Susceptibility and excitation spectrum of $(VO)_2P_2O_7$ in ladder and dimer-chain models

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We present numerical results for the magnetic susceptibility of a Heisenberg antiferromagnetic spin ladder as a function of temperature and the spin-spin interaction strengths $J_\perp$ and $J_\parallel$. These are contrasted with bulk-limit results for the dimer chain. A fit to the experimental susceptibility of the candidate spin-ladder compound vanadyl pyrophosphate, $(VO)_2P_2O_7$, gives the parameters $J_\perp = 7.82$ meV and $J_\parallel = 7.76$ meV. With these values we predict a singlet-triplet energy gap of $E_{\rm gap} = 3.9$ meV, and give a numerical estimate of the ladder triplet dispersion relation $\omega(k)$. In contrast, a fit to the dimer-chain model leads to $J_1 = 11.11$ meV and $J_2 = 8.02$ meV, which predicts a gap of $E_{\rm gap} = 4.9$ meV.

I. INTRODUCTION

Quantum antiferromagnetism in lower-dimensional systems has proven to be a very rich subject. Some of the more dramatic developments include the realization that half-integral and integral spin chains have very different excitation spectra,[1] and evidence that two-dimensional antiferromagnetism is a crucial component of high-temperature superconductivity (reviewed in Refs. 2–4).

The Heisenberg spin ladder is interesting theoretically as an intermediary between half-integer ($S=\frac{1}{2}$) and integer ($S=0,1$) spin chains. This system has isotropic nearest-neighbor interactions along the chains ($J_\parallel$) and along the rungs ($J_\perp$) of a ladder geometry,

$$H = J_\parallel \sum_{\leftrightarrow} \mathbf{S}_i \cdot \mathbf{S}_j + J_\perp \sum_{\updownarrow} \mathbf{S}_i \cdot \mathbf{S}_j \ . \tag{1}$$

Previous studies of the Heisenberg spin ladder have discussed the ground-state energy and the dependence of the singlet-triplet energy gap on $J_\perp/J_\parallel \equiv \alpha_l$ in the antiferromagnetic regime,[5–9] the triplet spin-wave dispersion relation,[7] the behavior of the system under doping,[5,10–15] and the dynamical structure factor.[7] Systems of coupled ladders have also been investigated.[11,15] Ferromagnetic rung couplings ($J_\perp < 0, J_\parallel > 0$) have been studied using Lanczos and Monte Carlo techniques and the renormalization group;[16] these references suggest that a gap exists for all $J_\perp < 0$.

In Ref. 7 we presented numerical and analytical results for the ground-state energy and triplet spin-wave dispersion relation of an $S=\frac{1}{2}$ spin ladder, as well as numerical results for the structure function $S(\mathbf{k},\omega)$. We found evidence that a single-triplet energy gap appears for any interchain coupling $J_\perp/J_\parallel > 0$, and that the spin-wave band minimum is at $k=\pi$, but the band is folded so the maximum energy occurs between $k=\pi/2$ (for $J_\perp/J_\parallel = 0$) and $k=0$ (for $J_\perp/J_\parallel = \infty$). The bandwidth was found to be relatively insensitive to the rung coupling $J_\perp$, and varied between $\pi J_\parallel/2$ (for $J_\perp/J_\parallel = 0$) and $2J_\parallel$ (for $J_\perp/J_\parallel = \infty$).

It has been suggested[5,7] that the antiferromagnetic spin ladder may be realized in nature by the antiferromagnet vanadyl pyrophosphate,[17,18] $(VO)_2P_2O_7$. This material has a ladder configuration of $S=\frac{1}{2}$ V^{+4} ions (Fig. 2 of Ref. 18), with spacings of 3.19(1) Å between rung ions and 3.864(2) Å between chain ions, and has a magnetic susceptibility characteristic of an antiferromagnet with an energy scale (from the susceptibility maximum) of ≈ 7 meV. The closely related material $VO(HPO_4)\cdot\frac{1}{2}H_2O$ has isolated V^{+4} ion pairs at a similar separation of 3.10 Å, and is well described magnetically by independent spin-$\frac{1}{2}$ Heisenberg pairs with a coupling (in our conventions) of $J = 7.81$ meV.[19]

Although the $(VO)_2P_2O_7$ lattice clearly shows a ladder configuration of V^{+4} ions, they might, in principle, interact magnetically as a different spin system. This was the case for $Cu(NO_3)_2\cdot\frac{5}{2}H_2O$ (Ref. 20), which was originally considered to be a possible spin ladder but was subsequently found to interact as a dimer chain, described by the Hamiltonian

$$H = \sum_i \{ J_1 \mathbf{S}_{2i} \cdot \mathbf{S}_{2i+1} + J_2 \mathbf{S}_{2i+1} \cdot \mathbf{S}_{2i+2} \} \ , \tag{2}$$

with $J_2/J_1 \equiv \alpha_d$ ($0 \le \alpha_d \le 1$). It was not practical to distinguish between ladder and dimer-chain models of copper nitrate from bulk thermodynamic properties alone, which were found to be very similar for the two systems.[20] The issue was finally decided in favor of the dimer-chain model by proton resonance[21] and neutron-diffraction[22] experiments.

Vanadyl pyrophosphate presents similar ambiguities. Although its susceptibility has been measured and is accurately described by the susceptibility of a dimer chain[18]

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(2), it is widely believed that the ladder Hamiltonian (1) will lead to a very similar $\chi(T)$ and so is not excluded by the good agreement with the dimer chain. Since no theoretical results have been published for the bulk-limit ladder susceptibility in the relevant $J_\perp \sim J_\parallel$ regime, comparison of the experimental susceptibility to the ladder model has not been possible. In this paper we present new numerical results for the bulk-limit susceptibility of ladders and dimer chains, and fit these to the data for $(VO)_2P_2O_7$. As we shall see, these models do indeed give very similar results for the susceptibility, and both give excellent fits to $(VO)_2P_2O_7$ with appropriate parameters. The ladder $\chi(T)$ is slightly preferred, although the differences are probably less important than the approximations made in the models and the experimental errors.

II. SPIN LADDER AND DIMER-CHAIN SUSCEPTIBILITIES

We determine the susceptibility on finite lattices by generating all energy levels $\{E_i\}$ and their multiplicities $\{d_i\}$ in each sector of fixed total S_z, using a Householder algorithm. The susceptibility is then obtained through its relation to the expected squared magnetization, summed over energy levels and total S_z sectors:

$$\chi(T)=g^2\mu_B^2\beta\frac{\sum_{S_z}\sum_i S_z^2 d_i e^{-\beta E_i}}{\sum_{S_z}\sum_i d_i e^{-\beta E_i}} . \tag{3}$$

This approach has the advantage that explicit eigenvectors are not required.

For the ladder geometry we used (3) to determine χ on $2\times L$ lattices with $L=3,4,\ldots,8$ for couplings $J_\perp/J_\parallel=\alpha_l=0.5$, 0.7, 0.9, 1.0, and 1.1 and for a range of $T/J_\parallel$ values; the results for $\alpha_l=1.0$ are shown in Fig. 1(a). To estimate the bulk-limit susceptibility the $2\times L$ results were fitted to the form $\chi_L=\chi_\infty+ae^{-bL}$ at each T and coupling, independently for $L=$odd and $L=$even, since these approached the bulk limit from opposite directions. This gave independent estimates of the bulk-limit susceptibility, which allowed a test of the accuracy of our extrapolation in L. Our bulk-limit estimate was taken (rather arbitrarily) to be a weighted average of $0.6\times$even-L $+0.4\times$odd-L extrapolated values, since the even-L series showed better convergence.

For the dimer chain we considered parameter values $J_2/J_1=\alpha_d=0.2$, 0.4, 0.6, 0.7, and 0.8 and $L=6,8,\ldots,16$; the $\alpha_d=0.7$ results are shown in Fig. 2(a). Here the even dimer number ($L=6,10,14$) and odd dimer number ($L=8,12,16$) series converged on the bulk limit from opposite sides, but the latter series showed much better convergence and hence was used by itself for our bulk-limit estimate.

Our extrapolations indicate that the bulk limit $\chi(T)$ for parameters relevant to $(VO)_2P_2O_7$ is quite accurately determined for temperatures above the susceptibility maximum, but at lower temperatures important finite-size effects remain. The systematic L-extrapolation error is difficult to determine at the lower temperatures, but it

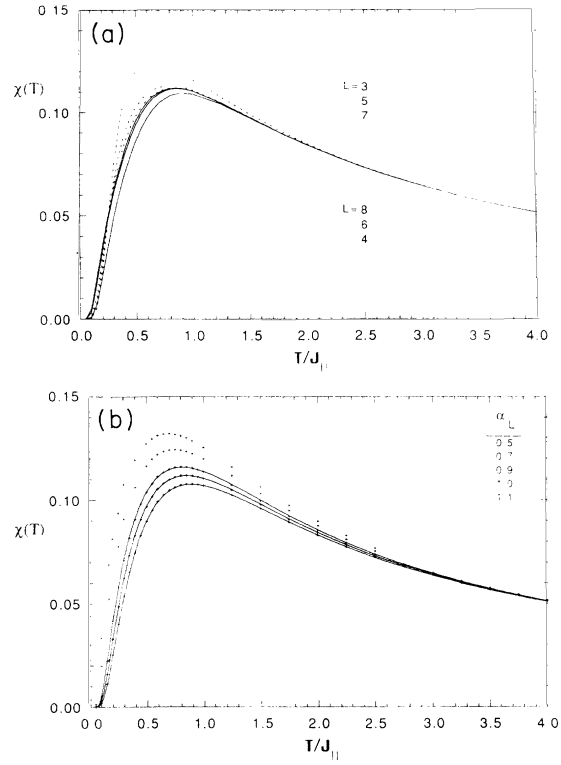

FIG. 1. (a) $2\times L$ finite ladder susceptibility for $L=3,5,7$ (dashed) and $L=4,6,8$ (solid). $\alpha_L=1.0$, χ in units of $g^2\mu_B^2/k_B$. (b) Bulk-limit results for the ladder susceptibility in units of $g^2\mu_B^2/k_B$ and the parametrization Eq. (4). $J_\parallel=1.0$ in both figures.

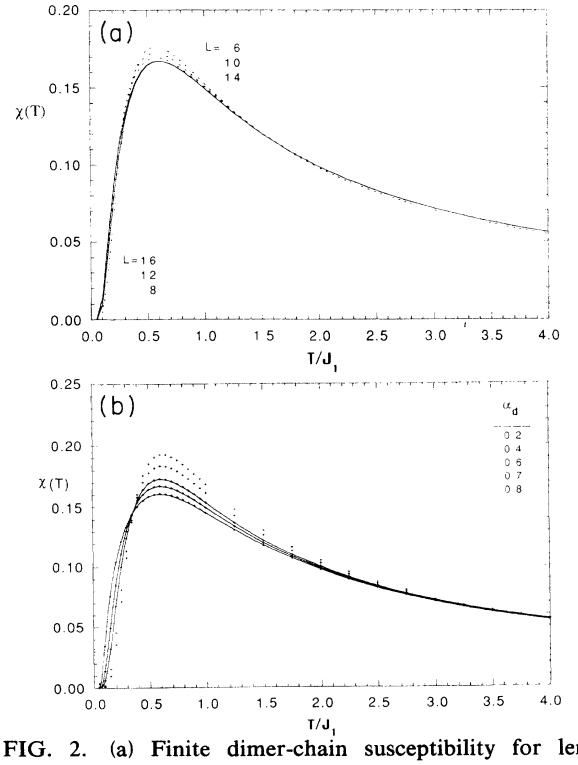

FIG. 2. (a) Finite dimer-chain susceptibility for length $L=6,10,14$ (dashed) and $L=8,12,16$ (solid). $\alpha_d=0.7$, χ in units of $g^2\mu_B^2/k_B$. (b) Bulk-limit results for the dimer-chain susceptibility in units of $g^2\mu_B^2/k_B$ and the parametrization Eq. (4). $J_1=1.0$ in both figures.

is presumably somewhat smaller than the difference $\Delta\chi=|\chi-\chi_{16}|$ between our fitted bulk limit χ and the value χ_{16} measured on the largest (16-spin) lattices. Taking the ladder first, with $\alpha_l=1.0$ and $T/J_\parallel=1.0$ we have $\chi_{16}=0.110\,454$ and $\chi=0.110\,465$, so our systematic extrapolation error should be well below $\Delta\chi=1.1\times10^{-5}$; at $T/J_\parallel=2.0$ this has fallen to $\Delta\chi<10^{-6}$. At smaller T the extrapolation is considerably larger, for example we find $\Delta\chi=8.2\times10^{-4}$ at $T/J_\parallel=0.50$ and $\Delta\chi=6.9\times10^{-4}$ at $T/J_\parallel=0.25$. Thus the systematic error in $\chi(T)$ should be below $\approx10^{-3}$ at these low temperatures. The increased error at low temperatures is compensated somewhat by a decrease in the magnitude of $\chi(T)$, so these inaccuracies have a reduced effect on fitted parameter values. The dimer chain (with $\alpha_d=0.7$) shows significantly better convergence than the ladder, and has $\Delta\chi=1.0\times10^{-4}$ at $T/J_1=0.25$ and $\Delta\chi\le10^{-6}$ for $T/J_1\ge0.5$.

For fitting purposes it is useful to have a parametrization of these results that allows accurate interpolation in T and interaction strengths. We tested several forms and found that the six-parameter function

$$\chi(T)=\frac{c_1}{T}[1+(T/c_2)^{c_3}(e^{c_4/T}-1)]^{-1}\times[1+(c_5/T)^{c_6}]^{-1} \quad (4)$$

adequately describes both the experimental data[23] and the theoretical ladder and dimer-chain susceptibilities over a range of parameters relevant to $(VO)_2P_2O_7$. This form also has the advantage that it incorporates the exponential behavior expected at low temperatures, unlike other parameterizations used previously for the dimer chain, and at high temperature it gives the correct Curie form $g^2\mu_B^2/4k_BT$. (The overall normalization c_1 is identically equal to $g^2\mu_B^2/4k_B$ in all cases, but the V^{4+} ion g factor is unknown *a priori* and is determined when c_1 is fitted to the data.)

For the two theoretical susceptibilities we fitted our numerical bulk-limit results to (4), with each of the coefficients $c_2,\ldots,c_6$ taken to be quadratic in the ratio of the two coupling constants. Preliminary fits to the $(VO)_2P_2O_7$ susceptibility indicated that the values $\alpha_l=1.0$ and $\alpha_d=0.7$ were close to optimum, so we parametrized our bulk limit results in terms of the departure from these values. The fitted coefficients $c_2\ldots c_6$ for the ladder over the range $0.9\lesssim\alpha_l\lesssim1.1$ (with $J_\parallel=1$) were found to be

$$c_2=+2.315-4.035(\alpha_l-1.0)+7.050(\alpha_l-1.0)^2\,, \quad (5a)$$

$$c_3=+0.403-1.025(\alpha_l-1.0)+1.850(\alpha_l-1.0)^2\,, \quad (5b)$$

$$c_4=+0.443+0.225(\alpha_l-1.0)+0.850(\alpha_l-1.0)^2\,, \quad (5c)$$

$$c_5=+0.745-0.390(\alpha_l-1.0)+0.200(\alpha_l-1.0)^2\,, \quad (5d)$$

$$c_6=+1.628-0.110(\alpha_l-1.0)+0.600(\alpha_l-1.0)^2\,, \quad (5e)$$

and for the dimer chain (with $J_1=1$) over the range $0.6\lesssim\alpha_d\lesssim0.8$ we found

$$c_2=+8.145-61.76(\alpha_d-0.7)+406.3(\alpha_d-0.7)^2\,, \quad (6a)$$

$$c_3=+0.562+2.84(\alpha_d-0.7)+7.30(\alpha_d-0.7)^2\,, \quad (6b)$$

$$c_4=+0.456-0.435(\alpha_d-0.7)+1.750(\alpha_d-0.7)^2\,, \quad (6c)$$

$$c_5=+0.592+1.090(\alpha_d-0.7)+0.400(\alpha_d-0.7)^2\,, \quad (6d)$$

$$c_6=+1.663+0.160(\alpha_d-0.7)-1.400(\alpha_d-0.7)^2\,. \quad (6e)$$

Due to the presence of large coefficients this parametrization is not useful far from the parameter ranges cited; if required the coefficients could be determined directly from the bulk-limit numerical results.

Our numerical results for the extrapolated bulk-limit susceptibility of the ladder and dimer chain are shown in Figs. 1(b) and 2(b), respectively, together with the interpolating functions defined by (4)–(6). The interpolating functions reproduce the bulk-limit susceptibility with a typical accuracy of a few times 10^{-4} over the parameter ranges quoted above.

III. COMPARISON WITH THE EXPERIMENTAL $(VO)_2P_2O_7$ SUSCEPTIBILITY

In a previous study Johnston *et al.*[18] presented results for the susceptibility of $(VO)_2P_2O_7$, and noted that the susceptibility of a dimer spin chain gives a very good description of the data for $\alpha_d\equiv J_2/J_1=0.7$ (fixed from an interpolation of theoretical curves for 0.6 and 0.8 from Ref. 20), $J_1=11.32$ meV (hence $J_2=7.93$ meV), and $g=2.00$. (Note that in our conventions the $\{J_n\}$ are twice as large as in Refs. 17 and 18.) A similar coupling of $J=7.81$ meV was determined for $VO(HPO_4)\cdot\frac{1}{2}H_2O$, which consists of isolated V^{+4} dimers,[19] and the g factor of the V^{+4} ion is known to be quite close to 2 from studies of other vanadium phosphates.[24] Of course it is not clear how the fitted dimer-chain parameters relate to $(VO)_2P_2O_7$ if it proves to be a spin ladder.

To confirm these results we fitted our three-parameter (J_1,α_d,g) dimer-chain susceptibility, described by (4) and (6), to the data of Ref. 18, which consists of 606 values for $\chi(T)$ from $T=7.2$ K to 344.34 K. We found the optimum parameter values to be $J_1=11.11$ meV, $\alpha_d=0.722$, and $g=1.99$. These are essentially the parameters found by Johnston *et al.*, and the minor differences are presumably due to the systematic errors in interpolation (perhaps 1% in parameter values). The fitted dimer-chain susceptibility and the data for $(VO)_2P_2O_7$ are shown in Fig. 3.

We similarly fitted the three-parameter $(J_\parallel,\alpha_l,g)$ ladder susceptibility (4), (5) to the experimental $(VO)_2P_2O_7$ $\chi(T)$ over the full temperature range. The optimum ladder parameters were found to be

$$J_\parallel=7.76\text{ meV}\,, \quad (7a)$$

$$\alpha_l\equiv J_\perp/J_\parallel=1.007\,, \quad (7b)$$

$$g=2.03\,. \quad (7c)$$

The proximity of g to 2 provides a plausibility test of the fit, as does the fitted value of $J_\perp=7.82$ meV, which is al-

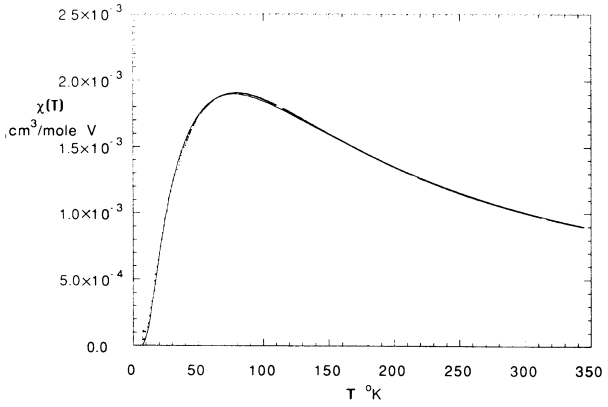

FIG. 3. Dimer-chain fit to $(VO)_2P_2O_7$ susceptibility; $J_1=11.11$ meV, $\alpha_d=0.722$, $g=1.99$.

most identical to the isolated dimer $J=7.81$ meV found previously[19] in $VO(HPO_4)\cdot\frac{1}{2}H_2O$. These results suggest that $(VO)_2P_2O_7$ is very close to a uniform ladder ($J_\perp=J_\parallel$), which is presumably accidental because the rungs are bridged by two oxygens, whereas the chains have only single oxygens between V^{+4} ions. The fitted ladder $\chi(T)$ is shown in Fig. 4, and this model evidently also gives a very good description of the experimental data. The goodness of fit, defined by the residual $f=\sum_i[\chi_{\rm expt}(T_i)-\chi_{\rm thy}(T_i)]^2$, slightly favors the ladder model over the dimer chain. We cannot choose between the models definitively from the susceptibility data, however, because the variation in $(VO)_2P_2O_7$ susceptibility estimated from samples with different annealing histories [Fig. 1(b) of Ref. (17)] is somewhat larger than the difference between predictions of ladder and dimer-chain models.

Finally, to test how well $J_\perp$ and $J_\parallel$ are determined, we studied the residual f in constrained two-parameter fits with g and $J_\parallel$ variable but $\alpha_l=J_\perp/J_\parallel$ fixed. As we changed α_l from the optimum value 1.007 we found that by 0.90 and 1.12 the residual had increased by a factor of 2. As we increase α_l through the range $[0.9,1.1]$ the fitted value of $J_\parallel$ decreases from 8.2 to 7.3 meV, which can be taken as a conservative estimate of the accuracy to which $J_\parallel$ is determined by the susceptibility. The fitted g factor remains close to 2.03 over this range. Outside this range of α_l there is a rapid decrease in the quality of fit, reaching a factor of 5 increase in f by $\alpha_l=0.81$ and 1.24.

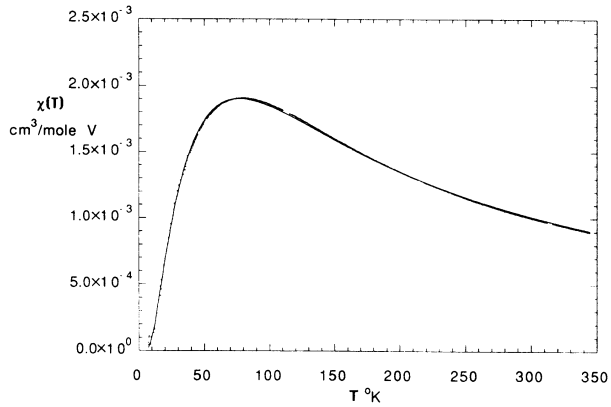

FIG. 4. Ladder fit to $(VO)_2P_2O_7$ susceptibility; $J_\parallel=7.76$ meV, $\alpha_L=1.007$, $g=2.03$.

IV. PREDICTIONS OF THE LADDER AND DIMER MODELS

Since we have determined ladder parameters for $(VO)_2P_2O_7$ from our fit to the susceptibility, we can use the results of Ref. 7 to give predictions for the gap and spin-wave excitation spectrum. From Fig. 2 of that reference we can see that the gap near $J_\perp/J_\parallel=1$ is quite well determined by the Lanczos and Monte Carlo studies. An approximate linear interpolation gives

$$\left.\frac{E_{\rm gap}}{J_\parallel}\right|_{\alpha_l\approx1}\approx0.50+0.65(\alpha_l-1)\ , \tag{8}$$

so for the optimum fitted parameters we predict

$$E_{\rm gap}=3.9\ \text{meV}\ . \tag{9}$$

Over the parameter range $0.9\le J_\perp/J_\parallel\le1.1$ discussed above the predicted gap increases from 3.6 to 4.1 meV.[25]

As was noted in Fig. 3 of Ref. 7, for $J_\perp=J_\parallel$ the minimum energy required to excite a triplet spin wave on the ladder as a function of k resembles the dispersion relation of a spin-$\frac{1}{2}$ chain, except for the presence of excitation gaps. The lowest excitation is at $k=\pi$, where the gap is $\omega(\pi)=0.50J_\parallel$, the maximum is shifted to a $k<\pi/2$, and a secondary minimum is at $k=0$. The dispersion relation is symmetric about $k=\pi$.

A complication not noted in Ref. 7 is that the lowest-lying triplet spin waves with these parameters arise from two distinct bands. The "primary" band, which contains the lowest gap, is odd under chain interchange ($k_\perp=\pi$), and is shown as solid lines for $J_\perp/J_\parallel=0.5$, 1.0, and 2.0 in Fig. 5. For large $J_\perp$ these are excitations of a single rung, with energy $\omega\approx J_\perp$. The "secondary" band (dashed lines in Fig. 5) is even under chain interchange, and for large $J_\perp$ these states consist of two excited rungs (hence

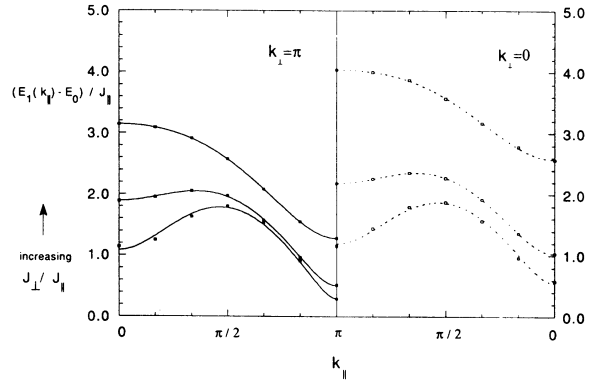

FIG. 5. Primary and secondary bands of $S=1$ spin-wave excitations for $J_\perp/J_\parallel=0.5$, 1.0, 2.0. Both bands are symmetric about $k_\parallel=\pi$.

$\omega' \approx 2J_\perp$ and the even symmetry), with the two $S=1$ excited rungs coupled to give $S_{tot}=1$. Thus the secondary band may be interpreted as the excitation of two spin-wave quanta of the primary band. This interpretation leads us to anticipate several features of the secondary dispersion relation in the bulk limit, for example $\omega'(k=0)=2\omega(k=\pi)$, so the band minimum of the secondary band in $(VO)_2P_2O_7$ should lie at 7.8 meV given our parameters. One may similarly construct the entire secondary $\omega'(k')$ given the primary $\omega(k)$ (assuming there are no bound states), by finding the minimum-energy combination of two quanta with specified k'. Presumably $\omega'(k')$ is the bottom of a two-magnon continuum.

In our representation in Fig. 5 we fitted the function

$$\omega(k)=[\omega(0)^2\cos^2(k/2)+\omega(\pi)^2\sin^2(k/2) + c_0^2\sin(k)^2]^{1/2}, \quad (10)$$

which interpolates between the known analytic chain and dimer limits, to the 2×12 lattice data. [The points $\omega'(k=0)$ in the secondary band showed large finite-size effects, and were replaced by $2\omega(\pi)$ as argued above.] For $J_\perp=J_\parallel=J$ the fitted constants were found to be $\omega(0)=1.890J$, $\omega(\pi)=0.507J$, and $c_0=1.382J$. In Fig. 6 we show the triplet dispersion relation which this parametrization predicts for $(VO)_2P_2O_7$, together with a similar result for the secondary band, using the mean value $J_\perp=J_\parallel=7.79$ meV and the physical lattice spacing. The primary triplet band ranges from 3.9 meV at $k=0.813$ A^{-1} to 16 meV at about 0.3 A^{-1}, and then falls to 15 meV at $k=0$. The secondary band begins at 7.9 meV at $k=0$ and rises to a broad plateau at an energy of about 17–18 meV centered on $k=0.813$ A^{-1}. Structure factor calculations on the 2×12 lattice[26] suggest that both bands should appear most clearly near the $k=\pi$ point (0.813 A^{-1}).

For comparison we quote predictions for the triplet spin-wave dispersion relation in the dimer-chain model. Of course the lattice spacing a and the direction of the continuous momentum variable k are problematical for $(VO)_2P_2O_7$ in the dimer model because there is no obvious dimer-chain interaction pathway. Since the dimer unit cell has length $2a$ the dispersion relation repeats with period $\Delta k=\pi/a$; this implies that the two different gaps we found for the ladder at 0 and π/a are equal in the dimer chain. Another characteristic feature of the dimer-chain dispersion relation is that it is symmetric about $\pi/2a$, due to inversion symmetry. For the parameters J_1, J_2, and g found in our susceptibility fits the dimer-chain model predicts a somewhat larger gap of $E_{gap}\approx 0.44J_1=4.9$ meV and a bandwidth of ≈ 11 meV. It is interesting that one can apparently distinguish between the dimer-chain and ladder models by an accurate measurement of the gap alone, using parameters derived from susceptibility fits.

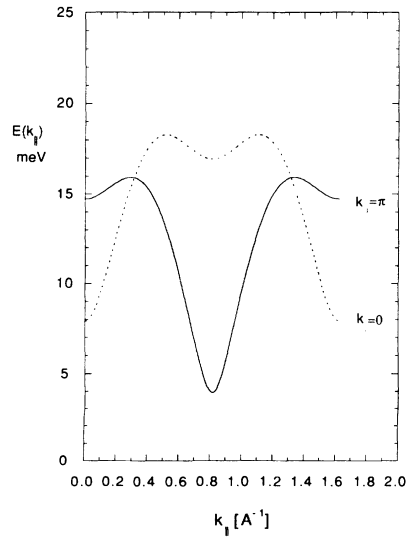

FIG. 6. Primary and secondary triplet spin-wave excitation spectra for $J_\parallel=J_\perp=7.79$ meV and $(VO)_2P_2O_7$ lattice spacing.

V. SUMMARY AND CONCLUSIONS

In this paper we used numerical techniques to study the susceptibility of a Heisenberg antiferromagnetic spin ladder and a dimerized Heisenberg spin chain. We used exact numerical diagonalization to generate all energy eigenvalues and their degeneracies, which were then used to determine $\chi(T)$ on ladders and dimer chains of up to 16 spins. We presented results for a range of temperatures and interaction ratios $J_\perp/J_\parallel$ (ladder) and J_2/J_1 (chain). These were extrapolated to give bulk-limit estimates, which we parametrized using an interpolating function. We fitted the bulk limit $\chi(T)$ to the susceptibility data for $(VO)_2P_2O_7$, which is a candidate spin-ladder system but is known to be accurately described by the dimer-chain susceptibility. Our best fit to the dimer-chain model accurately reproduces previous parameter values. Our best fit to the ladder is in slightly better agreement with the data, and indicates that $(VO)_2P_2O_7$ has very similar $J_\perp$ and $J_\parallel$ values. With these parameters we give numerical predictions for the spin-wave excitation gap of $(VO)_2P_2O_7$ and for other properties of the spin-wave dispersion relation.

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