltonian (2) that ansatz (23) creates a dimerization in the spin sector, so a natural assumption for the dimerization operator average is
\[
\langle D_{mn} \rangle = -[t + (-1)^{m+n}\delta]
\]
(24)  
Then the average mean-field energy per rung in the thermodynamic limit $\mathcal{M} \mathcal{N} \rightarrow \infty$ is
\[
\mathcal{E}_{\text{MF}} \equiv \frac{\langle \mathcal{H} \rangle}{\mathcal{M} \mathcal{N}} = -m_z - \frac{1}{2} gm_z^2 - \left( Jt + \lambda t m_x^2 + 2 \varepsilon \delta m_x \right)
\]
(25)  

The molecular Weiss fields $h^z_{mn} = -h_z$, $h^x_{mn} = (1)^{m+n+1}h_x$, $J_{mn}^{\text{eff}} = a + (1)^{m+n}b$ are defined by the following equations:

\[
\begin{align*}
h^z_{mn} &= -\frac{\partial E_{\text{MF}}}{\partial m^z} = 1 + 2\lambda t m^z \\
h^x_{mn} &= -\frac{\partial E_{\text{MF}}}{\partial m^x} = g m^x + 2\varepsilon \delta \\
a &= -\frac{\partial E_{\text{MF}}}{\partial t} = J + \lambda m^2_z \\
b &= -\frac{\partial E_{\text{MF}}}{\partial \delta} = 2\varepsilon m^x
\end{align*}
\]

The order parameters \[ \text{obtained as partial derivatives of the corresponding quantities } f_S, f_T \text{ with respect to their conjugate Weiss fields } \] should be determined from the system of four coupled equations

\[
\begin{align*}
m^z &= \frac{1}{2} \frac{1 + 2\lambda t m^z}{h} \tanh \frac{\beta h}{2} \\
m^x &= \frac{m^x g + 2\varepsilon \eta}{2} \tanh \frac{\beta h}{2} \\
t &= \frac{1}{\pi} \int_0^{\pi/2} d\varphi \cos^2 \varphi \tan \beta \xi(\varphi) \equiv \frac{1}{\pi} t_n(\Delta, \tilde{\beta}) \\
\eta &= \frac{\Delta}{\pi m^x} \int_0^{\pi/2} d\varphi \sin^2 \varphi \tan \beta \xi(\varphi) \equiv \frac{\Delta}{\pi m^x} \eta_n(\Delta, \tilde{\beta})
\end{align*}
\]

where $h = \sqrt{h_z^2 + h_x^2}$ is the absolute value of the Ising molecular field, and instead of $\delta$ we use the new parameter $\eta$ \[ \equiv m^x \eta \] (28)

We also introduced the auxiliary parameters

\[
\begin{align*}
\xi(\varphi) &\equiv \sqrt{\cos^2 \varphi + \Delta^2 \sin^2 \varphi} \\
\Delta &= \frac{2\varepsilon m^x}{J + \lambda m^2_z} \\
\tilde{\beta} &= \frac{\beta}{2(J + \lambda m^2_z)}
\end{align*}
\]

The following useful relationship holds at any temperature

\[
m^x + m^z = \frac{1}{4} \tanh^2 \frac{\beta h}{2}
\]

Alternatively, the mean-field equations \[ \text{can be derived from the free energy (per rung, } h_M = 0) \]

\[
f = 2\lambda t m^z + \frac{1}{2} gm^x + 2\varepsilon \delta m^x - T \ln 4 - T \left( \ln \cosh \frac{\beta h}{2} + \frac{1}{\pi} \int_0^{\pi/2} d\varphi \ln \cosh \beta \xi(\varphi) \right),
\]

understood as $f(m^z, m^x, t, \delta)$, by minimization over its variables. \[ \text{[h is defined according to Eqs. (26).]} \] Thus, $f$ plays a role of the Landau functional, defining parameters $m^z, m^x, t, \delta$.

Before presenting the analysis of Eqs. (27), let us describe their main properties more qualitatively. The system of these four coupled equations determines four unknown mean-field parameters $m^z, m^x, t, \eta$ (and $\delta$) as functions of the effective Hamiltonian couplings and temperature. The first couple of Eqs. (27) in the case $\lambda = \varepsilon = 0$ is familiar from the mean-field treatment of the pure IMTF (see, e.g., Ref. [34]), and has some similar properties with the pure case even for non-zero $\lambda$ and $\varepsilon$. There is always a non-trivial solution $0 < m^z \leq \frac{1}{2}$ for the field-induced magnetization (the lower bound value $m^z = 0$ is attained in the limit $g \to \infty$). Below certain critical temperature $T_c$, a non-trivial solution $m^x \neq 0$ appears. $m^z$ varies continuously across $T_c$, contrary to the case of the IMTF ($\lambda = \varepsilon = 0$) when it
stays constant \((= 1/g)\) for all \(T < T_c\). Non-zero \(m_x\) results in generation of the alternating term \(b\) in the exchange interaction \(J_{\text{eff,1}}\) and the non-zero dimerization parameter \(\delta\). Consequence of \(b \neq 0\) is a spin gap. Note that in the disordered phase \((m_x = 0)\), Eq. (27) is an empty statement at \(T \neq 0\), since \(\delta = 0\) as well. The parameter \(t\) is non-critical: it is some continuous non-negative function.

At \(T \leq T_c\) one equation from the pair (27a,b) can be written in the form

\[
m_{z}^{-1} = g + \frac{4\varepsilon^2}{\pi (J + \lambda m_{z}^2)} \eta_n - \frac{2\lambda}{\pi} t_n, \quad T \leq T_c
\]

more convenient for further analyses.

### IV. CRITICAL TEMPERATURE

At \(T = T_c\) we have Eqs. (27) as

\[
m_z = \frac{1}{2} \tanh \frac{\beta_c}{2} \left( 1 + \frac{2\lambda m_z}{\pi} t_n \right),
\]

another equation for \(m_{z}\), and parameters \(t_n, \eta_n\) are given by Eqs. (27c,d) with \(\Delta = 0\). The latter two functions have the following expansions:

\[
t_n(0, x) \approx \begin{cases} \frac{\pi}{4} x \left( 1 - \frac{1}{4} x^2 \right) + \mathcal{O}(x^5), & x < 1 \\ \frac{1}{2} x^2 \frac{1}{24}, & x > 1 \end{cases}
\]

and

\[
\eta_n(0, x) \approx \begin{cases} \frac{\pi}{4} x \left( 1 - \frac{1}{12} x^2 \right) + \mathcal{O}(x^5), & x < 1 \\ \ln A x + \mathcal{O}(\frac{1}{x}), & x > 1 \end{cases}
\]

where \(\Lambda \equiv \frac{8}{\pi e^{1/\gamma}} \approx 1.6685\), and \(\gamma \approx 0.5772\) is Euler’s constant.

#### A. Case \(\varepsilon = 0\): re-entrance

In order to better understand the model, let us start with the analysis of the mean-field equations for the case when the dimerization coupling is absent, i.e, \(\varepsilon = 0\). Then from Eqs. (27,33) we find the critical temperature \(T_c \equiv 1/\beta_c\)

\[
T_c = \frac{m_z g}{\ln \left( 1 + \frac{2m_z}{1 - 2m_z} \right)}
\]

where \(m_z\) is the solution of the following equation

\[
m_{z}^{-1} = g - \frac{2\lambda}{\pi} t_n \left( 0, \frac{J + \lambda m_{z}^2}{2m_{z} g} \ln \left( 1 + \frac{2m_{z}}{1 - 2m_{z}} \right) \right).
\]

It can be shown from Eqs. (32,33) that for large Ising coupling \(g \gg g_\lambda\) where

\[
g_\lambda = 2 \left( 1 + \frac{\lambda}{\pi} \right)
\]

the critical temperature \(T_c \approx g/4\) is large , while \(m_z \approx 1/g\) is small. So, the behavior of these parameters is the same as in the pure IMTF. It can also be shown that at \(g = g_\lambda\) the critical temperature \(T_c\) becomes zero (\(m_z = 1/\pi\)). Thus \(g_\lambda\) plays a role of the renormalized (due to the coupling \(\lambda\)) mean-field critical point \(g^* = g_\lambda(\lambda = 0) = 2\) of the pure IMTF, where \(T_c\) vanishes and no ordering in \(m_z\) occurs for \(g < g^*\). The mean-field \(g^*\) is two times less than the value known for the exactly solvable 1D IMTF. For 2D and 3D IMTF the values of \(g^*\) are known only from approximations and/or numerics, but they are bigger then the mean-field predictions.

However, the role of \(\lambda\) is more subtle than a simple shift in the parameters comparatively to the pure IMTF. The critical line \(T_c(g)\) manifests re-entrant behavior in the region \(g \gtrsim g_\lambda\) or, to state it differently, the function \(g(T_c)\) is non-monotonic, contrary to the case \(\lambda = 0\). (See Fig. 2) Also, \(T_c(g_\lambda) = 0\) does not mean that \(g_\lambda\) is the minimal
coupling \( g_{\text{min}} \) at which \( m_x \)-ordering can occur. How pronounced the re-entrance is, i.e., the width of the re-entrance region

\[
g_{\text{min}} < g < g_{\lambda}, \tag{39}\]

depends on the relative values of model’s couplings \( J, \lambda, g \). A more detailed analysis of the re-entrance in this model will be presented in a separate paper. At this time, we say that any value of the coupling \( \lambda \neq 0 \) always generates the re-entrance on the phase diagram \( g(T_c) \), i.e., \( g_{\text{min}} \rightarrow g_{\lambda} \rightarrow 2 \) only when \( \lambda \rightarrow 0 \). Characteristic results of the numerical solution of our equations are given in Fig. 2. At \( J \ll \lambda \) this feature is pronounced most [the case \( J = 0, \lambda = 1 \) shown in this figure], while at \( J \sim \lambda \) it is more smeared [see the curve at \( J = \lambda = 1 \)]. Notice that when \( g \rightarrow g_{\lambda} \), the curve \( T_c(g) \) approaches zero normally to the \( g \)-axis.

More qualitatively, the origin of the re-entrance can be understood from the free energy \( f \) which is minimized by our mean-field equations. First two terms on the r.h.s. of Eq. (31) augment the free energy \( f \) with the increase of \( m_z \) and/or \( m_x \), while at the same time the last two terms on the r.h.s. of that equation, explicitly proportional to the temperature, decrease \( f \) via the parameters \( \beta, \Delta \). The interplay of this contributions to \( f \) involves, apart of the temperature, the couplings \( g, J, \lambda \). The latter possess a range [the re-entrance region \( 39 \)] wherein the minimal free energy is achieved by re-distribution of the values of \( m_z(T) \) and \( m_x(T) \). In particular, within this region, upon say, decreasing \( T \), the system goes smoothly from the disordered phase \( m_z(T) = 0 \) to the ordered one \( m_z(T) \neq 0 \), and back to \( m_z(T) = 0 \). Note also from Eq. (33), that \( \lambda \) creates a self-consistent renormalization of the external field \( \Omega \), in terms of the dimensionful Hamiltonian (1), resulting in, particularly, the temperature dependence of \( m_z \) in the ordered phase. (Compare to the pure IMTF where at \( T < T_c \) it is frozen at \( m_z = 1/g \)). According to Eq. (32) the effective external field \( 1 + 2\lambda_m m_z/\pi \) grows at low temperature, bringing \( m_z \) closer to its maximal value \( 1/2 \) and in the same time, forcing \( m_z \) to decrease [cf. Eq. (30)]. This is another argument in the way to qualitatively explain why the system can re-enter the disordered phase upon decreasing the temperature. However, we should point out that these results and arguments are based on the mean-field analysis, so whether or not the re-entrance survives a more sophisticated treatment of the model, is not clear at this time.

**B. Case \( \varepsilon \neq 0 \): double re-entrance**

An important qualitative change in the critical behavior of the model occurs upon switching on the dimerization coupling \( \varepsilon \). At any \( \varepsilon \neq 0 \) the critical temperature is never zero, even if \( g \ll g_{\lambda} \). Let us note first that when \( \{ \Delta, T \} \rightarrow 0 \) then \( \eta_n \sim -\ln(\min\{ \Delta, T \}) \) is divergent, so this limit should be treated with care. When \( g \gg g_{\lambda} \) [cf. also conditions \( 31 \)] the behavior of the critical temperature and magnetization \( m_z \) is the same as in the case \( \varepsilon = 0 \). Upon decreasing \( g \), the critical temperature \( T_c \) decreases, and according to Eq. (35) \( m_z \approx \frac{1}{2} \), while from Eqs. (32) \( m_z^{-1} \approx c_1 - c_2 \ln T_c \), where \( c_1, c_2 \) are some constants. From the consistency of those equations we conclude that for any finite \( \varepsilon \) the critical temperature never vanishes. To put it differently, coupling \( \varepsilon \) destroys the quantum critical point of the IMTF. This constitutes a very important feedback effect of the spin chains on the charge degrees of freedom.

Characteristic numerical results for \( T_c(g) \) are shown in Fig. 2. Similar to the case \( \varepsilon = 0 \), the critical temperature shows the re-entrance in the region \( g \lesssim g_{\lambda} \). Moreover, instead of going to zero at \( g = g_{\lambda} \) as for \( \varepsilon = 0 \), the curve \( T_c(g) \) turns left, and \( T_c \) remains finite even at \( g = 0 \), albeit exponentially small in \( \varepsilon \). For the two curves discussed above in the case \( \varepsilon = 0 \), we show in Fig. 2 their counterparts at \( \varepsilon = 0.1 \). As one can see from the whole curve \( T_c(g) \), at bigger values of \( \lambda \) the system manifests a well-pronounced double re-entrance.

It follows from Eqs. (32, 33, 34, 35) that the left turn of the critical temperature and its failure to vanish at \( g = g_{\lambda} \) can be analytically described as a BCS-type solution for \( T_c(g) \), generated by finite \( \varepsilon \). Approximate solutions for \( T_c(g) \) found in two regions of \( g \) are

\[
T_c \approx \begin{cases} 
\frac{g}{4}, & g \gg g_{\lambda} \\
\frac{\lambda j}{2} \exp\left[-\frac{\pi j}{4\varepsilon}(g_{\lambda} - g)\right], & \text{BCS regime}
\end{cases} \tag{40}
\]

where

\[
\bar{J} = J + \frac{\lambda}{4} \tag{41}
\]

The boundary where the low-temperature BCS regime sets in and the related formulas are applicable, is given by the condition

\[
\text{BCS regime} : \quad g < g_{\lambda} + \frac{4\varepsilon^2}{\pi J} \tag{42}
\]
The field-induced magnetization $m_z$ in these regions is

$$m_z \approx \begin{cases} \frac{1}{2}, & g \gg g_\lambda \\ \frac{1}{2}, & \text{BCS regime} \end{cases}$$

(43)

C. Case $\varepsilon \neq 0, \lambda = 0$: no re-entrance

The analytical treatment in the intermediate regime when $g \sim g_\lambda$ is involved due to re-entrance. The situation simplifies when $\lambda = 0$ and the re-entrance is absent (see the curve for $J = 1, \lambda = 0, \varepsilon = 0.1$ in Fig. 2). Then both the intermediate $g \gtrsim g^* (g^* = 2)$ and the BCS regimes are well described by the approximate equation

$$g - g^* + \frac{4\varepsilon^2}{\pi J} \ln \frac{\beta_J}{2T_c} = 4e^{-1/T_c}$$

(44)

In particular, at the IMTF quantum critical point one finds that it is destroyed by the inter-ladder dimerization coupling $\varepsilon$, resulting in

$$T_c \approx \ln^{-1} \left( \frac{\pi J}{\varepsilon^2} \right), \quad g = g^*$$

(45)

So when the re-entrance is absent, upon decreasing $g$ the critical temperature $T_c$ monotonically decreases from the IMTF linear regime $T_c \propto g$ via the logarithmic dependence (45) towards the exponential BCS regime (40). It is interesting to note that a similar inverse-log dependence of the transition temperature near quantum criticality has been found in a recent molecular-field study of Cu$_2$Te$_2$O$_5$Br$_2$, a spin-system containing coupled spin-tetrahedra.

V. PROPERTIES OF THE AFE PHASE

A. AFE order parameter

In the ordered phase we can derive from Eqs. (26, 27, 30) the following equation

$$\frac{1}{2} \sqrt{m_x^2 + m_z^2} \tanh \left( \frac{\beta g_x}{2} \sqrt{m_x^2 + m_z^2} \right) = 1$$

(46)

where

$$g_x \equiv g + \frac{4\varepsilon^2}{\pi (J + \lambda m_z^2)} \eta_n$$

(47)

To determine the temperature behavior of the AFE order parameter $m_x$ in the immediate vicinity of the critical temperature where

$$\tau \equiv \frac{T_c - T}{T_c} \ll 1,$$

(48)

we need to expand Eq. (46) near $T_c$, taking into account that all parameters $m_x, m_z, g_x$ entering this equation are temperature-dependent and related via the mean-field equations.

It can be shown that to leading order

$$t_n(\Delta, \tilde{\beta}) \approx t_n(0, \tilde{\beta}) + \Delta^2 t_{n, \Delta^2}$$

$$\eta_n(\Delta, \tilde{\beta}) \approx \eta_n(0, \tilde{\beta}) + \Delta^2 \eta_{n, \Delta^2}$$

(49)

(50)

with the partial derivatives given by the following equations

$$i_{n, \Delta^2} = \frac{\partial t_n(\Delta, \tilde{\beta})}{\partial \Delta^2} \bigg|_{\Delta = 0} = - \left[ a(\tilde{\beta}) - t_n(0, \tilde{\beta}) \right]$$

(51)

$$\eta_{n, \Delta^2} = \frac{\partial \eta_n(\Delta, \tilde{\beta})}{\partial \Delta^2} \bigg|_{\Delta = 0} = - \left[ \frac{1}{2} a(\tilde{\beta}) + \frac{1}{4} b(\tilde{\beta}) - \eta_n(0, \tilde{\beta}) \right]$$

(52)
and

\[
a(\beta) \equiv \frac{1}{2} \int_{0}^{1} \frac{\beta dz}{\cosh^2 \beta z} \ln \frac{1 + \sqrt{1 - z^2}}{z} \\
b(\beta) \equiv \int_{0}^{1} \frac{\beta dz}{\cosh^2 \beta z} \left[ \frac{1}{\sqrt{1 - z^2}} + 2\beta \sqrt{1 - z^2} \tanh \beta z \right]
\]

Using the above equations in the leading-order expansion of Eqs. (32, 46) near \( T_c \), we can show that

\[
m_x^2 \propto \tau
\]

along the whole line of the critical temperature depicted in Fig. 2, i.e., \( T_c \) defines a second-order phase transition, and the order parameter has the mean-field critical index \( 1/2 \). In general, the coefficient of proportionality in the above relationship has to be defined numerically. For the regimes of strong Ising coupling and BCS, the order parameter can be calculated analytically.

For the functions defined by Eqs. (53) the following expansions are obtained:

\[
a(x) \approx \begin{cases} \\
\frac{\pi}{4} x(1 - \frac{1}{6} x^2) + O(x^5), & x < 1 \\
\frac{1}{2} \ln \frac{\pi}{4} x + O(\frac{1}{x}), & x > 1
\end{cases}
\]

and

\[
b(x) \approx \begin{cases} \\
\frac{\pi}{4} x(1 + \frac{1}{6} x^2) + O(x^5), & x < 1 \\
\frac{1}{2} + \frac{14}{3} \zeta(3) x^2 + O(\frac{1}{x^2}), & x > 1
\end{cases}
\]

where \( A \) is defined below Eq. (35), \( e \) is the exponential constant and \( \zeta(x) \) - Riemann’s zeta-function. Combining these asymptotics with those given by Eqs. (34, 35), we find

\[
m_x^2 \approx \begin{cases} \\
\frac{\pi^2}{9} x^2 \tau, & g \gg g_x \\
\frac{x^2}{2} \ln \frac{\pi}{4} x + \frac{1}{x} \zeta(3) x^2 \tau, & \text{BCS regime}
\end{cases}
\]

Notice that \( m_x \) is continuous across \( T_c \), however as follows from Eqs. (32, 49, 50) in the AFE phase \( m_x(T, m_x) = m_x(T, 0) + O(m_x^2) \), so its derivative with respect to temperature has a finite discontinuity at \( T_c \).

**B. \( T = 0 \)**

At zero temperature the parameters \( t_n \) and \( \eta_n \) are given in terms of the complete elliptic integral of the first and second kind, such that

\[
t_n = \frac{E(1 - \Delta^2) - \Delta^2 K(1 - \Delta^2)}{1 - \Delta^2} \\
\eta_n = \frac{K(1 - \Delta^2) - E(1 - \Delta^2)}{1 - \Delta^2}
\]

According to Eq. (30) at \( T = 0 \): \( m_x^2 + m_z^2 = \frac{1}{4} \), so we can establish the range within which \( \Delta \) can lie [cf. Eq. (26)]

\[
0 \leq m_x \leq \frac{1}{2} \iff 0 \leq \Delta \leq \frac{\varepsilon}{J} \ll 1
\]

where the last strong inequality is just the condition for Hamiltonian’s couplings we are implying from physical grounds. Note that \( \Delta_{\text{max}} \equiv \varepsilon/J \) is the absolute maximum \( \Delta \) can reach. Then we can safely make expansions up to terms \( O(\Delta^2 \ln \Delta) \)

\[
t_n \approx 1 \\
\eta_n \approx \ln \frac{4}{e \Delta}
\]
which can be used in Eq. (62) for any $\epsilon$. At large Ising couplings $g \gg g_\lambda$, $m_z$ is small, while $m_x$ is close to its maximal value ($\Delta \approx 2\epsilon m_x/J$):

$$m_x^{-1} \approx g - \frac{2\lambda}{\pi} + \frac{4\epsilon^2}{\pi J} \ln \frac{4J}{\epsilon \epsilon}$$  \hspace{1cm} (63)

$$m_x \approx \frac{1}{2} - m_z^2$$  \hspace{1cm} (64)

For smaller couplings $g \leq g_\lambda$ the order parameter $m_x$ is small ($\Delta \approx \frac{2\epsilon m_x}{J}$):

$$m_x \approx \begin{cases} \frac{\epsilon}{\sqrt{2\pi J}} \ln \left( \frac{8\pi^3 \pi}{\epsilon \epsilon} \right), & g \gg g_\lambda \\ \frac{2J}{\epsilon \epsilon} \exp \left[ - \frac{\pi J}{4\epsilon}(g_\lambda - g) \right], & \text{BCS regime} \end{cases}$$  \hspace{1cm} (65)

while

$$m_z \approx \frac{1}{2} - m_z^2$$  \hspace{1cm} (66)

is close to its maximal value. The dimerization parameter $\delta$ to leading order reads

$$\delta \approx \begin{cases} \left( \frac{1}{2} - \frac{1}{g_\lambda^2} \right) \ln \frac{2J}{\epsilon \epsilon}, & g \gg g_\lambda \\ \frac{\epsilon}{\sqrt{2\pi J}} \ln \left( \frac{8\pi^3 \pi}{\epsilon \epsilon} \right), & g = g_\lambda \\ \frac{\pi J}{2\epsilon \epsilon}(g_\lambda - g) \exp \left[ - \frac{\pi J}{4\epsilon}(g_\lambda - g) \right], & \text{BCS regime} \end{cases}$$  \hspace{1cm} (67)

C. Spin susceptibility

The zero-field spin susceptibility per rung (i.e., per spin) is given by

$$\chi_s = -\frac{\partial \langle s_x \rangle}{\partial h_M} \bigg|_{h_M=0} = -\frac{\partial^2 f_s}{\partial h_M^2} \bigg|_{h_M=0} = \frac{\beta}{2\pi} \int_0^{\frac{\pi}{2}} \frac{d\phi}{\cosh^2 \frac{\beta}{2} \xi(\phi)}$$

(68)

Its temperature dependence is similar to that of the dimerized Heisenberg or $XY$ spin chains. In the ordered AFE phase it shows the spin-gap behavior. At low temperatures the asymptotics of the integral (68) is given by the following expression

$$\chi_s = \frac{1}{J + \lambda m_z^2} \left( \frac{2\Delta_{SG}}{\pi T} \right)^\frac{\beta}{2} \exp \left[ - \frac{\Delta_{SG}}{T} \right] \left\{ 1 + \frac{3}{8} \frac{T}{\Delta_{SG}} + O \left( \frac{T^2}{\Delta_{SG}} \right) \right\}$$

(69)

The characteristic energy scale parameter $\Delta_{SG}$ defined as

$$\Delta_{SG} = 2\epsilon m_x$$

is natural to call the spin gap. Such definition coincides with the one stated in terms of a magnon. The latter is equal to the minimal energy needed to flip one spin, and according to Eqs. (18,20), to the one-particle fermion gap. The simple relation (70) between the spin gap and the alternating part of the exchange $b = 2\epsilon m_x$ [cf. Eq. (26)] holds because the $XY$ model is a free-fermion problem. Accounting for fermionic interactions in the $XYZ$ model breaks (70) already at the mean-field level.\footnote{Bosonization treatment of the interacting Jordan-Wigner fermions reveals the power-law relationship $\Delta_{SG} \propto b^{2/3}$ times some log correction.} It is interesting to compare the ratio of the zero-temperature spin gap $\Delta_{SG}^0$ and the critical temperature in different regimes of couplings. At large $g \gg g_\lambda$ this ratio [cf. Eqs. (48,49,40)]

$$\frac{\Delta_{SG}^0}{T_c} \approx \frac{4\epsilon}{g}$$

(71)

is small according to the assumption (41) on the range of the model parameters we are working with. Upon decreasing $g$ the ratio increases. It subtly involves interplay of the model couplings, and it is not easy to get its analytic form. At the critical coupling $g = g^*$ (when $\lambda = 0$) it can be found from Eqs. (47,65), and roughly

$$\frac{\Delta_{SG}^0}{T_c} \approx \left( \frac{8}{\pi J} \right)^\frac{1}{2} \epsilon^2 |\ln \epsilon|^{\frac{1}{2}}$$

(72)
More accurate evaluations of Eqs.(45,65), as well as direct numerical calculations show that within the parameter range (11) the ratio $\Delta_{SG}/T_c$ does not exceed 1. In the BCS region ($g < g\lambda$) the ratio is maximal, and moreover, it is the universal constant [cf. Eqs.(40,65)]

$$\frac{\Delta_{SG}}{T_c} \approx \frac{\pi}{e^\gamma} \approx 1.76, \quad \text{BCS regime}$$

(73)
of any BCS-type theory. Also, as we can see from Eq.(57), the temperature dependence of the spin gap in this regime is exactly as that of the superconductivity gap provided by the BCS theory:

$$\Delta_{SG} \approx 3.06 T_c \tau^{1/2}, \quad \tau << 1, \quad \text{BCS regime}.$$ (74)

D. Charge (pseudospin) susceptibilities

We will be interested in the pseudospin (i.e., charge) susceptibilities with respect to two external (electric) fields which have the same spatial dependence as the Weiss fields $h_{mn}^i$. Namely, the field along $z$ pseudospin “direction” is constant $E_{mn}^z = -E_z$, while the field $E_{mn}^x = \left(-1\right)^{m+n+1}E_x$ is staggered. We define the charge susceptibilities as

$$\chi^{c}_{ij} \equiv \left. \frac{\partial m_i}{\partial E_j} \right|_{E_j=0} : (i,j) = x \text{ or } z$$

(75)

These quantities can be calculated from equations of Section III with the Weiss fields shifted as $h_i \rightarrow h_i + E_i$.

1. $T > T_c$

After a straightforward but somehow lengthy algebra we obtain for the disordered paraelectric (PE) phase $T > T_c$:

$$\chi^{c}_{zz} = \chi^{c}_{zz} = 0$$

(76a)

$$\chi^{c}_{zz} = \frac{\beta}{4} \left[ \frac{1}{1 - \frac{4\pi}{\lambda} \left( 1 - 4m_z^2 \right) \left( n + \lambda \beta t_{n,\beta} \right) + \frac{2m}{\pi(1 + \lambda m) \left( m - 2\lambda \pi \tilde{t}_n \right)} \eta_n \right]$$

(76b)

$$\chi^{c}_{xx} = \frac{m_z}{1 - m_z \left( g + \frac{\lambda}{\pi \left( \tilde{J} + \lambda m_z \right)} \eta_n - \frac{2\lambda}{\pi} \tilde{t}_n \right)}$$

(76c)

where

$$\tilde{t}_{n,\beta} = \frac{\partial \tilde{t}_n(\Delta, \tilde{\beta})}{\partial \tilde{\beta}}.$$ (77)

Note that even in the limit $E_z \rightarrow 0$, $\chi^{c}_{zz}$ is in fact the susceptibility under the applied field $\Omega$ [cf. Eq.(1)] along this direction. It does not show critical behavior. On the contrary, from the comparison of Eqs.(32,76c) we immediately see that $\chi^{c}_{xx}$ diverges when $T \rightarrow T_c^+$. At large $g \gg g\lambda$ from Eqs. (10-13) we find

$$\chi^{c}_{xx} \approx \frac{1}{4T}$$

(78a)

$$\chi^{c}_{xx} \approx \frac{4T_c}{g^2} \frac{1}{\tau}$$

(78b)

so $\chi^{c}_{xx}$ diverges with the mean-field value of the critical index. The same divergence can be obtained from the analytical treatment in the BCS regime $g < g\lambda$ for $T \gtrsim T_c \ll 1$ where

$$\chi^{c}_{xx} \approx \frac{1}{T} \exp \left[ -\frac{\Delta_{CG}}{T} \right]$$

(79a)

$$\chi^{c}_{xx} \approx \frac{\pi \tilde{J}}{4\Delta^2 \ln \frac{T_c}{T}} \approx \frac{\pi \tilde{J}}{4\Delta^2 \left| \tau \right|}$$

(79b)

At these low temperatures one probes the $\Omega$-generated charge gap $\Delta_{CG} = 1 + \lambda/\pi \equiv g\lambda/2$ in $\chi^{c}_{xx}$, renormalized (comparatively to its bare value 1) via the coupling $\lambda$ by the dimer operator average $\tilde{t}$ [cf. Eq.(24)].
2. \( T < T_c \)

We will not give here the closed expressions for the four susceptibilities below \( T_c \), since there are too cumbersome. The important properties of these quantities are the following: \( \chi^c_{xz} \) is continuous across the transition, and \( \chi^c_{xx} \propto 1/\tau \) is divergent when approaching \( T_c \) from below. In the regimes of strong Ising coupling and BCS, \( \chi^c_{xx} \) can be calculated analytically, showing that

\[
\chi^c_{xx}(T \to T^-) = \frac{1}{2} \chi^c_{xx}(T \to T^+)
\]

as one should have expected in the mean-field theory. The transverse pseudospin susceptibilities are “diamagnetic” (indicating that the system tends to preserve the pseudospin vector’s length) and show weaker divergence:

\[
\chi^c_{xz} = \chi^c_{zx} = \begin{cases} 
- \frac{1}{g} \left[ \frac{2}{g^2 - 1} \right]^{1/2}, & g \gg g_{\lambda} \\
- \frac{\pi^2}{2 \sqrt{14} \zeta(3)} \tilde{J} T_c \tau^{-1/2}, & \text{BCS regime}
\end{cases}
\]

E. Specific heat capacity

The easiest way to obtain the specific heat capacity in our approach, is to calculate it directly from the mean-field energy (25)

\[
c = \frac{\partial \mathcal{E}_{\text{MF}}}{\partial T}
\]

(82)

(here, in fact \( c = c_v \)). As usual, at the second-order phase transition temperature the specific heat undergoes a finite jump. For two regimes of couplings \( c \) can be easily calculated analytically. At high temperatures \( T \gg 1 \) above \( T_c \)

\[
c \approx \frac{1}{4T^2} \left( 1 + \frac{J^2}{2} \right)
\]

(83)

As one can see from the above equation, spins and pseudospins \( (m_z) \) contribute to the specific heat in the same fashion. At large \( g \gg g_{\lambda}, T_c \) is also large. For this regime we find

\[
c^+ - c^- \approx \frac{g^2 - 4}{4gT_c}, \ g \gg g_{\lambda}
\]

(84)

where \( c^\pm \equiv c(T \to T_c^\pm) \). Low temperatures \( (T \ll 1) \) within the disordered PE phase are attainable only at smaller couplings \( g \lesssim g_{\lambda} \). In the BCS regime \( (T > T_c) \) we get

\[
c \approx \frac{\pi T}{3J} + \mathcal{O}(T^3), \ \text{BCS regime}
\]

(85)

Since pseudospins \( m_z \) are gapped, at these low temperatures their contribution to the specific heat is exponentially suppressed as \( \mathcal{O}(\exp[-\Delta_{\text{CG}}/T]) \), thus only spins contribute. The latter contribution is equivalent to the one of a degenerate free electron gas. Eqs. (30) for the AFE phase in the BCS regime gives

\[
\frac{\partial m_z}{\partial T} = \frac{\partial m_z^2}{\partial T^2} + \mathcal{O}(e^{-1/T})
\]

(86)

Using it along with asymptotics (34, 35, 40, 55, 56, 57), we obtain

\[
c^+ - c^- \approx \frac{4\pi}{7 \zeta(3) J} T_c, \ \text{BCS regime}
\]

(87)

Then the ratio

\[
\frac{c^-}{c^+} = 1 + \frac{12}{7 \zeta(3)} \approx 2.43
\]

(88)
is another universal constant known from the BCS theory. Note that the origin of this universality can be traced from Eq. (85), which results in that only gapped fermions contribute to the heat capacity in the vicinity of $T_c$ in the BCS regime.

Since the specific heat jump is positive at any regime of couplings, it indicates the free energy decrease in the ordered AFE phase $f_{AFE} - f_{FE} \propto -m_x^2$.

Upon approaching zero temperature the specific heat capacity decreases exponentially for any coupling $g$. The decay scale is defined by $\Delta_{SG}$ which is the smallest gap in the model (the zero temperature charge gap varies from $\Delta_{CG} = g_\lambda/2$ in the BCS regime to $\Delta_{CG} = g/2$ in the regime of large $g$), so $c \propto \exp[-\Delta_{SG}/T]$ similar to the spin susceptibility.

## VI. SUMMARY AND DISCUSSION

We study the spin-pseudospin model comprised from the Ising Model in Transverse Field (IMTF) Hamiltonian for pseudospins coupled to the spin-$\frac{1}{2}$ $XY$-Hamiltonian, on a triangular lattice. The effective Hamiltonian, similar to ours, appears from analyses of the quarter-filled ladder compound NaV$_2$O$_5$, which can be described by a set of spin and pseudospin variables, the latter representing the charge degrees of freedom. Following the proposal of Mostovoy and Khomskii for the scenario of the phase transition in NaV$_2$O$_5$, we include into the Hamiltonian a specific inter-ladder spin-pseudospin coupling term allowed by symmetries, linear over the pseudospin operators.

In the framework of the molecular-field approximation we explore the phase diagram of the model and find that it possesses two phases: the disordered phase without charge order or spin gap; and the low-temperature phase containing both the anti-ferroelectric (zigzag) charge order and spin gap (without long-range spin order). Such ordered phase is experimentally observed below the transition temperature $T_c = 34$ K in NaV$_2$O$_5$. The phase transition in our model is of the second kind. We calculate various physical quantities, as the order parameter, spin and pseudospin (charge) susceptibilities, specific heat, and find their (mean-field) temperature behavior near $T_c$.

Our analysis reveals an important property of the phase diagram, not envisaged in the original Mostovoy-Khomskii proposal: the inter-ladder spin-pseudospin coupling ($\varepsilon$) not only creates the spin dimerization (spin gap) triggered by the charge ordering in the region of Ising’s coupling where the IMTF can order ($g > g_\lambda$), but it also generates simultaneous appearance of the charge order and spin gap in the case when the IMTF is always disordered ($g < g_\lambda$). So, the coupling $\varepsilon$ destroys the IMTF quantum critical point at $g = g_\lambda$ and results in a continuous evolution of $T_c(g) \neq 0$ into the region $g < g_\lambda$. In that region $T_c$ has the exponential BCS-like dependence on model’s couplings.

An interesting feature of model’s phase diagram is that near the IMTF critical coupling $g_\lambda$ it shows regimes with a re-entrance (for couplings $\lambda \neq 0$, $\varepsilon = 0$) or with a double re-entrance (for couplings $\lambda \neq 0$, $\varepsilon \neq 0$). How pronounced the re-entrant behavior is, depends on the interplay of couplings, but the re-entrance is absent when $\lambda = 0$. Usually, re-entrance is due either to disorder effects or due to the stabilization of the ordered phase via the release of entropy by secondary degrees of freedom. The IMTF does not show any re-entrance at the mean-field level, but accounting for the first-order fluctuation corrections revealed some re-entrance in the 2D IMTF. In that study it was assumed to be an artifact of poor approximation. In our case, we have given some qualitative arguments on how the re-entrant behavior can be understood from the competition of model’s energy scales. However, we cannot exclude that the found re-entrance is a mean-field artifact. A more detailed study on this re-entrance will be presented elsewhere.

Now we proceed with the discussion on the applications of our results to NaV$_2$O$_5$. For NaV$_2$O$_5$ we take the following data: $\Omega = 700$ meV, $T_c = 34$ K, $\Delta_{SG} = 106$ K ($\Delta_{SG}/T_c \approx 3$), $J_H + \frac{1}{2}J_{ST} = 50$ meV. The microscopic calculations give $\tilde{\varepsilon} = 15$ meV. Using these data as model’s dimensionless parameters [cf. notations (3)], $\varepsilon = 0.021$, $J = 0.065$, $\lambda = 0.026$ we find that $T_c = 0.004$ lies on the curve $T_c(g)$ in the BCS-regime region with $g = 1.99$ ($J_H = 1.4$ eV), close to the critical coupling $g_\lambda = 2.02$. The estimates for $J_H$ in NaV$_2$O$_5$ which is proportional to the in-ladder Coulomb repulsion between neighboring rungs, vary, but do not exceed $J_H = 1.5$ eV. The whole curve $T_c(g)$ with the above parameters $J, \lambda, \varepsilon$ looks very much similar to that shown in Fig. 2 for $J = 1, \lambda = 0, \varepsilon = 0.1$, without appreciable re-entrance. (In any case $g_c = 1.99 < g_{min}$.) We would not insist that our mean-field analysis gives the quantitatively correct evaluation of $T_c$ in terms of the microscopic parameters for NaV$_2$O$_5$, since the uncertainties in their values left us with a freedom to make this reasonable fit. However, we think we have grasped the qualitatively correct physics. The question why $T_c$ is so low in NaV$_2$O$_5$, or what is the scale that gives such a low $T_c$, can be answered as follows: $g$ for NaV$_2$O$_5$ lies in the proximity of the quantum critical point of the IMTF (most likely on the “disordered side, i.e., $g < g_\lambda$). $T_c$ is determined by the two scales: $g - g_\lambda$ and the inter-ladder dimerization coupling $\varepsilon$. The latter destroys the IMTF quantum critical point and makes the ordering possible at $g < g_\lambda$.

A flaw of the present study is that we cannot account for the large BCS ratio $\Delta_{SG}/T_c$ observed in NaV$_2$O$_5$. The small value of $T_c$ indicates that we are either in the BCS regime described by the analytical result, or very close to it, so the ratio cannot exceed the universal BCS value. This gives the spin gap almost two times smaller than
the experimental value. This clearly indicates that for more realistic calculations the spin part of the Hamiltonian should be modified, and instead of the $XY$ spin model the full three component Heisenberg Hamiltonian should be considered. As we have already explained it in Section we will not speculate more on this point.

Experiments of various kinds unequivocally demonstrate that what occurs in NaV$_2$O$_5$ at $T_c = 34$ K is a (thermal) continuous structural phase transition. Also rather large regions of structural (charge-ordering) two-dimensional fluctuations are seen on the both sides of $T_c$. The order parameter critical index $\beta = 0.17 - 0.19$ is close to what one expects for a two-dimensional phase transition with a one-component order parameter. So we conclude that the universality class of this transition is the 2D Ising. In order to obtain a complete description of the transition in NaV$_2$O$_5$, thermal fluctuations in the IMTF-sector of the spin-pseudospin Hamiltonian must be taken into account. According to what we said above, we expect the Ising coupling for the NaV$_2$O$_5$-Hamiltonian to lie near the quantum critical point of the 2D IMTF. From the experimental results we expect that physics of NaV$_2$O$_5$ near $T_c$ is controlled by the region of the 2D IMTF phase diagram where 2D Ising (thermal) fluctuations dominate. Although at lower temperatures the change of the nature of these fluctuations into 3D Ising (quantum) may also become important. We believe that apart from the applications to NaV$_2$O$_5$, a study of the fluctuations in proximity of this quantum critical point is a very interesting problem of its own.

It is worth pointing out that the pseudospin sector of our Hamiltonian is only quasi two-dimensional, in the sense that pseudospins on neighboring ladders are coupled only via the dimerization coupling $\varepsilon$. Accounting for the Ising coupling between ladders, as one can see from the triangular lattice mapping shown in Fig.1 will cause a frustration. The neglect of this coupling for the applications to NaV$_2$O$_5$ is justified from microscopic grounds, but from a general point of view it would be interesting to study a Hamiltonian, similar to ours, with a truly two-dimensional IMTF part.

Acknowledgments

We are grateful to E. Orignac and R. Valentí for helpful discussions. We are also thankful to F. Capraro and K. Pozgajec for their help with the software and numerical calculation. This work is supported by the German Science Foundation.

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Contrary to NaV$_2$O$_5$, the quasi-2D compound Na$_2$Ti$_2$Pn$_2$O is less studied experimentally and less understood, so in the following we will restrict our discussion to the former.

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In a recent study of a similar one-dimensional spin-pseudospin model by Y. Ohta, T. Nakaegawa, and S. Ejima, cond-mat/0309433 the estimates were such that $J/I = 3.2$ eV. At this coupling the 1D IMTF has a phase transition at $T = 0$, and this is how the charge ordering occurs in their scenario. We however find their value of $J/I$ being too overestimated.

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FIG. 1: Two-dimensional triangular lattice of coupled ladders. A vertical line denotes a single ladder where a dot denotes its rung. At each rung $(m, n)$ there is pseudospin $T_{mn}$ and spin $S_{mn}$ (not shown). In the region encircled by the dashed line we show four sites involved in each term of the sum in the Hamiltonian (2). Spins and pseudospins residing on two sites of the same $m$-th ladder are coupled via the exchange interaction terms, while two pseudospins from $(m-1)$-th and $(m+1)$-th ladders are coupled via the dimerization interaction constant $\varepsilon$. The pseudospin $T_{mn}$ ordering pattern shown in the figure corresponds to the ordered anti-ferroelectric (zigzag) phase.

FIG. 2: Critical temperature of the AFE phase transition as a function of the Ising coupling $g$ at different values of $\lambda, \varepsilon$. The critical couplings $g^* = 2$ and $g_\lambda = 2.6366$. Large empty circles on the curves at $\varepsilon = 0.1$ indicate the right boundary for the BCS-region described by the exponential dependence (40). At large values of $g$ (not shown) all curves $T_c(g)$ approach the asymptotic line $T_c = g/4$. 