PHYSICAL REVIEW B **91**, 041108(R) (2015)

Elastic constants and ultrasound attenuation in the spin-liquid phase of Cs_2CuCl_4

S. Streib and P. Kopietz
Institut für Theoretische Physik, Universität Frankfurt, Max-von-Laue Strasse 1, 60438 Frankfurt, Germany

P. T. Cong, B. Wolf, M. Lang, N. van Well, F. Ritter, and W. Assmus
Physikalisches Institut, Universität Frankfurt, Max-von-Laue Strasse 1, 60438 Frankfurt, Germany
(Received 16 September 2014; published 9 January 2015)

The spin excitations in the spin-liquid phase of the anisotropic triangular lattice quantum antiferromagnet Cs_2CuCl_4 have been shown to propagate dominantly along the crystallographic b axis. To test this dimensional reduction scenario, we have performed ultrasound experiments in the spin-liquid phase of Cs_2CuCl_4 probing the elastic constant c_{22} and the sound attenuation along the b axis as a function of an external magnetic field along the a axis. We show that our data can be quantitatively explained within the framework of a nearest-neighbor spin-1/2 Heisenberg chain, where fermions are introduced via the Jordan-Wigner transformation and the spin-phonon interaction arises from the usual exchange-striction mechanism.

DOI: 10.1103/PhysRevB.91.041108 PACS number(s): 75.10.Kt, 43.35.+d, 72.55.+s, 75.10.Pq

Spin-liquid behavior can occur either in a spin-liquid ground state or in a spin-liquid phase at finite temperatures. One of the characteristic properties of spin liquids are strong short-range spin correlations in the absence of long-range magnetic order. Such a behavior has been observed in the magnetic insulator Cs_2CuCl_4, for example in inelastic neutron scattering experiments [1], at temperatures between 0.6 K and 2.6 K in magnetic fields below the saturation field $B_c = 8.5$ T. Experimentally, the boundary between the spin-liquid phase and the conventional paramagnetic phase has been characterized by broad peaks in the specific heat [2] and in the magnetic susceptibility [3]. Cs_2CuCl_4 can be modeled by a spin-1/2 Heisenberg antiferromagnet on a spatially anisotropic triangular lattice with nearest-neighbor exchange couplings $J = 4.34$ K along the crystallographic b axis and $J' = 1.49\ \mathrm{K} \approx J/3$ along the diagonal links within the bc plane (see Fig. 1) [4]. The interplane interaction $J'' = 0.2$ K and the Dzyaloshinskii-Moriya interaction $D = 0.23$ K can be neglected in the temperature range of the spin-liquid phase. Given the fact that the diagonal coupling J' is nonnegligible, one would naively expect an anisotropic two-dimensional spin liquid state. However, several independent calculations [5–15] found that the spin excitations in the anisotropic triangular lattice antiferromagnet are quasi-one-dimensional and propagate dominantly along the direction corresponding to the largest exchange coupling, which is the crystallographic b axis in Cs_2CuCl_4. In this work we shall give further evidence for this dimensional reduction scenario [16] by showing that ultrasound experiments probing the sound propagation along the b axis can be quantitatively explained using a one-dimensional Heisenberg chain which is coupled to lattice vibrations via the usual exchange-striction mechanism [17].

The spin-phonon interaction and the ultrasonic attenuation in two-dimensional spin liquids have recently been discussed by Zhou and Lee [18] and by Serbyn and Lee [19]. In one-dimensional Heisenberg [20] and XY chains [21] the interaction between the spin degrees of freedom and the phonons were studied a long time ago, but these older works mainly focused on the spin-Peierls transition and treated the spin-phonon interaction in an adiabatic approximation. Moreover, only the first derivative of the exchange coupling with respect to the phonon coordinates were considered, which turns out to be insufficient to explain our ultrasound experiments for the c_{22} mode in Cs_2CuCl_4.

From the exact Bethe ansatz solution of the spin-1/2 antiferromagnetic Heisenberg chain we know that the ground state is a spin liquid, exhibiting algebraic correlations but no long-range magnetic order. The elementary excitations above this ground state are spinons carrying spin 1/2. A combination of numerical and analytical methods has lead to an excellent understanding of this model [22]; for example, exact numerical results for the magnetization [23], magnetic susceptibility [24], specific heat [25], and the dynamic structure factor [26,27] are available. However, a proper microscopic calculation of ultrasound propagation and attenuation in the Heisenberg chain cannot be found in the literature. Below we shall present a simple solution of this problem using the Jordan-Wigner representation of the spin algebra in terms of spinless fermions [28]. We shall also present some new data of the elastic constant c_{22} and the corresponding ultrasound damping rate in the spin-liquid phase of Cs_2CuCl_4 which agree very well with our theory. For details concerning the experiment and the sample preparation we refer to Ref. [29] and Ref. [30], respectively. The ultrasound physics of Cs_2CuCl_4 has previously been studied for magnetic fields along the a axis in the ordered phase using spin-wave theory [29], and for magnetic fields along the b axis in the spin-liquid phase by combining phenomenological expressions for the ultrasound propagation and attenuation with calculations for two-dimensional spin models [31].

Assuming that the relevant spin excitations can propagate only along the crystallographic b axis in the spin-liquid phase of Cs_2CuCl_4, we expect that ultrasound experiments probing the c_{22} mode along the b axis can be explained by the following one-dimensional spin-phonon Hamiltonian,

$$\mathcal{H} = \sum_n J_n [\boldsymbol{S}_n \cdot \boldsymbol{S}_{n+1} - 1/4] - h \sum_n S_n^z + \mathcal{H}_2^p, \qquad (1)$$

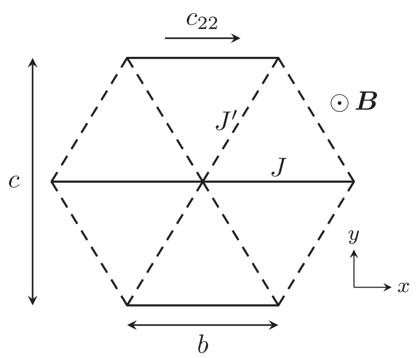

FIG. 1. Part of the anisotropic triangular lattice formed by the spins in Cs_2CuCl_4. The largest exchange coupling J connects nearest-neighbor spins along the crystallographic b axis. The corresponding elastic constant is denoted by c_{22}. In this work we consider only the case where the magnetic field $\boldsymbol{B}$ is along the a axis perpendicular to the plane of the lattice.

where $\boldsymbol{S}_n$ are spin-1/2 operators localized at the positions x_n on a chain with N spins and periodic boundary conditions, and $h = g\mu_B B$ (with $g \approx 2.2$) [4] is the Zeeman energy associated with an external magnetic field $\boldsymbol{B}$ along the crystallographic a axis; see Fig. 1. The spin-phonon coupling arises from the fact that for a vibrating lattice the spins are located at $x_n = nb + X_n$, where nb (with $n = 1,\dots,N$) are the points of a one-dimensional lattice with lattice spacing b, and X_n denote the deviations from the lattice points. Since the exchange coupling J_n between a pair of spins $\boldsymbol{S}_n$ and $\boldsymbol{S}_{n+1}$ located at x_n and x_{n+1} depends on the actual distance between the spins, J_n is a function of $X_{n+1} - X_n$. Assuming that this difference is small, we may expand to second order,

$$J_n \approx J + J^{(1)}(X_{n+1} - X_n) + \frac{J^{(2)}}{2}(X_{n+1} - X_n)^2, \quad (2)$$

where $J^{(1)}$ and $J^{(2)}$ are the first and second derivative of the exchange coupling along the b axis with respect to the phonon coordinates. As usual, we quantize the lattice vibrations by introducing the conjugate momenta P_n and demanding that $[X_n, P_m] = i\delta_{n,m}$, where we have set $\hbar = 1$. The last term in Eq. (1) describes noninteracting phonons with dispersion $\omega_q = c|q|$ [32],

$$\mathcal{H}_2^p = \sum_q \left[\frac{P_{-q}P_q}{2M} + \frac{M}{2}\omega_q^2 X_{-q}X_q \right], \quad (3)$$

where M is the mass attached to the vibrating sites, and the operators X_q and P_q are defined via the Fourier expansions $X_n = N^{-1/2}\sum_q e^{iqnb}X_q$ and $P_n = N^{-1/2}\sum_q e^{iqnb}P_q$.

To explain ultrasound experiments, we calculate the self-energy correction $\Pi(q,i\omega)$ to the phonon propagator, which arises from the coupling of the phonons to the spins. Let us therefore represent the spin operators in terms of spinless fermions using the Jordan-Wigner transformation [28]

$$S_n^+ = (S_n^-)^\dagger = c_n^\dagger(-1)^n e^{i\pi\sum_{j<n} c_j^\dagger c_j}, \quad S_n^z = c_n^\dagger c_n - 1/2, \quad (4)$$

where c_n annihilates a fermion at site x_n and the phase factor $(-1)^n$ is introduced for convenience. Our spin-phonon Hamiltonian (1) then assumes the form

$$\mathcal{H} = -\frac{1}{2}\sum_n J_n(c_n^\dagger c_{n+1} + c_{n+1}^\dagger c_n + c_n^\dagger c_n + c_{n+1}^\dagger c_{n+1}) + \sum_n J_n c_n^\dagger c_n c_{n+1}^\dagger c_{n+1} - h\sum_n c_n^\dagger c_n + Nh/2 + \mathcal{H}_2^p. \quad (5)$$

In this work, we shall treat the two-body interaction in the second line of Eq. (5) within the self-consistent Hartree-Fock approximation, which amounts to approximating the two-body term by

$$c_n^\dagger c_n c_{n+1}^\dagger c_{n+1} \approx \rho(c_{n+1}^\dagger c_{n+1} + c_n^\dagger c_n) - \rho^2 - \tau(c_n^\dagger c_{n+1} + c_{n+1}^\dagger c_n) + \tau^2, \quad (6)$$

where the dimensionless variational parameters ρ and τ satisfy the self-consistency conditions

$$\rho = \langle c_n^\dagger c_n\rangle, \quad \tau = \langle c_n^\dagger c_{n+1}\rangle. \quad (7)$$

In the absence of phonons, the solution of these equations was worked out a long time ago by Bulaevskii [33]. Within the Hartree-Fock approximation the fermion dispersion is $\xi_k = -ZJ\cos k + 2sJ - h$, where $Z = 1 + 2\tau$ is the dimensionless renormalization factor of the nearest-neighbor hopping, $s = \rho - 1/2$ is the dimensionless magnetization, and k is the fermion lattice momentum in units of the inverse lattice spacing $1/b$. In Fig. 2 we show the numerical result for $Z(h)$ at $T = 0$ and we compare our mean-field result for $s(h)$ with the exact magnetization curve of the Heisenberg chain obtained via the Bethe ansatz [23].

To obtain the change of the elastic constant and the sound attenuation, we should calculate the self-energy of the phonons due to the coupling to the spins. Substituting the gradient expansion (2) for the exchange coupling and the Hartree-Fock decoupling (6) into Eq. (5), we arrive at the approximate spin-phonon Hamiltonian

$$\mathcal{H} = F_0 + \sum_k \xi_k c_k^\dagger c_k + \mathcal{H}_2^p + \delta\mathcal{H}_2^p + \mathcal{H}_3^{sp} + \mathcal{H}_4^{sp}, \quad (8)$$

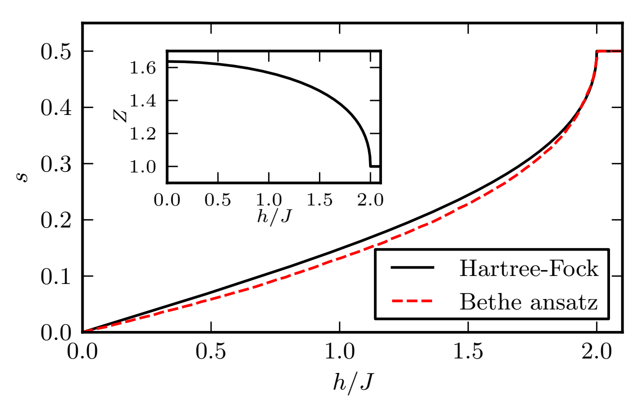

FIG. 2. (Color online) Comparison of the Hartree-Fock result for the magnetization curve $s(h)$ of the Heisenberg chain (without phonons) at $T = 0$ with the exact Bethe ansatz [23]. Inset: Renormalization $Z(h)$ of the nearest-neighbor hopping.

where $F_0/N = h/2 + J(\tau^2 - \rho^2)$ and

$$\delta\mathcal{H}_2^p = 2J^{(2)}(\tau^2 - \rho^2)\sum_q \sin^2(q/2)X_{-q}X_q, \tag{9a}$$

$$\mathcal{H}_3^{sp} = \frac{1}{\sqrt{N}}\sum_{k'kq}\delta^*_{k',k+q}\Gamma_3(k,q)c^\dagger_{k'}c_kX_q, \tag{9b}$$

$$\mathcal{H}_4^{sp} = \frac{1}{2N}\sum_{k'kq_1q_2}\delta^*_{k',k+q_1+q_2}\Gamma_4(k,q_1,q_2)c^\dagger_{k'}c_kX_{q_1}X_{q_2}. \tag{9c}$$

Here $\delta^*_{k',k} = \sum_m \delta_{k',k+2\pi m/b}$ enforces momentum conservation modulo a vector of the reciprocal lattice, $c_k = N^{-1/2}\sum_n e^{-ikbn}c_n$, and the cubic and quartic interaction vertices are for small phonon momenta given by

$$\Gamma_3(k,q) \approx -iqJ^{(1)}[Z\cos k - 2s], \tag{10a}$$

$$\Gamma_4(k,q_1,q_2) \approx q_1q_2J^{(2)}[Z\cos k - 2s]. \tag{10b}$$

The coupling to the fermions gives rise to a momentum- and frequency-dependent self-energy correction $\Pi(q,i\omega)$ to the propagator of the phonon field X_q, which is proportional to $[\omega^2 + \omega_q^2 + \Pi(q,i\omega)]^{-1}$. To second order in the derivatives of the exchange coupling the phonon self-energy has three contributions, $\Pi(q,i\omega) = \Pi_2(q) + \Pi_3(q,i\omega) + \Pi_4(q)$, where

$$\Pi_2(q) = [J^{(2)}/M][\tau^2 - \rho^2]4\sin^2(q/2), \tag{11a}$$

$$\Pi_3(q,i\omega) = \frac{1}{MN}\sum_k \frac{f_k - f_{k+q}}{\xi_k - \xi_{k+q} + i\omega}|\Gamma_3(k,q)|^2, \tag{11b}$$

$$\Pi_4(q) = \frac{1}{MN}\sum_k f_k\Gamma_4(k,q,-q), \tag{11c}$$

and $f_k = [e^{\beta\xi_k} + 1]^{-1}$ is the occupation of the fermion state with momentum k in self-consistent Hartree-Fock approximation. From the analytic continuation of the self-energy $\Pi(q,i\omega)$ to real frequencies we obtain the renormalized phonon energy and the phonon damping [29],

$$\tilde{\omega}_q = \omega_q + \frac{\mathrm{Re}\Pi(q,\omega_q + i0)}{2\omega_q}, \quad \gamma_q = -\frac{\mathrm{Im}\Pi(q,\omega_q + i0)}{2\omega_q}. \tag{12}$$

The renormalized phonon velocity can be obtained from $\tilde{c}/c = \lim_{q\to 0}\tilde{\omega}_q/\omega_q$, which yields for the shift $\Delta c = \tilde{c} - c$,

$$\Delta c/c = g_1c^{(1)} + g_2c^{(2)}, \tag{13a}$$

$$c^{(1)} = ⨍_{-\pi}^{\pi}\frac{dk}{2\pi}Jf'(\xi_k)\frac{v_k}{v_k - c}[2s - Z\cos k]^2, \tag{13b}$$

$$c^{(2)} = s^2 - Z^2/4, \tag{13c}$$

where $⨍$ denotes the Cauchy principal value, $f'(\xi_k) = -\beta f_k[1 - f_k]$ is the derivative of the Fermi function, $v_k = ZJb\sin k$ is the group velocity of the fermionic excitations, and we have introduced the dimensionless coupling constants $g_1 = [J^{(1)}b]^2/(2Mc^2J)$ and $g_2 = J^{(2)}b^2/(2Mc^2)$. In principle it should be possible to calculate these coupling using *ab initio* methods, but here we simply determine g_1 and g_2 by fitting our theoretical prediction (13) to our experimental data. In Fig. 3 we show a comparison between theory and experiment as a function of the magnetic field. We find that the relative change of the sound velocity $\Delta c/c$ is best fitted by $g_1 \approx 0$ and $g_2 \approx -1.1 \times 10^{-3}$, which gives $J^{(2)}b^2 \approx -238J$. Only for our experimental data at the highest temperature $T = 1.15$ K, we get a finite value $g_1 = 0.85 \times 10^{-3}$, which gives $J^{(1)}b \approx \pm 14J$. Because of the large value of $c/(Jb) \approx 6.8$, the term $c^{(1)}$ is more than one order of magnitude smaller than $c^{(2)}$ for Cs_2CuCl_4. While in the ordered phase ($T = 50$ mK, upper panel) our theoretical prediction (13) agrees only for magnetic fields up to 5 T with our experimental data, in the spin-liquid phase ($T = 1$ K, lower panel) we obtain excellent agreement between theory and experiment for magnetic fields up to 7 T. A natural explanation for the deviations at larger fields is that in this regime the fluctuations are controlled by the dilute Bose gas quantum critical point at $B_c = 8.5$ T [2], which of course cannot be described by our one-dimensional model.

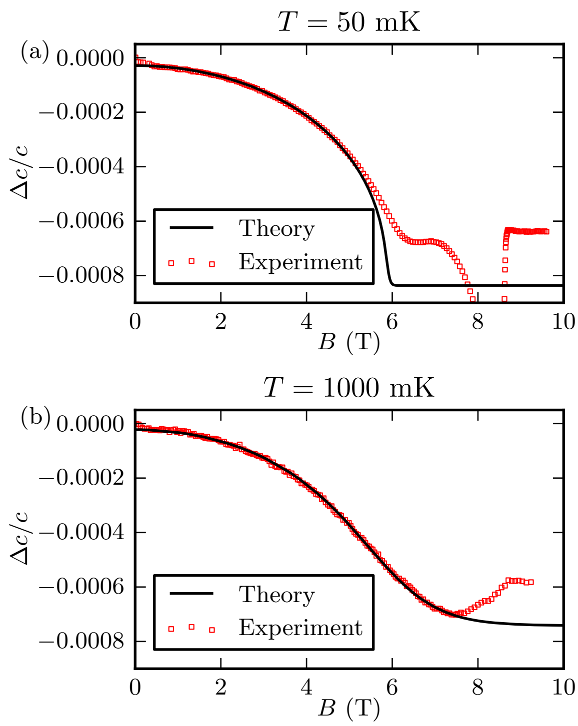

FIG. 3. (Color online) Comparison of theory and experiment for the relative change of the sound velocity of the c_{22} mode: (a) in the ordered phase ($T = 50$ mK) with coupling constants $g_1 = 0$ and $g_2 = -1.2 \times 10^{-3}$, (b) in the spin-liquid phase ($T = 1$ K) with coupling constants $g_1 = 0$ and $g_2 = -1.1 \times 10^{-3}$. In the fitting procedure we have allowed a constant offset for $\Delta c/c$ which is necessary due to a small anomaly in the experimental data close to zero magnetic field.

Finally, let us discuss the ultrasound attenuation of the c_{22} mode in Cs_2CuCl_4. Our experimental data for three different temperatures as a function of the magnetic field are shown in Fig. 4. In the regime $B \lesssim 7$ T where the fluctuations controlled by the quantum critical point are negligible and our theoretical prediction for the renormalization of the phonon velocity agrees with experiment, the sound attenuation is very small and practically constant. This can easily be explained within our one-dimensional model. Using Eqs. (11b) and (12) we

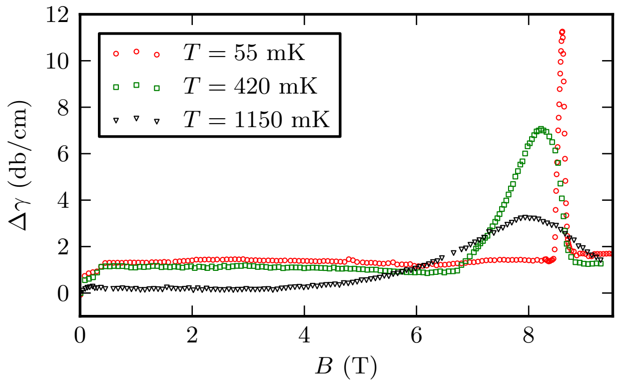

FIG. 4. (Color online) Experimental results for the relative change $\Delta\gamma$ of the sound attenuation of the c_{22} mode as a function of the magnetic field for three different temperatures.

obtain for the damping

$$\gamma_q = \frac{\pi}{2M\omega_q}\int_{-\pi}^{\pi}\frac{dk}{2\pi}(f_k - f_{k+q})|\Gamma_3(k,q)|^2\delta(\xi_k - \xi_{k+q} + \omega_q). \tag{14}$$

This expression is only finite if the absolute value of the maximal group velocity $v_* = ZJb$ of the fermions exceeds the phonon velocity c. Because in Cs_2CuCl_4 this condition is never satisfied ($c/(Jb) \approx 6.8$), the attenuation of the c_{22} mode vanishes in our approximation. Higher orders in perturbation theory will give a finite result, but it will involve more than two derivatives of the exchange coupling which are expected to be small.

On the other hand, the condition $v_* > c$ can possibly be realized in some other quasi-one dimensional quantum antiferromagnet. Let us therefore evaluate Eq. (14) in the regime $v_* > c$. In the long-wavelength limit $q \to 0$ we obtain

$$\frac{\gamma_q}{\omega_q} \sim \frac{g_1 c J \Theta(v_*^2 - c^2)}{2v_*\sqrt{1 - c^2/v_*^2}}[-f'(\xi_+)V_+^2 - f'(\xi_-)V_-^2], \tag{15}$$

where $\xi_\pm = JV_\pm - h$ and $V_\pm = 2s \pm Z\sqrt{1 - c^2/v_*^2}$. A numerical evaluation of this expression is shown in the upper panel of Fig. 5.

The damping exhibits strong peaks as function of the magnetic field, corresponding to the resonance conditions $\xi_\pm = 0$ imposed by the broadened delta functions $-f'(\xi_\pm)$ in Eq. (14). In the lower panel of Fig. 5 we show that close to the resonance the corresponding shift $c^{(1)}$ in the phonon

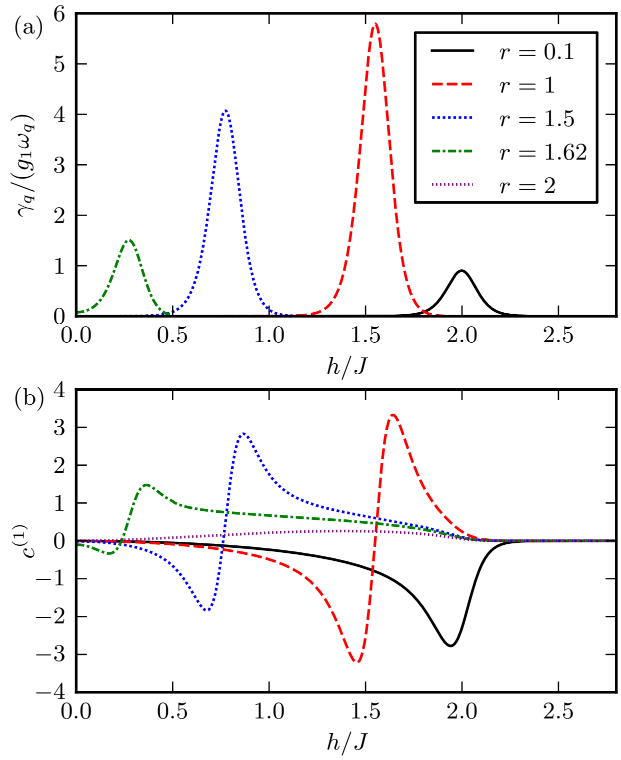

FIG. 5. (Color online) (a) Damping γ_q for different values of $r = c/(Jb)$ as a function of the magnetic field at temperature $T/J = 0.05$. (b) Corresponding contribution $c^{(1)}$ to $\Delta c/c$ defined in Eq. (13).

velocities can exhibit a sign change depending on the value of $r = c/(Jb)$.

In summary, we have developed a simple microscopic theory which explains ultrasound experiments probing the propagation and the attenuation of the c_{22} mode in the spin-liquid phase of Cs_2CuCl_4. Our basic assumption is that in the spin-liquid phase the elementary excitations are one-dimensional fermions. The excellent agreement between theory and experiments shown in Fig. 3 gives further support to the dimensional reduction scenario advanced by Balents [16]. It would be interesting to test our theoretical predictions for the ultrasound attenuation shown in Fig. 5 using suitable antiferromagnetic spin chains with sufficiently small phonon velocities.

We thank Andreas Kreisel for detailed comments on the manuscript. Financial support by the DFG via SFB/TRR49 is gratefully acknowledged.

[1] R. Coldea, D. A. Tennant, and Z. Tylczynski, Phys. Rev. B **68**, 134424 (2003).

[2] T. Radu, H. Wilhelm, V. Yushankhai, D. Kovrizhin, R. Coldea, Z. Tylczynski, T. Lühmann, and F. Steglich, Phys. Rev. Lett. **95**, 127202 (2005).

[3] Y. Tokiwa, T. Radu, R. Coldea, H. Wilhelm, Z. Tylczynski, and F. Steglich, Phys. Rev. B **73**, 134414 (2006).

[4] R. Coldea, D. A. Tennant, K. Habicht, P. Smeibidl, C. Wolters, and Z. Tylczynski, Phys. Rev. Lett. **88**, 137203 (2002).

[5] M. Q. Weng, D. N. Sheng, Z. Y. Weng, and R. J. Bursill, Phys. Rev. B **74**, 012407 (2006).

[6] S. Yunoki and S. Sorella, Phys. Rev. B **74**, 014408 (2006).

[7] Y. Hayashi and M. Ogata, J. Phys. Soc. Jpn. **76**, 053705 (2007).

[8] O. A. Starykh and L. Balents, Phys. Rev. Lett. **98**, 077205 (2007).

[9] M. Kohno, O. A. Starykh, and L. Balents, Nat. Phys. **3**, 790 (2007).
[10] M. Kohno, Phys. Rev. Lett. **103**, 197203 (2009).
[11] D. Heidarian, S. Sorella, and F. Becca, Phys. Rev. B **80**, 012404 (2009).
[12] T. Tay and O. I. Motrunich, Phys. Rev. B **81**, 165116 (2010).
[13] O. A. Starykh, H. Katsura, and L. Balents, Phys. Rev. B **82**, 014421 (2010).
[14] T. Herfurth, S. Streib, and P. Kopietz, Phys. Rev. B **88**, 174404 (2013).
[15] L. F. Tocchio, C. Gros, R. Valentí, and F. Becca, Phys. Rev. B **89**, 235107 (2014).
[16] L. Balents, Nature (London) **464**, 199 (2010).
[17] See, for example, B. Lüthi, *Physical Acoustics in the Solid State* (Springer, Berlin, 2005).
[18] Y. Zhou and P. A. Lee, Phys. Rev. Lett. **106**, 056402 (2011).
[19] M. Serbyn and P. A. Lee, Phys. Rev. B **87**, 174424 (2013).
[20] E. Pytte, Phys. Rev. B **10**, 4637 (1974).
[21] R. A. T. de Lima and C. Tsallis, Phys. Rev. B **27**, 6896 (1983).
[22] See, for example, T. Giamarchi, *Quantum Physics in One Dimension* (Oxford University Press, Oxford, 2003).
[23] R. B. Griffiths, Phys. Rev. **133**, A768 (1964).
[24] S. Eggert, I. Affleck, and M. Takahashi, Phys. Rev. Lett. **73**, 332 (1994).
[25] A. Klümper, Eur. Phys. J. B **5**, 677 (1998).
[26] M. Mourigal, M. Enderle, A. Klöpperpieper, J.-S. Caux, A. Stunault, and H. M. Rønnow, Nat. Phys. **9**, 435 (2013).
[27] B. Lake, D. A. Tennant, J.-S. Caux, T. Barthel, U. Schollwöck, S. E. Nagler, and C. D. Frost, Phys. Rev. Lett. **111**, 137205 (2013).
[28] E. Lieb, T. Schultz, and D. Mattis, Ann. Phys. **16**, 407 (1961).
[29] A. Kreisel, P. Kopietz, P. T. Cong, B. Wolf, and M. Lang, Phys. Rev. B **84**, 024414 (2011).
[30] N. Krüger, S. Belz, F. Schossau, A. A. Haghighirad, P. T. Cong, B. Wolf, S. Gottlieb-Schoenmeyer, F. Ritter, and W. Assmus, Cryst. Growth Des. **10**, 4456 (2010).
[31] A. Sytcheva, O. Chiatti, J. Wosnitza, S. Zherlitsyn, A. A. Zvyagin, R. Coldea, and Z. Tylczynski, Phys. Rev. B **80**, 224414 (2009).
[32] The phonon velocity c in x direction should not be confused with the lattice parameter c in Fig. 1. From here on, c denotes the phonon velocity.
[33] L. N. Bulaevskii, Zh. Eksp. Teor. Fiz. **43**, 968 (1962) [Sov. Phys. JETP **16**, 685 (1963)].