Dynamical structure factor in copper benzoate and other spin-1/2 antiferromagnetic chains

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(Received 11 April 2003; published 8 August 2003)

Recent experiments of the quasi-one-dimensional spin-$\frac{1}{2}$ antiferromagnet copper benzoate established the existence of a magnetic field induced gap. The observed neutron scattering intensity exhibits resolution limited peaks at both the antiferromagnetic wave number and at incommensurate wave numbers related to the applied magnetic field. We determine the ratio of spectral weights of these peaks within the framework of a low-energy effective field theory description of the problem.

DOI: 10.1103/PhysRevB.68.064410 PACS number(s): 75.10.Jm

I. INTRODUCTION

Recent experiments[1,2] have investigated the behavior of the quasi-one-dimensional spin-$\frac{1}{2}$ antiferromagnet copper benzoate in a magnetic field. Neutron scattering experiments[2] established the existence of field-dependent incommensurate low-energy modes in addition to low-energy modes at the antiferromagnetic wave number. The incommensurability was found to be consistent with the one predicted by the exact solution of the Heisenberg model in a magnetic field. However, the system exhibited an unexpected excitation gap, induced by the applied field. A theory for this effect was put forward by Oshikawa and Affleck in Ref. 3. Application of a uniform magnetic field $\boldsymbol{H}$ induces a staggered field perpendicular to $\boldsymbol{H}$. The staggered field is generated both by a staggered g-tensor[4] and a Dzyaloshinskii-Moriya (DM) interaction. The same physical mechanism has been found also in other materials.[5] The effective Hamiltonian describing such field-induced gap systems is given by[3]

$$\hat{H}=\sum_i \; J\boldsymbol{S}_i\cdot\boldsymbol{S}_{i+1}-HS_i^z+h(-1)^iS_i^x, \tag{1}$$

where

$$h=\gamma H. \tag{2}$$

The constant γ is given in terms of the staggered g-tensor[4] and the DM interaction. For copper benzoate the exchange constant is $J\approx 1.57$ meV and the induced staggered field is much smaller than the applied uniform field, $h\ll H$.

As long as $h\ll J$, or equivalently as long as the field induced gap Δ is much smaller than J, it is possible to describe the low-energy degrees of freedom of (1) in terms of a massive, relativistic quantum field theory. This low-energy effective theory is obtained by Abelian bosonization and is given by a Sine-Gordon model with Hamiltonian density,[3]

$$\mathcal{H}=\frac{v}{2}[(\partial_x\Phi)^2+(\partial_x\Theta)^2]-\mu(h)\cos(\beta\Theta). \tag{3}$$

Here Φ is a canonical Bose field, Θ is the dual field and the coupling β depends on the value of the applied uniform field and has been calculated in Refs. 6–8 by using the results of Ref. 9. The spin velocity v also depends on H and is shown in Fig. 9 of Ref. 6. It is useful to define

$$\xi=\frac{\beta^2}{8\pi-\beta^2}. \tag{4}$$

The spectrum of the Sine-Gordon model (3) in the relevant range of β consists of a soliton–antisoliton doublet and several soliton–antisoliton bound states called "breathers."[3,7] The soliton gap as a function of H and h was determined in Ref. 6 in the regime $\Delta\ll H$, where

$$\frac{\Delta}{J}\simeq\left(\frac{h}{J}\right)^{(1+\xi)/2}\left[B\left(\frac{J}{H}\right)^{(2\pi-\beta^2)/4\pi}\left(2-\frac{\beta^2}{\pi}\right)^{1/4}\right]^{-(1+\xi)/2} \tag{5}$$

with $B=0.422\,169$. Equation (5) is applicable as long as H is sufficiently smaller than J or to be more precise as long as the magnetization is small. For magnetic fields comparable to J it is necessary to take into account the magnetic field dependencies of the spin velocity and the normalization $c(H)$ of the spin operator [see Eq. (8)], whereas the effects of the current–current interaction may be neglected. Using the results of Ref. 10 we obtain the following expression for the gap in the regime of H comparable to J (but still $h\ll J$):

$$\frac{\Delta}{J}\simeq\frac{2\tilde{v}(H)}{\sqrt{\pi}}\,\frac{\Gamma\left(\frac{\xi}{2}\right)}{\Gamma\left(\frac{1+\xi}{2}\right)}\left[\frac{c(H)\pi}{2\tilde{v}(H)}\,\frac{\Gamma\left(\frac{1}{1+\xi}\right)}{\Gamma\left(\frac{\xi}{1+\xi}\right)}\,\frac{h}{J}\right]^{(1+\xi)/2}. \tag{6}$$

Here $\tilde{v}=v/(Ja_0)$ is the "dimensionless spin velocity," a_0 is the lattice constant, and $c(H)$ is given below. The breather gaps are given by

$$\Delta_n=2\Delta\sin\left(\frac{\pi\xi n}{2}\right),\quad n=1,\ldots,\left[\frac{1}{\xi}\right]. \tag{7}$$

II. DYNAMICAL STRUCTURE FACTOR

The staggered/oscillating components of the spin operators are expressed in terms of the continuum fields Φ and Θ as

$$S_n^z \sim (-1)^n a(H) \sin\left(\frac{2\pi}{\beta}\Phi - \frac{2\delta}{a_0}x\right),$$

$$S_n^x \sim (-1)^n c(H) \cos(\beta\Theta),$$

$$S_n^y \sim (-1)^n c(H) \sin(\beta\Theta). \tag{8}$$

Here $x = na_0$ and the parameter β and the incommensuration δ are determined from the exact solution of the Heisenberg model in a uniform magnetic field[8,9,11] [that is the Hamiltonian (1) for $h=0$]. The amplitudes $a(H)$ and $c(H)$ are at present not known analytically, but can be determined numerically with high accuracy. We note that these amplitudes are also calculated in the absence of a staggered field, the expectation being that the changes due to a small $h \ll H$ will be negligible. The data used in this work are obtained in the scheme of Refs. 12 and 13: We calculate the spin polarization $\langle S_n^z \rangle$ and the two-spin correlation function $\langle S_n^x S_{n'}^x \rangle$ in the Heisenberg chain of 200 spins using the density-matrix renormalization group method, and then, fit them to analytic formulas obtained from the Abelian bosonization taking $a(H)$ and $c(H)$ as fitting parameters. The results as well as other parameters, which are determined exactly,[6,7] are listed in Table I for several typical values of the magnetization m.

The inelastic neutron scattering intensity is proportional to

$$I(\omega,\boldsymbol{k}) \propto \sum_{\alpha,\beta} \left(\delta_{\alpha\beta} - \frac{k_\alpha k_\beta}{\boldsymbol{k}^2}\right) S^{\alpha\beta}(\omega,\boldsymbol{k}), \tag{9}$$

where $\alpha,\beta = x,y,z$ and the dynamical structure factor $S^{\alpha\beta}$ is defined by

$$S^{\alpha\beta}(\omega,k) = \sum_{l=1}^{N} \int_{-\infty}^{\infty} \frac{dt}{2\pi} e^{-ikla_0 + i\omega t} \langle S_{l+1}^\alpha(t) S_1^\beta(0) \rangle. \tag{10}$$

Here k denotes the component of $\boldsymbol{k}$ along the chain direction. A schematic representation of which excited states will contribute to the various components of the dynamical structure factor at $k=\pi/a_0$ and $k=(\pi \pm 2\delta)/a_0$ is shown in Fig. 1. At the antiferromagnetic wave number there are several breather excitations and at higher energies multiparticle continua.[7] These contribute to the xx and yy components of the dynamical structure factor, which have been determined in detail in Ref. 7. At the incommensurate wave numbers $k=(\pi \pm 2\delta)/a_0$ there are soliton and antisoliton states and at higher energies multiparticle scattering continua. The composition of these continua can be inferred by observing that the corresponding intermediate states must have topological charge ∓ 1 at wave number $(\pi \pm 2\delta)/a_0$ in order to contribute to S^{zz}. For example, at $(\pi + 2\delta)/a_0$ there is a two-particle soliton-breather continuum above an energy $\Delta + \Delta_1$.

TABLE I. Amplitudes a and c, the spin velocity v, the coupling β, and the field H as functions of the magnetization m. The amplitudes are determined numerically except for $m=0.5$, where exact values are shown. The figures in parentheses for a and c indicate the error on the last quoted digits.

m	a	c	v	β	H
0.02	0.591(3)	0.4937(3)	1.54271	2.35016	0.17599
0.04	0.550(5)	0.4883(2)	1.51707	2.31088	0.34214
0.06	0.520(4)	0.4863(2)	1.48415	2.27738	0.50013
0.08	0.4947(6)	0.4853(2)	1.44425	2.24653	0.65001
0.10	0.475(1)	0.4847(2)	1.39796	2.21731	0.79164
0.12	0.454(1)	0.4842(2)	1.34593	2.18927	0.92489
0.14	0.437(2)	0.4835(2)	1.28879	2.16216	1.04965
0.16	0.422(2)	0.4825(2)	1.22720	2.13587	1.16589
0.18	0.4070(7)	0.4810(2)	1.16178	2.11029	1.27360
0.20	0.3938(8)	0.4790(2)	1.09314	2.08538	1.37287
0.22	0.3813(6)	0.4764(2)	1.02184	2.06107	1.46380
0.24	0.3700(8)	0.4731(2)	0.94844	2.03735	1.54656
0.26	0.3596(7)	0.4690(2)	0.87347	2.01418	1.62134
0.28	0.3499(4)	0.4639(2)	0.79741	1.99153	1.68839
0.30	0.3406(4)	0.4578(2)	0.72074	1.96940	1.74794
0.32	0.3330(2)	0.4504(2)	0.64387	1.94775	1.80030
0.34	0.3262(2)	0.4416(2)	0.56722	1.92658	1.84575
0.36	0.3200(3)	0.4310(2)	0.49116	1.90586	1.88462
0.38	0.3145(4)	0.4183(2)	0.41602	1.88559	1.91723
0.40	0.3094(2)	0.4029(1)	0.34212	1.86574	1.94390
0.42	0.3070(8)	0.3841(1)	0.26973	1.84631	1.96497
0.44	0.3058(2)	0.3601(1)	0.19912	1.82727	1.98079
0.46	0.3062(6)	0.3284(1)	0.13049	1.80863	1.99168
0.48	0.309(1)	0.2802(1)	0.06407	1.79036	1.99797
0.50	0.3183	0	0	1.77245	2

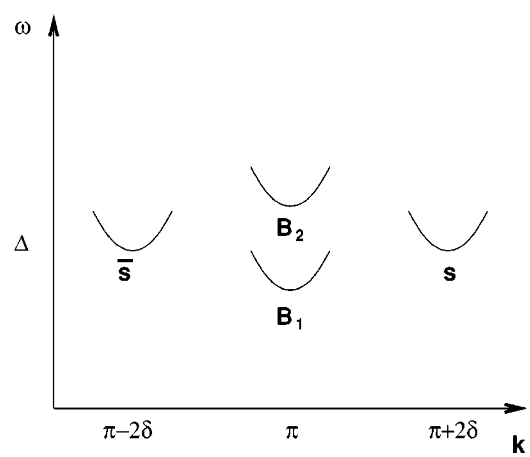

FIG. 1. Schematic structure of the lowest-energy excited states relevant to neutron scattering experiments. Soliton and antisoliton occur in the vicinity of the incommensurate wave numbers $\pi \pm 2\delta$ and are seen in S^{zz}, whereas the first breather B_1 occurs in the vicinity of π and contributes to S^{yy}. At higher energies further breather bound states as well as multiparticle scattering continua are present.

In this paper we calculate the single particle soliton/antisoliton contributions to S^{zz} and compare them to the dominant feature in the dynamical structure factor, the contribution of the lightest breather bound state B_1 to S^{yy}.

The lightest breather B_1 has a gap Δ_1 and contributes to S^{yy} as[7]

$$S^{yy}\left(\omega,\frac{\pi}{a_0}+q\right)\Big|_{B_1}=C_y(H)\,\delta(\omega^2-(vq)^2-\Delta_1^2),\quad (11)$$

where

$$C_y(H)=2\tilde{v}Jc^2(H)\left[2\cos(\pi\xi/2)\sqrt{2\sin(\pi\xi/2)}\exp\left(-\int_0^{\pi\xi}\frac{dt}{2\pi}\frac{t}{\sin t}\right)\right]^2\left(\frac{\Delta}{J\tilde{v}}\frac{\sqrt{\pi}}{2}\frac{\Gamma((1+\xi)/2)}{\Gamma(\xi/2)}\right)^{\beta^2/2\pi}$$
$$\times\exp\left[2\int_0^\infty\frac{dt}{t}\left(\frac{\sinh^2(2\beta^2 t)}{2\sinh(\beta^2 t)\sinh(8\pi t)\cosh[(8\pi-\beta^2)t]}-\frac{\beta^2}{4\pi}e^{-16\pi t}\right)\right].\quad (12)$$

Here we have used the normalizations of Ref. 14. The leading contributions to the longitudinal structure factor at the incommensurate wave numbers $k=(\pi\pm2\delta)/a_0$ are due to soliton and antisoliton. Using the results of Ref. 15 we obtain

$$S^{zz}\left(\omega,\frac{\pi\pm2\delta}{a_0}+q\right)\Big|_{s,\bar{s}}=C_z(H)\,\delta(\omega^2-(vq)^2-\Delta^2),\quad (13)$$

where

$$C_z(H)=\frac{\tilde{v}J}{2}a^2(H)\left(\frac{\mathcal{C}_1^4\xi}{4\mathcal{C}_2}\right)^{-1/4}\left(\frac{\sqrt{\pi}\Delta\Gamma\left(\frac{3}{2}+\frac{\xi}{2}\right)}{J\tilde{v}\Gamma\left(\frac{\xi}{2}\right)}\right)^{2\pi/\beta^2}$$
$$\times\exp\left[\int_0^\infty\frac{dt}{t}\left(\frac{\exp[-(1+\xi)t]-1}{2\sinh(\xi t)\sinh[(1+\xi)t]\cosh(t)}+\frac{1}{2\sinh(t\xi)}-\frac{2\pi e^{-2t}}{\beta^2}\right)\right].\quad (14)$$

Here the constants $\mathcal{C}_{1,2}$ are given by

$$\mathcal{C}_1=\exp\left(-\int_0^\infty\frac{dt}{t}\frac{\sinh^2(t/2)\ \sinh[t(\xi-1)]}{\sinh(2t)\sinh(\xi t)\cosh(t)}\right),$$
$$\mathcal{C}_2=\exp\left(4\int_0^\infty\frac{dt}{t}\frac{\sinh^2(t/2)\sinh[t(\xi-1)]}{\sinh(2t)\sinh(\xi t)}\right).\quad (15)$$

As was pointed out in Ref. 6, at $H=0$ the low-energy effective theory of (1) is SU(2) symmetric. In our notations this implies that

$$\lim_{H\to0}\frac{C_y}{c^2(H)}=2\lim_{H\to0}\frac{C_z}{a^2(H)}.\quad (16)$$

Equation (16) is easily verified numerically. In order to evaluate $C_{y,z}$ we need to know the constant of proportionality γ that relates the induced staggered field h with the applied uniform field H. This constant differs from compound to compound. On the other hand, γ enters the expressions for $C_{y,z}$ only via the soliton gap Δ. Hence it is useful to isolate the γ dependence and consider the quantities,

$$C_y'(H)=C_y(H)\left(\frac{\Delta}{J}\right)^{-(\beta^2/2\pi)}J^{-1},$$
$$C_z'(H)=C_z(H)\left(\frac{\Delta}{J}\right)^{-(2\pi/\beta^2)}J^{-1}.\quad (17)$$

The amplitudes $C_{y,z}'(H)$ are shown as functions of the magnetization in Fig. 2.

III. COPPER BENZOATE

We are now in a position to determine the ratio between the spectral weights of the first breather (seen in the transverse structure factor S^{yy}) and the soliton (seen in the longitudinal structure factor S^{zz}). In an ideal situation one would carry out measurements with momentum transfers only along

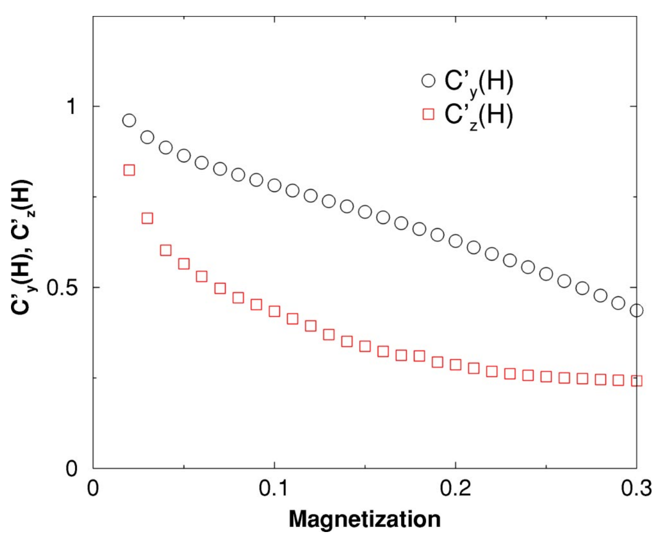

FIG. 2. Amplitudes $C_{y,z}'(H)$ as functions of the magnetization.

the y and z directions, respectively. In practice, the experiments on Copper Benzoate were carried out with momentum transfers $\boldsymbol{k}_{1,2}$ respectively, where

$$(\boldsymbol{k}_1\cdot\boldsymbol{a},\boldsymbol{k}_1\cdot\boldsymbol{b},\boldsymbol{k}_1\cdot\boldsymbol{c})=2\pi(-0.3,0,1),$$

$$(\boldsymbol{k}_2\cdot\boldsymbol{a},\boldsymbol{k}_2\cdot\boldsymbol{b},\boldsymbol{k}_2\cdot\boldsymbol{c})=2\pi(-0.3,0,1.12). \tag{18}$$

Here $\boldsymbol{c}$ points along the chain direction and the antiferromagnetic wave number corresponds to $2\pi/c$ ($c=6.30A$) as there are two copper atoms per unit cell along the c-axis. To make contact with our previous notations we need to set $a_0=c/2$. The measurements with momentum transfers $\boldsymbol{k}_1$ and $\boldsymbol{k}_2$ probe the dynamical structure factor around π/a_0 and the incommensurate wave number $(\pi+2\delta)/a_0$, respectively. In order to make direct comparisons with the experiments we need to relate the (a,b,c) coordinate system describing the crystal axes to the (x,y,z) spin coordinates. By definition z and x are the directions of the uniform and staggered fields, respectively. In the experiments of Ref. 2 the uniform field was applied along the b-direction. Based on a polarization analysis it was suggested in Ref. 6 that the staggered field lies in the ac plane and encloses an angle of $\alpha=-72°$ with the a-axis. In the vicinity of π/a_0 the dominant contribution to the structure factor comes from the transverse correlators. This implies that

$$\begin{aligned}I(\omega,\boldsymbol{k}_1)&\propto(0.083\cos^2\alpha+0.917\sin^2\alpha)S^{yy}\left(\omega,\frac{\pi}{a_0}\right)\\&\quad+(0.083\sin^2\alpha+0.917\cos^2\alpha)S^{xx}\left(\omega,\frac{\pi}{a_0}\right)\\&\approx 0.84\ S^{yy}\left(\omega,\frac{\pi}{a_0}\right)+0.16\ S^{xx}\left(\omega,\frac{\pi}{a_0}\right).\end{aligned} \tag{19}$$

On the other hand at momentum transfer $\boldsymbol{k}_2$ the dominant contribution to the structure factor is due to the longitudinal component

$$I(\omega,\boldsymbol{k}_2)\propto S^{zz}\left(\omega,\frac{\pi+2\delta}{a_0}\right). \tag{20}$$

As was pointed out in Ref. 6 there are unresolved issues concerning the polarization analysis and the estimate of the angle α should be regarded with some caution. It is possible to infer α by analyzing other experiments such as specific heat and ESR measurements. The analysis of the specific heat data suggests that $\alpha\approx-82°$,[8] which leads to a contribution of about 90% of S^{yy} in (19).

The neutron scattering experiments of Ref. 2 were performed in a uniform magnetic field of 7 T, which corresponds to a magnetization per site of $m\approx0.06$. The breather and soliton gaps were observed at

$$\Delta\approx0.22\text{ meV},\ \Delta_1\approx0.17\text{ meV}. \tag{21}$$

Using the expression (5) for the soliton gap we can infer the coefficient of proportionality between the uniform field H and the staggered field h as $\gamma\approx0.06$. Taking $\gamma=0.06$ and $m=0.06$, Eq. (5) gives $\Delta=0.215$ meV, $\Delta_1=0.171$ meV and we will use this set of parameters for our further analysis.

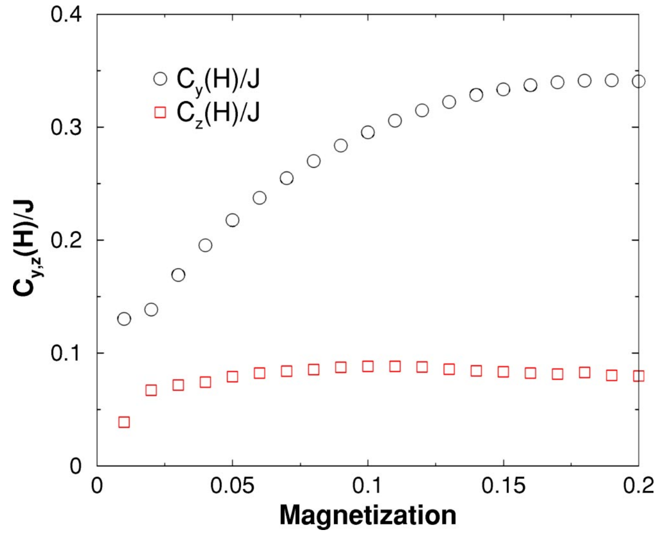

FIG. 3. Amplitudes $C_{y,z}(H)$ as functions of the magnetization.

Under the above assumptions we may now determine the spectral weights of the coherent soliton and breather peaks in the dynamical structure factor. The results are shown in Fig. 3.

At a magnetization of $m=0.06$ we have

$$\frac{C_y(H)}{C_z(H)}\approx2.88. \tag{22}$$

The spectral weights are obtained by integrating the respective structure factors over frequency at fixed momentum, i.e.,

$$I_{B_1}=\int d\omega S^{yy}\left(\omega,\frac{\pi}{a_0}\right)\bigg|_{B_1},$$

$$I_s=\int d\omega S^{zz}\left(\omega,\frac{\pi+2\delta}{a_0}\right)\bigg|_s. \tag{23}$$

The ratio of spectral weights between the first breather I_{B_1} and the soliton I_s is approximately

$$\frac{I_{B_1}}{I_s}=\frac{C_y(H)}{C_z(H)}\frac{\Delta}{\Delta_1}\approx3.64. \tag{24}$$

In order to compare to experiment, we need to take into account the different momentum transfers in the measurements of S^{yy} and S^{zz} respectively. From (19) and (20) we arrive at the following theoretical prediction for the ratio of intensities,

$$R=0.84\frac{I_{B_1}}{I_s}\approx3.06. \tag{25}$$

The experimentally observed[2] ratio of peak heights between the breather and soliton peaks is approximately 2.8. This is in reasonable agreement with our result. For a better comparison one should take into account the resolution function of the instrument in both momentum and energy, but this goes beyond the scope of our present analysis.

IV. DISCUSSION

We have determined the (anti)soliton contribution to the dynamical structure factor for field-induced gap systems such as copper benzoate within a low-energy field theory description. We find reasonable agreement with neutron scattering experiments. Our calculations are valid as long as the induced gap is small compared to the exchange J and the uniform magnetization m is not too small. The latter condition has to be imposed in order to justify neglecting the interaction of spin currents. Although this interaction is irrelevant in the renormalization group sense, its scaling dimension is close to 2 for $m \ll 1$ and its effects on, e.g., the definition of the amplitudes $c(H)$ and $a(H)$ needs to be taken into account.

ACKNOWLEDGMENTS

We thank I. Affleck and I. Zaliznyak for discussions. Work at Brookhaven National Laboratory was carried out under Contract No. DE-AC02-98 CH10886, Division of Material Science, U.S. Department of Energy. The research of A.F. was supported in part by a Grant-in-Aid for Scientific Research on Priority Areas from the Ministry of Education, Culture, Sports, Science, and Technology (Grant No. 12046238).

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