Origin of the second coherent peak in the dynamical structure factor of an asymmetric spin-ladder

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(Received 17 August 2006; revised manuscript received 27 November 2006; published 20 March 2007)

Appearance of the second coherent peak in the dynamical structure factor of an asymmetric spin-ladder is suggested. The general arguments are confirmed by the first-order (with respect to the asymmetry) calculation for a spin-ladder with singlet-rung ground state. Based on this result, an interpretation is proposed for the inelastic neutron scattering data in the spin gap compound CuHpCl, for which some of the corresponding interaction constants are estimated.

DOI: 10.1103/PhysRevB.75.094420 PACS number(s): 75.10.Jm, 75.50.Ee, 75.40.Gb

I. INTRODUCTION

Inelastic neutron scattering is an effective method for analysis of low-energy excitations in low-dimensional spin systems.[1] The dynamical structure factor (DSF) obtained from this experiment produces an essential information about the low-energy spectrum. Sharp peaks of the DSF line shape correspond to coherent modes, while broad bands originate from incoherent excitation continuums.

Theoretical study of a spin ladder DSF was developed in the papers of Refs. 2 and 3. A strong antiferromagnetic rung coupling corresponds to the DSF with a single coherent-mode peak,[2] while for a weak coupling the line shape has only an incoherent background.[3] The models studied in Refs. 2 and 3 are symmetric under exchange of the legs because their couplings along both legs are equal to each other and the same is true for the couplings along both diagonals. Such requirement fails for an asymmetric spin-ladder.

The compound $Cu_2(C_5H_{12}N_2)_2Cl_4$ (CuHpCl) was first interpreted as an asymmetric spin ladder[4] (with nonequal couplings along diagonals). However, neutron scattering[5,6] revealed two coherent peaks in the DSF line shape for CuHpCl. Since this type of behavior does not agree with the results of Refs. 2 and 3 (obtained for the symmetric case), it was suggested in Ref. 6 that the magnetic structure of CuHpCl is inconsistent with the spin-ladder model.

In this paper, we show the principal difference between excitation spectra of symmetric and asymmetric spin-ladders and we suggest argumentation confirming the existence of the second coherent peak in DSF of an asymmetric spin-ladder. As an example, we calculate the DSF for a weakly asymmetric spin ladder with singlet-rung ground state and produce an evidence for the second coherent peak.

II. HAMILTONIAN FOR AN ASYMMETRIC SPIN LADDER

The general Hamiltonian of an asymmetric spin-ladder has the following form:

$$\hat{H}=\hat{H}^{symm}+\hat{H}^{asymm}, \tag{1}$$

where $\hat{H}^{symm}=\sum_n H^{symm}_{n,n+1}$ and $\hat{H}^{asymm}=\sum_n H^{asymm}_{n,n+1}$. The local Hamiltonian densities are given by the following expressions:

$$H^{symm}_{n,n+1}=H^{rung}_{n,n+1}+H^{leg}_{n,n+1}+H^{frust}_{n,n+1}+H^{cyc}_{n,n+1}, \tag{2}$$

where

$$H^{rung}_{n,n+1}=J_{\perp}\mathbf{S}_{1,n}\cdot\mathbf{S}_{2,n},$$

$$H^{leg}_{n,n+1}=J^{symm}_{\parallel}(\mathbf{S}_{1,n}\cdot\mathbf{S}_{1,n+1}+\mathbf{S}_{2,n}\cdot\mathbf{S}_{2,n+1}),$$

$$H^{frust}_{n,n+1}=J^{symm}_{frust}(\mathbf{S}_{1,n}\cdot\mathbf{S}_{2,n+1}+\mathbf{S}_{2,n}\cdot\mathbf{S}_{1,n+1}),$$

$$H^{cyc}_{n,n+1}=J_c[(\mathbf{S}_{1,n}\cdot\mathbf{S}_{1,n+1})(\mathbf{S}_{2,n}\cdot\mathbf{S}_{2,n+1})+(\mathbf{S}_{1,n}\cdot\mathbf{S}_{2,n})\times(\mathbf{S}_{1,n+1}\cdot\mathbf{S}_{2,n+1})-(\mathbf{S}_{1,n}\cdot\mathbf{S}_{2,n+1})(\mathbf{S}_{2,n}\cdot\mathbf{S}_{1,n+1})], \tag{3}$$

and

$$H^{asymm}_{n,n+1}=J^{asymm}_{\parallel}(\mathbf{S}_{1,n}\cdot\mathbf{S}_{1,n+1}-\mathbf{S}_{2,n}\cdot\mathbf{S}_{2,n+1})+J^{asymm}_{frust}(\mathbf{S}_{1,n}\cdot\mathbf{S}_{2,n+1}-\mathbf{S}_{2,n}\cdot\mathbf{S}_{1,n+1}). \tag{4}$$

This structure is schematically represented in Fig. 1.

It is convenient to extract from the general $\hat{H}^{symm}$ the "singlet-rung" part $\hat{H}^{s-r}$ commuting with the following operator:

$$\hat{Q}=\sum_n Q_n, \tag{5}$$

where $Q_n=\frac{1}{2}(\mathbf{S}_{1,n}+\mathbf{S}_{2,n})^2$. The commutativity condition,

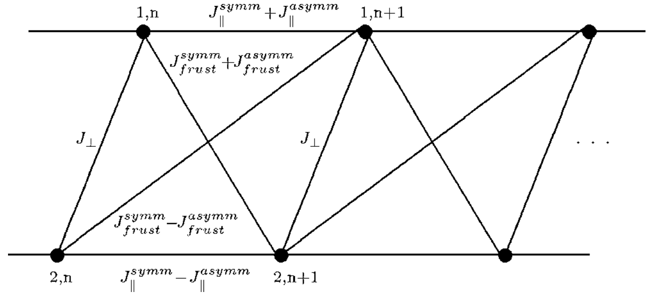

FIG. 1. Schematic of the magnetic structure of an asymmetric spin-ladder.

$$[\hat{H}^{s\text{-}r},\hat{Q}]=0, \tag{6}$$

or in equivalent form,

$$[H^{s\text{-}r}_{n,n+1},Q_n+Q_{n+1}]=0, \tag{7}$$

results to the following restriction on the interaction constants for $\hat{H}^{s-r}$:

$$J^{s\text{-}r}_{frust}=J^{s\text{-}r}_{\parallel}-\frac{1}{2}J^{s\text{-}r}_{c}. \tag{8}$$

According to Eq. (8), the Hilbert space for $\hat{H}^{s-r}$ splits on the infinite set of eigenspaces corresponding to different eigenvalues of $\hat{Q}$,[7,8]

$$\mathcal{H}=\sum_{m=0}^{\infty}\mathcal{H}^m, \quad \hat{Q}|_{\mathcal{H}^m}=m. \tag{9}$$

The one-dimensional subspace $\mathcal{H}^0$ is generated by the single vector,

$$|0\rangle=\prod_n|0\rangle_n, \tag{10}$$

where $|0\rangle_n$ is the nth rung singlet. The following restrictions,

$$J^{s\text{-}r}_{\perp}>2J^{s\text{-}r}_{\parallel}, \quad J^{s\text{-}r}_{\perp}>\frac{5}{2}J^{s\text{-}r}_{c}, \quad J^{s\text{-}r}_{\perp}+J^{s\text{-}r}_{\parallel}>\frac{3}{4}J^{s\text{-}r}_{c}, \tag{11}$$

guarantee that the state [Eq. (10)] is the (singlet rung) ground state for $\hat{H}^{s-r}$. The operator $\hat{Q}$ has a sense of the magnon number[8] associated with $\hat{H}^{s-r}$.

We will use the decomposition,

$$\hat{H}^{symm}=\hat{H}^{s\text{-}r}+\Delta\hat{H}^{symm}, \tag{12}$$

with the following additional restrictions on the interaction constants of ΔH^{symm}:

$$\Delta J^{symm}_{\perp}=\Delta J^{symm}_{c}=0, \quad \Delta J^{symm}_{\parallel}=-\Delta J^{symm}_{frust}. \tag{13}$$

These restrictions guarantee the uniqueness of the decomposition [Eq. (12)]. Moreover, under Eqs. (13) and (4) the local exchange relations between $\Delta\hat{H}^{symm}$, $\hat{H}^{asymm}$, and $\hat{Q}$ have the following forms:

$$\{\Delta H^{symm}_{n,n+1},Q_n+Q_{n+1}\}=2\Delta H^{symm}_{n,n+1}, \tag{14}$$

$$\{H^{asymm}_{n,n+1},Q_n+Q_{n+1}\}=3H^{asymm}_{n,n+1}, \tag{15}$$

where {.,.} means anticommutator.

As follows from Eq. (14), the term $\Delta\hat{H}^{symm}$ does not mix even and odd components in Eq. (9). Therefore, the Hilbert space $\mathcal{H}$ splits on two invariant subspaces of the operator $\hat{H}^{symm}$,

$$\mathcal{H}=\mathcal{H}^{even}+\mathcal{H}^{odd}, \quad \mathcal{H}^{even}=\sum_{m=0}^{\infty}\mathcal{H}^{2m}, \quad \mathcal{H}^{odd}=\sum_{m=0}^{\infty}\mathcal{H}^{2m+1}. \tag{16}$$

From Eq. (15) it follows that $\hat{H}^{asymm}$ mixes $\mathcal{H}^{even}$ and $\mathcal{H}^{odd}$; however, on the sector $\mathcal{H}^0$ its action is trivial. Really, according to Eq. (15), $H^{asymm}_{n,n+1}|0\rangle_n|0\rangle_{n+1}$ have to lie in the sector with $Q_n+Q_{n+1}=3$ that is impossible because the operator Q_n has only eigenvalues 0 and 1. So we have

$$\hat{H}^{asymm}|0\rangle=0. \tag{17}$$

More detailed analysis of the 16×16 matrix H^{asymm} (which represents the action of $H^{asymm}_{n,n+1}$ on the product of nth and $n+1$ rungs) shows that it has only three (degenerative) eigenvalues: 0 and $\pm\sqrt{(J^{asymm}_{\parallel})^2+(J^{asymm}_{frust})^2}$. Therefore, for small $J^{asymm}_{\parallel}$ and J^{asymm}_{frust} the state $|0\rangle$ remains as the ground state for $\hat{H}^{s-r}+\hat{H}^{asymm}$.

Now we may suggest the following interpretation for the appearance of the second coherent mode in the DSF line shape of an asymmetric spin-ladder. It is known[7–9] that in the strong rung-coupling regime, an excitation spectrum of an symmetric spin-ladder has coherent modes of two types, the one-magnon triplet state lying in $\mathcal{H}^{odd}$ and three-bound two-magnon states (with total spin 0,1,2) lying in $\mathcal{H}^{even}$. The ground state also lies in $\mathcal{H}^{even}$. In the Born approximation, a scattering neutron creates a new state by flipping a single elementary spin. It is a principal fact that the excited state lies in $\mathcal{H}^{odd}$. For this reason, in the symmetric case, only the subspace $\mathcal{H}^{even}$ excites during the scattering process. However, even a little asymmetry results to excitations from $\mathcal{H}^{even}$ and, in particular, the bound two-magnon mode with a total spin of 1 which is respective for the appearance of the second coherent peak in the DSF.

In the next sections, we shall confirm our arguments by studying the simplest model for which $\Delta\hat{H}^{symm}=0$ and the ground state exactly has form (10).

III. ONE AND TWO-MAGNON STATES FOR $\hat{H}^{s-r}$

The eigenstates of $\hat{H}^{s-r}$ in the sectors with $\hat{Q}=1$ and $\hat{Q}=2$ may be obtained exactly.[7,8] From now we shall focus on this special model omitting the upper indices $s-r$ or *symm* in the notation of interaction constants J_c, $J_{\parallel}$, and J_{frust}. In other words, we shall study the model [Eqs. (2) and (3)] with additional restrictions [Eqs. (8) and (11)] on $J_{\perp}$, $J_{\parallel}$, J_{frust}, and J_c.

According to the following formulas:

$$H^{s-r}_{n,n+1}|0\rangle_n|1\rangle^{\alpha}_{n+1}=\left(\frac{1}{2}J_{\perp}-\frac{3}{4}J_c\right)|0\rangle_n|1\rangle^{\alpha}_{n+1}+\frac{J_c}{2}|1\rangle^{\alpha}_n|0\rangle_{n+1},$$

$$H^{s-r}_{n,n+1}|1\rangle^{\alpha}_n|0\rangle_{n+1}=\left(\frac{1}{2}J_{\perp}-\frac{3}{4}J_c\right)|1\rangle^{\alpha}_n|0\rangle_{n+1}+\frac{J_c}{2}|0\rangle_n|1\rangle^{\alpha}_{n+1}, \tag{18}$$

$$H^{s-r}_{n,n+1}\varepsilon_{\alpha\beta\gamma}|1\rangle^{\beta}_n|1\rangle^{\gamma}_{n+1}=(J_{\perp}-J_{\parallel}-J_c/4)\varepsilon_{\alpha\beta\gamma}|1\rangle^{\beta}_n|1\rangle^{\gamma}_{n+1}, \tag{19}$$

where $\alpha,\beta,\gamma=1,2,3$ and $|1\rangle_n^\alpha$ is the triplet associated with an nth rung,

$$|1\rangle_n^\alpha=(\mathbf{S}_{1,n}^\alpha-\mathbf{S}_{2,n}^\alpha)|0\rangle,\quad(\mathbf{S}_{1,n}^\alpha+\mathbf{S}_{2,n}^\alpha)|1\rangle_n^\beta=i\varepsilon_{\alpha\beta\gamma}|1\rangle_n^\gamma,\tag{20}$$

one- and (spin 1) two-magnon states for $\hat{H}^{s-r}$ have the following forms:[7,8]

$$|k,magn\rangle_0^\alpha=\frac{1}{\sqrt{N}}\sum_n e^{ikn}\cdots|0\rangle_{n-1}|1\rangle_n^\alpha|0\rangle_{n+1}\cdots,$$

$$|k_1,k_2,scatt\rangle_0^\alpha=Z_{scatt}^{-1}(k_1,k_2)\sum_{m=-\infty}^{\infty}\sum_{n=m+1}^{\infty}\varepsilon_{\alpha\beta\gamma}\times a^{scatt}(m,n;k_1,k_2)\cdots|1\rangle_m^\beta\cdots|1\rangle_n^\gamma\cdots,$$

$$|k,bound\rangle_0^\alpha=Z_{bound}^{-1}(k)\sum_{m=-\infty}^{\infty}\sum_{n=m+1}^{\infty}\varepsilon_{\alpha\beta\gamma}\times a^{bound}(m,n;k)\cdots|1\rangle_m^\beta\cdots|1\rangle_n^\gamma\cdots,\tag{21}$$

where

$$a^{scatt}(m,n;k_1,k_2)=C_{12}e^{i(k_1m+k_2n)}-C_{21}e^{i(k_2m+k_1n)},$$

$$a^{bound}(m,n;k)=e^{iu(m+n)+v(m-n)},\quad u=\frac{k}{2}+\left(1-\frac{\Delta_1}{|\Delta_1|}\right)\frac{\pi}{2}.\tag{22}$$

Here, $C_{ab}=\cos[(k_a+k_b)/2]-\Delta_1e^{i[(k_a-k_b)/2]}$, $\Delta_1=5/4-J_\parallel/J_c$, $v>0$, and

$$\cos\frac{k}{2}=|\Delta_1|e^{-v}.\tag{23}$$

The normalization factors,

$$Z_{scatt}(k_1,k_2)=2N\sqrt{\cos^2\frac{k_1+k_2}{2}-2\Delta_1\cos\frac{k_1+k_2}{2}\cos\frac{k_1-k_2}{2}+\Delta_1^2},\quad Z_{bound}(k)=\sqrt{2\frac{N\cos^2\frac{k}{2}}{\Delta_1^2-\cos^2\frac{k}{2}}},\tag{24}$$

depend on N the number of rungs.

The corresponding dispersion laws are the following:

$$E^{magn}(k)=J_\perp-\frac{3}{2}J_c+J_c\cos k,\tag{25}$$

$$E^{scatt}(k_1,k_2)=2J_\perp-3J_c+J_c(\cos k_1+\cos k_2),\tag{26}$$

$$E^{bound}(k)=2J_\perp+(\Delta_1-3)J_c+\frac{J_c}{\Delta_1}\cos^2\frac{k}{2}.\tag{27}$$

As follows from Eq. (25), the one-magnon gap E_{gap}^{magn} and the one-magnon zone width ΔE^{magn} are given by the following formulas:

$$E_{gap}^{magn}=J_\perp-\frac{3}{2}J_c-|J_c|,\quad\Delta E^{magn}=2|J_c|.\tag{28}$$

IV. FIRST-ORDER DSF FOR $\hat{H}^{s-r}+\hat{H}^{asymm}$

From Eqs. (4) and (20) it follows that

$$H_{n,n+1}^{asymm}\varepsilon_{\alpha\beta\gamma}|1\rangle_n^\beta|1\rangle_{n+1}^\gamma=-i(J_\parallel^{asymm}-J_{frust}^{asymm})|1\rangle_n^\alpha|0\rangle_{n+1}+i(J_\parallel^{asymm}+J_{frust}^{asymm})|0\rangle_n|1\rangle_{n+1}^\alpha,\tag{29}$$

so,

$${}_0^\alpha\langle q,magn|\hat{H}^{asymm}|k_1,k_2,scatt\rangle_0^\beta=\frac{2iJ^{asymm}(q)\cos\frac{q}{2}\sin\frac{k_1-k_2}{2}\delta_{k_1+k_2\,q}\delta_{\alpha\beta}}{\sqrt{N\left(\cos^2\frac{q}{2}-2\Delta_1\cos\frac{q}{2}\cos\frac{k_1-k_2}{2}+\Delta_1^2\right)}},$$

$${}_0^\alpha\langle q,magn|\hat{H}^{asymm}|k,bound\rangle_0^\beta=\sqrt{2}iJ^{asymm}(q)\frac{\sqrt{\Delta_1^2-\cos^2\frac{q}{2}}}{|\Delta_1|}\delta_{kq}\delta_{\alpha\beta},\tag{30}$$

where

$$J^{asymm}(q)=J_{frust}^{asymm}\cos\frac{q}{2}-iJ_\parallel^{asymm}\sin\frac{q}{2}.\tag{31}$$

Considering $\hat{H}^{asymm}$ as a small perturbation, we may obtain the corresponding corrections for the one- and two-magnon states. In the simplest case when the one-magnon mode does not intersect the two-magnon sector, all the first-order corrections to one- and two-magnon dispersions vanish. First-order one-magnon contributions to the $S=1$ two-magnon states are the following:

$$|k_1,k_2,scatt\rangle_1 = \frac{2iJ^{asymm}(k_1+k_2)\cos\frac{k_1+k_2}{2}\sin\frac{k_1-k_2}{2}}{\sqrt{N\left(\cos^2\frac{k_1+k_2}{2}-2\Delta_1\cos\frac{k_1+k_2}{2}\cos\frac{k_1-k_2}{2}+\Delta_1^2\right)}} \times\frac{1}{[E^{scatt}(k_1,k_2)-E^{magn}(k_1+k_2)]}|k_1+k_2,magn\rangle_0,$$

$$|k,bound\rangle_1=\sqrt{2}i\frac{\sqrt{\Delta_1^2-\cos^2\frac{k}{2}}J^{asymm}(k)}{\Delta_1[E^{bound}(k)-E^{magn}(k)]}|k,magn\rangle_0. \tag{32}$$

We use the following expression for the zero-temperature dynamical structure factor,[1,5,6]

$$S_{\alpha\beta}(\mathbf{q},\omega)=\lim_{N\to\infty}\frac{1}{N}\sum_{\mu}\langle 0|\hat{\mathbf{S}}^{\alpha}(\mathbf{q})|\mu\rangle\langle\mu|\hat{\mathbf{S}}^{\beta}(-\mathbf{q})|0\rangle\delta(\omega-E_{\mu}), \tag{33}$$

where $\hat{\mathbf{S}}(\mathbf{q})$ is the Fourier transformation of spin associated with the two-dimensional vector $\mathbf{q}=(q,q_{rung})$. Here, q and q_{rung} are the corresponding leg and rung components. Since the latter has only two possible values, 0 and π, we may study them separately,

$$\hat{\mathbf{S}}(q,0)=\sum_{n}e^{-iqn}(\mathbf{S}_{1,n}+\mathbf{S}_{2,n}),$$

$$\hat{\mathbf{S}}(q,\pi)=\sum_{n}e^{-iqn}(\mathbf{S}_{1,n}-\mathbf{S}_{2,n}). \tag{34}$$

According to the following two formulas,

$$[\hat{Q},\hat{\mathbf{S}}(q,0)]=0,\quad \{\hat{Q},\hat{\mathbf{S}}(q,\pi)\}=\hat{\mathbf{S}}(q,\pi), \tag{35}$$

we may reduce the matrix elements in Eq. (33),

$$\langle\mu|\hat{\mathbf{S}}(q,0)|0\rangle=0,\quad \langle\mu|\hat{\mathbf{S}}(q,\pi)|0\rangle=\sum_{\nu\in\mathcal{H}^1}\langle\mu|\nu\rangle\langle\nu|\hat{\mathbf{S}}(q,\pi)|0\rangle, \tag{36}$$

so $S_{\alpha\beta}(q,0,\omega)=0$. For calculation of $S_{\alpha\beta}(q,\pi,\omega)$, let us note that from Eqs. (20) and (21) it follows that

$$\langle 0|\hat{\mathbf{S}}^{\alpha}(q,\pi)|k,magn\rangle_0^{\beta}=\sqrt{N}\delta_{\alpha\beta}\delta_{kq}, \tag{37}$$

so, the DSF has a purely diagonal form, $S_{\alpha\beta}(q,\pi,\omega)=\delta_{\alpha\beta}S(q,\pi,\omega)$, where

$$S(q,\pi,\omega)=\sum_{\mu}|\langle\mu|q,magn\rangle_0^3|^2\delta(\omega-E_{\mu}). \tag{38}$$

The unperturbed DSF corresponding only to $\hat{H}^{s-r}$ consists of a single one-magnon coherent peak

$$S^{(0)}(q,\pi,\omega)=\delta[\omega-E^{magn}(q)]. \tag{39}$$

In the first order with respect to the asymmetry, we have to take into account only the two-magnon contributions. Using the substitution $2\pi\Sigma_k\to N\int_{-\pi}^{\pi}dk$, we obtain the following formula:

$$S^{(1)}(q,\pi,\omega)=A_{bound}(q)\delta[\omega-E^{bound}(q)]+A_{scatt}(q,\omega), \tag{40}$$

where

$$A_{bound}(q)=\frac{2|J^{asymm}(q)|^2\left(\Delta_1^2-\cos^2\frac{q}{2}\right)}{\Delta_1^2[E^{magn}(q)-E^{bound}(q)]^2}, \tag{41}$$

$$A_{scatt}(q,\omega)=\frac{2|J^{asymm}(q)|^2\left[\cos^2\frac{q}{2}-x^2(\omega)\right]\Theta[1-x^2(\omega)]}{\pi\left[\cos^2\frac{q}{2}-2\Delta_1x(\omega)+\Delta_1^2\right][\omega-E^{magn}(q)]^2}. \tag{42}$$

Here, $\Theta(x)$ is the step function and $x(\omega)=(\omega-2J_{\perp}+3J_c)/(2J_c)$.

The first term in Eq. (40) corresponds to the second coherent peak carried from the sector $\mathcal{H}^{even}$.

Formulas (41) and (42) will be correct only when the energy of the one-magnon mode is smaller than the energy of the bound state and the lower bound of the two-magnon continuum. Contrary to the asymmetry mixing between $\mathcal{H}^1$ and $\mathcal{H}^2$, any intersection of the one- and two-magnon scattering sectors will result to magnon decay.[10] In order to avoid this possibility, we shall obtain the "nonintersection" condition.

According to Eqs. (25) and (26)

$$2J_{\perp}-3J_c-2|J_c|\cos\frac{k_1+k_2}{2} \leqslant E^{scatt}(k_1,k_2)\leqslant 2J_{\perp}-3J_c+2|J_c|\cos\frac{k_1+k_2}{2}, \tag{43}$$

and the condition $E^{magn}(k_1+k_2)<E^{scatt}(k_1,k_2)$ reduces to the following form, $J_c[\cos k/2+|J_c|/(2J_c)]^2<J_{\perp}/2$. This inequality will be automatically satisfied for $J_c<0$, while for $J_c>0$ it results to $2J_{\perp}>9J_c$ or, using Eq. (28), to an equivalent form,

$$E_{gap}^{magn}>\Delta E^{magn}. \tag{44}$$

The last formula has a clear interpretation. Really, $E_{gap}^{magn}-\Delta E^{magn}$ measures the difference between the one- and two-magnon sectors. When it is satisfied, these sectors do not intersect, a magnon decay is impossible, and formula (42) is correct.

V. COMPARISON WITH THE EXPERIMENTAL DATA FOR CuHpCl

As it was suggested in Ref. 5, the compound CuHpCl corresponds to the case $J_{\parallel}^{asymm}=0$, and $J_{frust}^{asymm}=J_{frust}$. In other words, it may be described by the Hamiltonian

$$H^{\mathrm{CuHpCl}}_{n,n+1} = H^{rung}_{n,n+1} + H^{leg}_{n,n+1} + H^{diag}_{n,n+1} + H^{cyc}_{n,n+1}, \quad (45)$$

where the terms $H^{rung}_{n,n+1}$, $H^{leg}_{n,n+1}$, and $H^{cyc}_{n,n+1}$ are given by Eq. (3) and

$$H^{diag}_{n,n+1} = J_{diag}\mathbf{S}_{1,n}\mathbf{S}_{2,n+1}. \quad (46)$$

Here, $J_{diag}=J_{frust}+J^{asymm}_{frust}=2J_{frust}$.

If one suggests that the state [Eq. (10)] is the exact ground state, then condition (8) reduces to

$$2J_{diag} = J_{\parallel} - \frac{1}{2}J_c. \quad (47)$$

Under this condition (however, not proven experimentally), it is possible to estimate the parameters $J_{\perp}$ and J_c from formulas (25) and (28) and experimental data. As it was presented in Ref. 11, $E^{magn}_{gap} \approx 10.8$ K corresponds to $k=\pi$; however, as it was shown in Ref. 12 by $k=0$ electron spin resonance (ESR) measurements $E_{gap}+2\Delta E^{magn}=13.1$ K. These data agree with the neutron scattering experiments.[5,6] From Eqs. (25) and (28) it follows that $J_c \approx 1.2$ K and $J_{\perp} \approx 13.8$ K.

Unfortunately, any quantitative interpretation fails for the neutron scattering data obtained in Ref. 6. Really, all the scans presented here correspond to the scattering with $q_{rung}=0$. However, as it was shown in the previous section, $S_{\alpha\beta}(q,0,\omega)=0$. Therefore, the appearance of the scattering peaks in Figs. 9 and 10(a) of Ref. 6 may be explained only by a deviation of the initial state of the ladder from the singlet-rung vacuum [Eq. (10)]. The strength of this deviation may be estimated only by comparison of the data presented in Ref. 6 with the same one related to the scattering with $q_{rung}=\pi$. However, the latter is not yet obtained.

In spite of the quantitative disagreement at $q_{rung}=0$, our argumentation qualitatively confirms the appearance of the second coherent peak in the structure factor.

VI. SUMMARY

In this paper, we have demonstrated the principal difference between the excitation spectra of symmetric and asymmetric spin-ladders. For the symmetric one, the Hilbert space splits into two invariant subspaces, $\mathcal{H}^{even}$ and $\mathcal{H}^{odd}$. In this case, only the sector $\mathcal{H}^{odd}$ gives a nonzero contribution to the dynamical structure factor. However, the picture is quite different for an asymmetric spin-ladder. The asymmetry term mixes both the subspaces and the two-magnon bound state from $\mathcal{H}^{even}$ contribute to the DSF resulting to the appearance of the second coherent peak.

As an illustration, we have obtained the first-order DSF for the special model of asymmetric spin-ladder with exact singlet-rung ground state. The suggested model was applied to the probably asymmetric spin-ladder compound CuHpCl for which the existence of the second coherent peak was observed experimentally. Despite the full agreement between our special model and the experimental data was not confirmed, some of the interaction constants were estimated from the inelastic neutron scattering and ESR data.

ACKNOWLEDGMENT

The authors are very grateful to S. V. Maleev for helpful discussions.

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