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Rapid Note

A new model to describe the physics of $(VO)_2P_2O_7$

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Abstract. In the past different models for the magnetic salt vanadyl pyrophosphate $(VO)_2P_2O_7$ were discussed. Neither a spin ladder nor an alternating chain are capable to describe recently measured magnetic excitations. In this paper we propose a 2D model that fits better to experimental observations.

PACS. 75.10.-b General theory and models of magnetic ordering – 75.25.+z Spin arrangements in magnetically ordered materials (including neutron and spin-polarized electron studies, synchroton-source X-ray scattering, etc.) – 75.40.Mg Numerical simulation studies

Low dimensional quantum spin systems have been a field of intense theoretical and experimental research over the last decades. Special interest was given to spin ladder and chain materials. One compound that has been examined in this context is the insulating magnetic salt vanadyl pyrophosphate $(VO)_2P_2O_7$. Initially it was considered as a prototypical realization of a two-leg antiferromagnetic Heisenberg ladder [1]. However, susceptibility data on polycrystalline and single crystalline material could be well fitted with both, ladder or alternating chain models [1–3], stressing the fact that susceptibility is not too sensitive to the particular model. Early inelastic neutron scattering measurements on polycrystalline samples indicated a spin gap of about 3.7 meV and supported a two-leg ladder model with the coupling constants estimated from susceptiblity data [4].

Recent neutron scattering experiments with powder samples [6] and with an array of single crystals [7] provided detailed information on the low-energy excitation spectrum. Garrett *et al.* [7] observed a triplet branch with strongest (antiferromagnetic) dispersion in b-direction, weak (ferromagnetic) dispersion in a-direction, and a spin gap of 3.1 meV. Most notably they found an additional *second* branch, separated from the first by an energy smaller than the gap. This was inconsistent with the picture, of $(VO)_2P_2O_7$ being a spin ladder in a-direction, but also an alternating Heisenberg chain in b-direction can not explain a second triplet branch over the whole Brillouin zone, as was shown recently [8,9].

In this work, starting with the alternating Heisenberg chain, we check whether coupling of (two) chains resolves this puzzling situation. As we do not succeed proceeding this way, we consider a new, truly two-dimensional model. We perform exact diagonalizations of finite systems with up to 32 spins and periodic boundary conditions, supplemented by finite-size analysis if possible.

The Hamiltonian of the alternating Heisenberg chain (AHC) reads as follows

$$H_{\mathrm{AHC}} = J_b \sum_i (1 + \delta(-1)^i) \mathbf{S}_i \cdot \mathbf{S}_{i+1}, \qquad (1)$$

where $\mathbf{S}_i$ are spin-1/2 operators and i denotes the sites in b-direction (see Fig. 1). For $\delta > 0$ the spectrum has a gap; there is an one-magnon branch and a singlet branch, at least around momentum $\pi/2$, below a continuum of states.

As an example Figure 2 shows the low-lying excitations of a finite system of 32 sites for $\delta = 0.2$. The magnon branch is fitted to a sum of cosines $\omega_q^m = \sum_{n=0}^{5} a_n \cos(2nq)$ and the shaded region corresponds to the continuum of two-magnon excitations resulting from this dispersion. Recently it was stressed [8,10] that there exists a second well-defined triplet below the two-magnon continuum near momentum $\pi/2$, but as Figure 2 indicates, the second triplet occurs only very close to higher states, even for the relatively strong dimerization of $\delta = 0.2$. Therefore it was stated that an alternating chain will not explain the second triplet excitation observed in $(VO)_2P_2O_7$ at *all* q-momenta. However, it is known [8] that including frustration, *i.e.* an antiferromagnetic next nearest-neighbor interaction α between $\mathbf{S}_i$ and $\mathbf{S}_{i+2}$, into the alternating chain model, yields a second well-defined triplet branch

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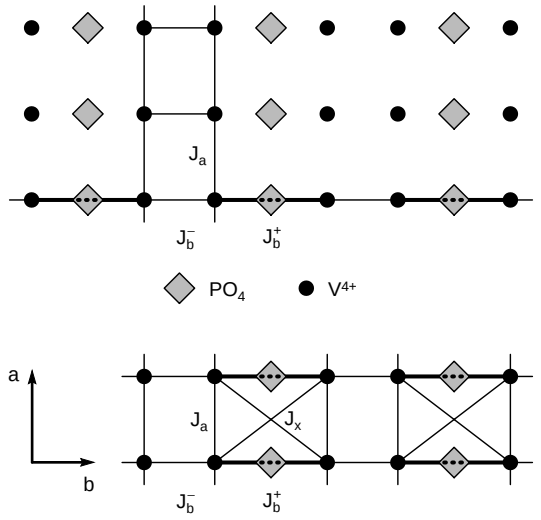

Fig. 1. Schematic structure of $(VO)_2P_2O_7$. The exchange couplings are depicted for (i) the ladder model ($J_\parallel = J_a, J_\perp = J_b^-$), (ii) the alternating chain model ($J_b^\pm = J_b(1 \pm \delta)$, and (iii) the new model ($J_b^\pm, J_a, J_\times$). Throughout we measure energies in units of J_b.

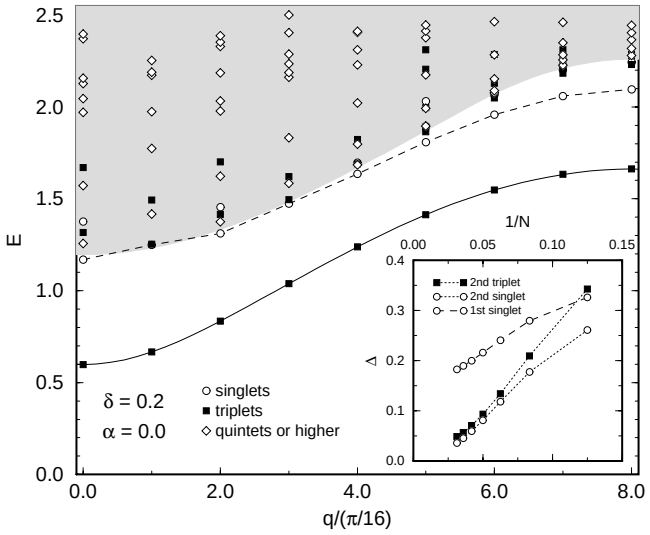

Fig. 2. Low-energy excitations of the AHC. The inset shows the difference Δ of the 1st and 2nd singlet and the 2nd triplet to the 1st quintet at momentum $\pi/2$ *versus* inverse chain length.

below the continuum in the whole Brillouin zone, provided α is sufficiently strong.

Since the nearest-neighbor exchange paths *via* the PO_4 groups in $(VO)_2P_2O_7$ are already relatively complicated [5], additional longer ranged couplings in b-direction, leading to an intra-chain frustration, would presumably involve neighboring chains in a nontrivial way. Therefore, in a first step, we will consider a simple perpendicular coupling of two alternating chains instead:

$$H_{\rm CC} = J_b \sum_{ij=1,2} (1+\delta(-1)^i)\mathbf{S}_{i,j}\cdot\mathbf{S}_{i+1,j} + J_a \sum_i \mathbf{S}_{i,1}\cdot\mathbf{S}_{i,2}. \quad (2)$$

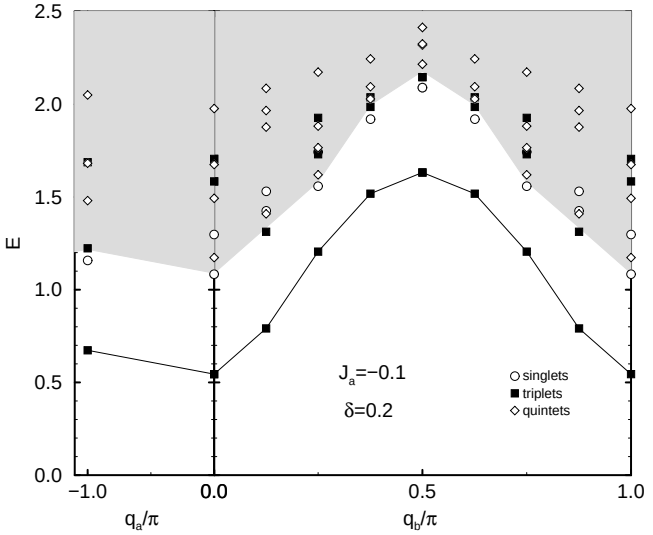

Fig. 3. Low-energy excitations of two alternating chains (16 sites each) coupled ferromagnetically; the parameters are: $\delta = 0.2, J_a = -0.1$.

Here j is numbering the chains. As was already suggested in [7], such a coupling in a-direction should be ferromagnetic to explain the observed dispersion. For illustration, in Figure 3 we plotted a few low-lying energies of a 2×16 system with $\delta = 0.2$ and $J_a = -0.1$. Similar results were obtained for a 3×8 system.

Again we have a well-defined magnon branch. As a guide to the eyes we shaded the region where one would expect a two-magnon continuum, approximated here by adding two magnon energies of the finite system. Close to the continuum edge there are several states: singlets, triplets, as well as quintets. To gain further insight one has to perform a finite-size analysis. For extrapolation to the infinite system we use the following formulas for the lowest singlets and triplets [8,11]:

$$E^{\rm S}(L) = E^{\rm S}(\infty) + \left(\frac{B}{L} + C\right) e^{-L/A} \quad (3)$$

$$E^{\rm T}(L) = E^{\rm T}(\infty) + \frac{B}{L} e^{-L/A}. \quad (4)$$

In Figure 4 a few low-lying excitations at momenta $(q_b, q_a) = (0,0)$, $(0,\pi)$ and $(\pi/2, 0)$ are given subject to the ferromagnetic interchain coupling J_a. At $(0,0)$ and $(0,\pi)$ the data is extrapolated to infinite system size. At $(\pi/2, 0)$ results for a 2×16 system are shown, because the small number of four different system sizes $(2\times 4, 8, 12, 16)$ makes finite size scaling questionable at this momentum. Nevertheless, in the inset we tried to extrapolate the second triplet at $(\pi/2, 0)$ to the infinite system, using the ansatz of equation (4). The plot indicates that this triplet shows a weak nonmonotonic behaviour, in contrast to the single chain case.

Obviously at momenta $(0,0)$ and $(0,\pi)$ there are no second triplets below the two-magnon continuum. Just at $(q_b, q_a) = (\pi/2, 0)$ a second triplet stays very close to the continuum edge, and the well-defined singlet excitation,

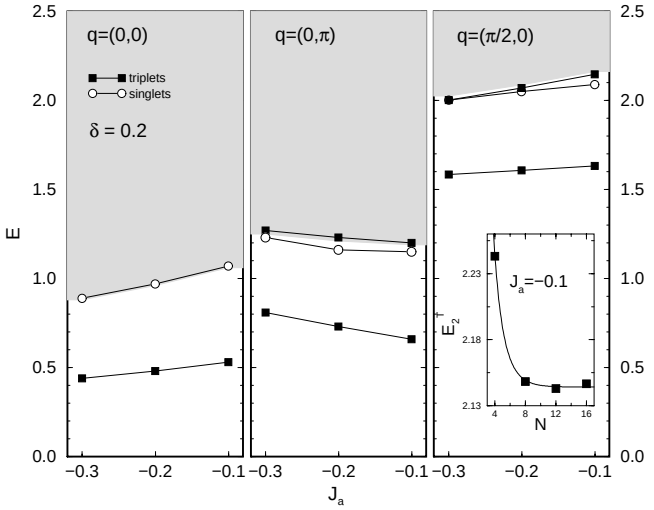

Fig. 4. Energy of the lowest singlet (open circles) and triplet (filled squares) excitations for different interchain coupling J_a and fixed dimerization $\delta = 0.2$.

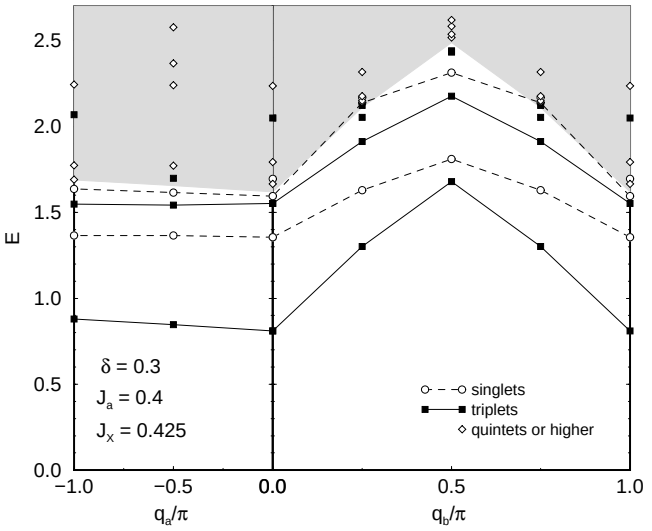

Fig. 5. Low-energy excitations of the 2D model; the system size is 4×8, $\delta = 0.3$, $J_a = 0.4$ and $J_\times = 0.425$.

known from the single alternating chain seems to disappear with increasing interchain coupling J_a.

From the above results we conclude that an interchain coupling of this simple type does not qualitatively change the structure of the low-energy excitations compared to the single alternating chain. Excitations are just shifted (as it seems linearly with J_a in most cases), but no new features appear. This is, why we propose another model for $(VO)_2P_2O_7$.

We mentioned above that frustration in the alternating chain can lead to a well-defined triplet below the two-magnon continuum. Thus going to the second dimension we include an additional, frustrating coupling $J_\times$. Then our model Hamiltonian reads

$$H_\times = J_b \sum_{i,j} (1 + \delta(-1)^i) \mathbf{S}_{i,j} \cdot \mathbf{S}_{i+1,j} + J_a \sum_{i,j} \mathbf{S}_{i,j} \cdot \mathbf{S}_{i,j+1} + J_\times \sum_{i,j} (\mathbf{S}_{2i,j} \cdot \mathbf{S}_{2i+1,j+1} + \mathbf{S}_{2i+1,j} \cdot \mathbf{S}_{2i,j+1}) \quad (5)$$

(*cf.* Fig. 1, lower panel). As yet there is no data available about the strength of such a coupling, but as a first step it seems not unreasonable in view of the oxygen-mediated superexchange paths in $(VO)_2P_2O_7$. We assume all exchange integrals to be antiferromagnetic, but still the parameter space is very large. It appears that $J_\times$ has to be bigger than J_a to get a ferromagnetic magnon dispersion in a-direction, what is plausible. On the other hand, both couplings should not differ too much for a second triplet branch to exist in the whole momentum space, and should have a sufficient strength. The size of the gap to the first triplet branch is (still) mainly controlled by the dimerization δ.

A good choice of parameters is $\delta = 0.3$, $J_a = 0.4$ and $J_\times = 0.425$, for which we diagonalized systems of two, three and four chains with a total number of up to 32 spins. The low-energy excitations of the 4×8 system are shown in Figure 5.

Beside two triplet branches (T_1 and T_2) we observe also a well-defined singlet (S_1), and there might even be a second singlet (S_2) near momentum $(q_b, q_a) = (\pi/2, 0)$. As the difference between $J_\times$ and J_a is small, the dispersion of the triplets is weak in a-direction, in accordance with experiments. We stress that the picture remains qualitatively unchanged going from the 3×8 to the 4×8 system, just the second triplet shifts downwards at momentum $(0, 0)$ with increasing system size. Thus we believe that these features will survive in the infinite system.

To provide some more information on the excitation spectrum, we calculated the dynamical spin structure factor – which is proportional to the neutron scattering cross section – and the integrated spectral weight

$$S(\mathbf{q}, \omega) = \sum_n |\langle n|\mathbf{S}^z(\mathbf{q})|0\rangle|^2 \delta(E_n - E_0 - \omega)\,, \quad (6)$$

$$N(\mathbf{q}, \omega) = \int_0^\omega d\omega' S(\mathbf{q}, \omega')\,, \quad (7)$$

where $\mathbf{S}^z(\mathbf{q}) = \sum_{i,j} e^{i\mathbf{q}\cdot\mathbf{r}_{i,j}} S^z_{i,j}$. In the plot the integral is normalized to one; its real value $N(\mathbf{q}) = N(\mathbf{q}, \infty)$ is noted in each panel.

The most pronounced feature of the structure factor is of course the first triplet, but it seems that we need a finite momentum component in b-direction to get some weight. For comparison take the dashed and solid lines in the upper two panels of Figure 6, corresponding to momenta (π, x) and $(0, x)$, respectively, that are equivalent in energy. The second triplet occurs only in a-direction and has very low spectral weight (note the small integrated intensity in the upper panel, where T_2 seems to be dominant). This is unsatisfactory in view of the experiments, where both triplets are comparable in intensity. Obviously, it is an effect of special selection rules, since we arranged the spins in a quadratic lattice for our calculation, not taking into account their real positions in the $(VO)_2P_2O_7$ crystal.

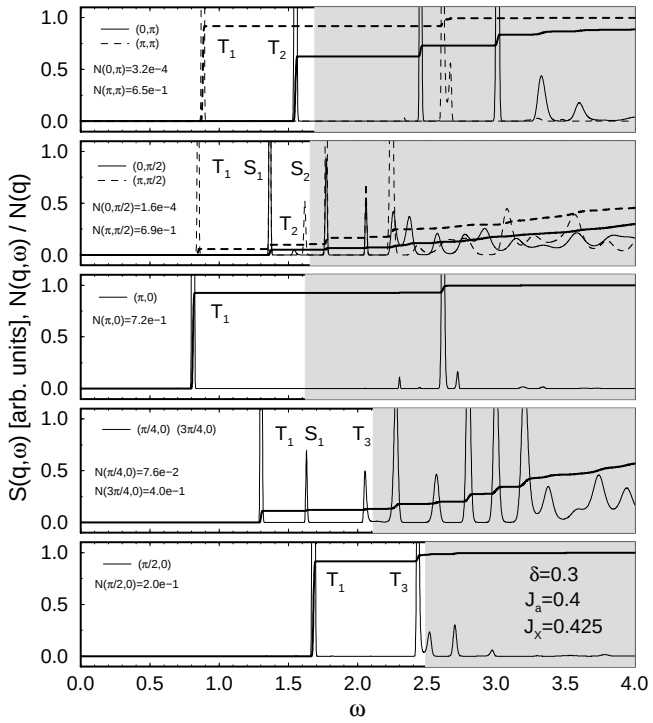

Fig. 6. Dynamical spin structure factor for the 2D model (4×8 system, $\delta = 0.3, J_a = 0.4, J_\times = 0.425$); the lowest singlet and triplet excitations are classified.

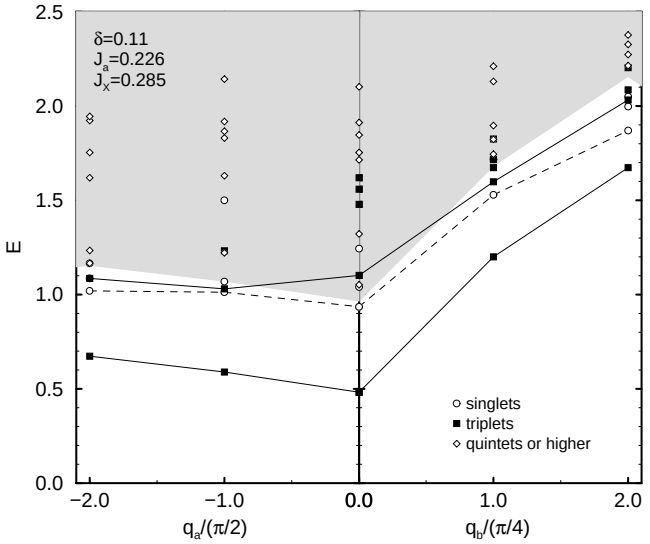

Fig. 7. Low-energy excitations of the 2D model; the system size is 4×8, $\delta = 0.11$, $J_a = 0.226$ and $J_\times = 0.285$.

As in the other figures we shaded the region of an assumed two-magnon continuum. Here the peak structure is expected to merge into a broad absorption band for the infinite system. The poorer resolution and the continously increasing integrated weight in Figure 6 are first signs of this behaviour.

To summarize, using exact diagonalization methods we have shown that a simple ferromagnetic coupling of alternating Heisenberg chains does not provide two well-defined triplet branches as were observed in inelastic neutron scattering experiments on vanadyl pyrophosphate $(VO)_2P_2O_7$. From our experience with frustrated alternating Heisenberg chains, we proposed an alternative model to describe the low-energy physics of $(VO)_2P_2O_7$, introducing a frustrating interchain coupling. Due to the large parameter space and the computational effort for sufficiently extended 2D systems, we made no attempt to fix the parameters for $(VO)_2P_2O_7$, but showed that the proposed model can describe the general feature of two triplet branches below a continuum of states. These triplets exhibit a ferromagnetic (antiferromagnetic) dispersion in *a*- (*b*-) direction. Thus we believe that our model is a good starting point for further analysis.

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Note added in proof

After submission of this contribution a similar study of Uhrig *et al.* [12] appeared at the cond-mat e-print archive. Using perturbation theoretic methods for the same 2D model the authors try to fit the free parameters to the observed magnon dispersion, which yields $(\delta, J_a, J_\times) = (0.115, 0.226, 0.285)$. Performing exact diagonalizations also for this parameter set, we found that the second triplet mode is *not* clearly separated from higher excitations (Fig. 7). Thus we think a better adaption of the model to $(VO)_2P_2O_7$ has still to be done.

References

1. D.C. Johnston, J.W. Johnson, D.P. Goshorn, A.J. Jacobson, Phys. Rev. B **35**, 219 (1987).
2. T. Barnes, J. Riera, Phys. Rev. B **50**, 6817 (1994).
3. A.V. Prokofiev, F. Büllesfeld, W. Assmus, H. Schwenk, D. Wichert, U. Löw, B. Lüthi, Eur. Phys. J. B **5**, 313 (1998).
4. R.S. Eccleston, T. Barnes, J. Brody, J.W. Johnson, Phys. Rev. Lett. **73**, 2626 (1994).
5. D.A. Tennant, S.E. Nagler, A.W. Garrett, T. Barnes, C.C. Torardi, Phys. Rev. Lett. **78**, 4998 (1997).
6. A.W. Garrett, S.E. Nagler, T. Barnes, B.C. Sales, Phys. Rev. B **55**, 3631 (1997).
7. A.W. Garrett, S.E. Nagler, D.A. Tennant, B.C. Sales, T. Barnes, Phys. Rev. Lett. **79**, 745 (1997).
8. G. Bouzerar, A.P. Kampf, G.I. Japaridze, Phys. Rev. B **58**, 3117 (1998); G. Bouzerar, S. Sil, cond-mat/9805042.
9. T. Barnes, J. Riera, D.A. Tennant, cond-mat/9801224.
10. G.S. Uhrig, H.J. Schulz, Phys. Rev. B **54**, R9624 (1996).
11. T. Barnes, E. Dagotto, J. Riera, E.S. Swanson, Phys. Rev. B **47**, 3196 (1993).
12. G.S. Uhrig, B. Normand, cond-mat/9807186.