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Analytic representation of a zero-frequency transport coefficient. General theory and application to ultrasonic attenuation in $CsNiCl_3$

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Starting from Mori's continued-fraction representation of spectral functions, we derive an analytical formula for a transport coefficient in the static limit. The formula then is applied to the phonon dynamics of a model Hamiltonian for a compressible one-dimensional magnetic system. The resulting expression for the ultrasonic attenuation coefficient is compared with an experiment for $CsNiCl_3$. Good qualitative agreement is obtained.

I. INTRODUCTION

The interaction of spin and phonon degrees of freedom in magnetic systems is a topic of much interest, both theoretical and experimental.[1] Such an interaction can be due to the dependence of the exchange on the magnetic ion-ion distance. Most studies of the subject have been devoted to three-dimensional (3D) systems.

In this paper we will investigate the effect of spin-phonon interaction on acoustic properties. More specifically, we will investigate the ultrasonic attenuation, which is interesting because it is directly accessible to experiment. As in Refs. 2–6, we consider one-dimensional (1D) magnets with isotropic exchange. This is not irrelevant since Almond and Rayne[7] obtained ultrasonic attenuation data in a large temperature range at a number of frequencies for $CsNiCl_3$, which is a 1D Heisenberg chain.[8] An anomaly at the onset of 3D magnetic ordering indicated that the attenuation indeed is of magnetic origin.

Other authors have studied theoretically the influence of magnetic interactions on ultrasonic attenuation. Tani and Mori[9] studied the effect in 3D substances near the magnetic critical point. Bennett and Pytte[10] and Laramore and Kadanoff[11] also studied 3D systems above and near the critical point. Leung and Huber[12] studied the 1D planar magnet $CsNiF_3$ at low temperature. Only Nagano and Okamoto[13] considered 1D isotropic magnetic interactions, but they found an incorrect frequency dependence for the attenuation.

We will study the attenuation starting from a simple model Hamiltonian for a compressible Heisenberg chain. We use Mori's continued-fraction expansion[14] for the dynamic-displacement correlation function, which describes the phonon dynamics. The sound damping is then given by the transport coefficient, for which we derive a general analytic expression in the static limit. Next a reasonable approximation is made and we compare the result with experiment.

II. GENERAL THEORY

A. The spectral function

The dynamic behavior of a variable A can be described in frequency space by the spectral function

$$\psi_{AA}(\omega)=\int_{-\infty}^{+\infty} e^{i\omega t}\langle A^*(t)A\rangle dt \; . \tag{2.1}$$

This is equivalent to the imaginary part of the Laplace transformed correlation function as follows:

$$\psi_{AA}(\omega)=-2\lim_{\epsilon\to 0+}\mathrm{Im}\phi_{AA}(\omega+i\epsilon) \; , \tag{2.2a}$$

$$\phi_{AA}(z)=\phi_{AA}(\omega+i\epsilon) = -i\int_0^{\infty} e^{izt}\langle A^*(t)A\rangle dt \; . \tag{2.2b}$$

According to Mori's theory, $\phi_{AA}(z)$ can be represented rigorously by a continued fraction[14]

$$\phi_{AA}(z)=\cfrac{\langle |A|^2\rangle}{z-\cfrac{\Delta_1^2}{z+\Sigma(z)}} \; , \tag{2.3}$$

where the transport coefficient $\Sigma(z)$ is given by the infinite continued fraction

$$\Sigma(z)=\cfrac{-\Delta_2^2}{z-\cfrac{\Delta_3^2}{z-\cfrac{\Delta_4^2}{\ddots}}} \; . \tag{2.4}$$

The coefficients Δ_n^2 are static quantities and they can be expressed entirely in terms of the frequency moments of $\psi_{AA}(\omega)$.[15] For example, the first three read

$$\Delta_1^2=\langle\omega^2\rangle \ , \qquad (2.5a)$$

$$\Delta_2^2=\frac{\langle\omega^4\rangle}{\langle\omega^2\rangle}-\langle\omega^2\rangle \ , \qquad (2.5b)$$

$$\Delta_3^2=\left[\frac{\langle\omega^6\rangle}{\langle\omega^2\rangle}-\frac{\langle\omega^4\rangle^2}{\langle\omega^2\rangle^2}\right]\Big/\Delta_2^2 \ , \qquad (2.5c)$$

$$\langle\omega^{2n}\rangle=\left\langle\left|\frac{d^nA}{dt^n}\right|^2\right\rangle\Big/\langle |A|^2\rangle \ . \qquad (2.5d)$$

Combining Eqs. (2.2)–(2.4), we find

$$\psi_{AA}(\omega)=2\langle |A|^2\rangle\frac{\langle\omega^2\rangle\Sigma''(\omega)}{[\omega^2-\langle\omega^2\rangle+\omega\Sigma'(\omega)]^2+[\omega\Sigma''(\omega)]^2} \ , \qquad (2.6)$$

where a single and a double prime, respectively, denote the real and imaginary part.

B. The transport coefficient

The problem now is to calculate $\Sigma(z)$. Let us write $\Sigma(z)$ as

$$\Sigma(z)=\cfrac{-\Delta_2^2}{z-\cfrac{\Delta_3^2}{z-\ddots\cfrac{\Delta_n^2}{z+\Sigma_n(z)}}} \ . \qquad (2.7)$$

Assuming that $\Delta_{n+m}^2=\Delta_n^2 (m\geq 0)$, we find

$$\Sigma_n(z)=\cfrac{-\Delta_n^2}{z-\cfrac{\Delta_n^2}{z-\cfrac{\Delta_n^2}{\ddots}}}=\frac{-z+(z^2-4\Delta_n^2)^{1/2}}{2} \ , \qquad (2.8)$$

whence

$$\Sigma_n(0)=i\Delta_n \ . \qquad (2.9)$$

Letting $n\to\infty$, (2.7) then yields

$$\Sigma(0)=\lim_{n\to\infty} i\frac{\Delta_2^2}{\Delta_3^2}\frac{\Delta_4^2}{\Delta_5^2}\cdots\Delta_n^{(-1)^n} \ . \qquad (2.10)$$

This is only convergent if

$$\lim_{n\to\infty}\frac{\Delta_{n+1}^2}{\Delta_n^2}=1 \ , \qquad (2.11)$$

which is likely to be generally true.[16] Then (2.10) is an exact analytic formula. Although (2.7) has only real coefficients, we correctly obtained a purely imaginary value at zero frequency. Therefore, no phenomenological damping has to be introduced. The same result (2.10) can also be obtained using the same assumption (2.11), but starting from Mori's long-time approximation[14] for the continued fraction.

In obtaining (2.10), we changed the order of limits $n\to\infty$ and $z=i\epsilon\to 0+$. Let us show by means of an example that (2.10) nevertheless represents the correct result. Suppose Σ is Gaussian:

$$\Sigma(t)=\Delta_2^2\exp(-\Delta_3^2t^2/2) \ , \qquad (2.12a)$$

$$\Sigma(z=0)=i\Delta_2^2\int_0^\infty\exp(-\Delta_3^2t^2/2)dt$$
$$=i\left[\frac{\pi}{2}\right]^{1/2}\frac{\Delta_2^2}{\Delta_3} \ . \qquad (2.12b)$$

With the use of the well-known expansion coefficients of the continued fraction of the Gaussian,[15] one has

$$\Delta_{2+n}^2=n\Delta_3^2, \quad n>0 \qquad (2.13)$$

and (2.10) yields

$$\Sigma(0)=i\frac{\Delta_2^2}{\Delta_3}\lim_{n\to\infty}\frac{(2^nn!)^2}{(2n+1)!}(2n+2)^{1/2}$$
$$=i\left[\frac{\pi}{2}\right]^{1/2}\frac{\Delta_2^2}{\Delta_3} \ , \qquad (2.14)$$

in agreement with (2.12b). Of course, the limit (2.10) can only be calculated in very few cases. In order to be useful, an approximation is necessary.

III. THE ULTRASONIC ATTENUATION IN THE COMPRESSIBLE HEISENBERG CHAIN

As in Refs. 2–6, let us take the following Hamiltonian as a starting point for the calculations

$$H=H_P+H_S+H_{SP} \ , \qquad (3.1a)$$

$$H_P=\sum_{i=1}^{N}\frac{p_i^2}{2m}+\frac{\alpha}{2}\sum_{i=1}^{N-1}(x_{i+1}-x_i)^2 \ , \qquad (3.1b)$$

$$H_S=-J\sum_{i=1}^{N-1}\vec{S}_i\cdot\vec{S}_{i+1}\ , \tag{3.1c}$$

$$H_{SP}=-\epsilon\sum_{i=1}^{N-1}(x_{i+1}-x_i)\vec{S}_i\cdot\vec{S}_{i+1}\ . \tag{3.1d}$$

The spin-phonon interaction H_{SP} has been restricted to terms linear in the ionic displacements. In the model the ultrasonic attenuation is merely due to the spin-phonon interaction, which is also true in the experiments of Ref. 7, at least at temperatures below 70 K. In spite of the quantum-mechanical character of $CsNiCl_3$ ($S=1$), we use classical mechanics for practical reasons.

For the phonon dynamics the relevant dynamic variable A in Sec. II is the Fourier transformed displacement x_q. Then Ref. 2 gives

$$\Delta_1^2=\frac{6\Omega^2(1-\cos q)}{3+\gamma u(1-3y_1^2+2y_2)}\ , \tag{3.2a}$$

$$\Delta_2^2=\Delta_1^2\gamma u(1-3y_1^2+2y_2)/3\ , \tag{3.2b}$$

$$\Delta_3^2=32\Omega^2\delta^2\left[\gamma+\frac{3}{u(1-3y_1^2+2y_2)}\right]\times\{\tfrac{1}{3}(1-\cos q)y_1(1-y_2)+\gamma[\tfrac{2}{9}(1-y_2)(\tfrac{1}{2}+y_2)-\tfrac{1}{5}y_1(y_1-y_3)\cos q]\}\ , \tag{3.2c}$$

where

$$u=\beta J,\ \ \beta=1/k_BT \tag{3.3a}$$

$$\gamma=\epsilon^2/\alpha J\ , \tag{3.3b}$$

$$\delta=J/2\sqrt{2}\Omega\ , \tag{3.3c}$$

$$\Omega=\sqrt{\alpha/m}\ , \tag{3.3d}$$

$$y_n=\lambda_n/\lambda_0\ , \tag{3.3e}$$

$$\lambda_m=\int_{-1}^{1}P_m(x)\exp\left[ux\left[1+\frac{\gamma}{2}x\right]\right]dx\ . \tag{3.3f}$$

In the limit of very long wavelengths ($q\ll 1$)

$$\Delta_2^2<\omega_q^2\ll\Delta_3^2 \tag{3.4}$$

(γ not too large), where ω_q is the frequency of a phonon of wave vector q. By using this together with the fact that the coefficients Δ_n tend to increase if n increases,[15,17] it seems justified to use the static limit (2.10) of (2.7) in (2.6) for the investigation of the ultrasound.

Because we only know the expressions of Δ_1^2, Δ_2^2, and Δ_3^2, we write (2.10) as

$$\Sigma(0)=Ci\Delta_2^2/\Delta_3\ , \tag{3.5a}$$

$$C=\lim_{n\to\infty}\frac{1}{\Delta_3}\frac{\Delta_4^2}{\Delta_5^2}\frac{\Delta_6^2}{\Delta_7^2}\cdots\Delta_n^{(-1)^n}\ . \tag{3.5b}$$

It is impossible to calculate C, but let us assume that it depends on T much less than Δ_2^2/Δ_3. This assumption is justified by the exact results

$$\lim_{T\to 0}\Delta_2^2=0\ , \tag{3.6a}$$

$$\lim_{T\to 0}\Delta_n^2\neq 0,\ \ n>2 \tag{3.6b}$$

and the condition (2.11). Furthermore, calculations for the rigid Heisenberg chain revealed a moderate temperature dependence for Δ_3^2 and Δ_4^2.[17] By (3.4) and (3.5), and under the condition that C is not too large,

$$\omega_q=(\langle\omega^2\rangle_q)^{1/2}\gg\Sigma_q''(0)\ . \tag{3.7}$$

Therefore, to a very good approximation (2.6) represents a pair of Lorentzian lines centered at $\pm\omega_q$ with a halfwidth at half maximum given by $\Sigma_q''/2$. Then the time correlation function behaves as

$$\exp(i\omega_qt)\exp(-t\Sigma_q''/2)\ . \tag{3.8}$$

The damping or ultrasonic attenuation in dB per unit length is therefore

$$D=4.34\Sigma_q''/c\sim\frac{\Delta_2^2}{\Delta_3c}\ , \tag{3.9}$$

where c is the speed of sound. From (3.2) one then obtains

$$D(\omega_q)\sim\omega_q^2\frac{[u(1-3y_1^2+2y_2)]^{3/2}}{[(1-y_2)(1+2y_2)/9-y_1(y_1-y_3)/5]^{1/2}}\ . \tag{3.10}$$

IV. RESULTS AND DISCUSSION

Equation (3.10) shows that the ultrasonic attenuation depends quadratically on frequency. This is in agreement with the experimental results of Almond and Rayne for the attenuation of longitudinal waves in $CsNiCl_3$.[7] Nagano and Okamoto erroneously obtained an $\omega^{3/2}$ dependence.[13] In order to find ω^2

they had to introduce 3D anisotropic interactions and a cutoff, and in our opinion this is somewhat artificial. In Fig. 1 we compare the results of Ref. 7 with the theoretical prediction (3.10) at the frequency $\nu=\omega/2\pi=150$ MHz. We chose $J=-27$ K (Ref. 8) and we assumed that the coupling strength γ is very small. Then, to a very good approximation, we can set $\gamma=0$ in (3.10), because the main γ dependence is absorbed in the proportionality factor. Since we are not able to determine the absolute height of the attenuation, we fitted the height so that the experiment and theory coincide at 50 K. Remark the broad maximum near 30 K, which is related to the 1D chainlike properties. The qualitative agreement is generally good. At temperatures below 25 K the disagreement can be ascribed to quantum-mechanical effects that are necessarily present in spin-one magnets like $CsNiCl_3$. For temperatures above 25 K the agreement is remarkably good.

We have used a model with one unit per cell. Although $CsNiCl_3$ has a more complicated structure, in the long-wavelength limit the simplified model is appropriate.

In summary, in contrast to earlier theory,[13] our theory gives both the correct frequency dependence and the correct qualitative temperature dependence in agreement with experiment. It would be interesting to have experimental results for other 1D magnets, for example $(CH_3)_4NMnCl_3$ [tetramethyl ammonium manganese chloride (TMMC)], which behaves much more classically and is a better 1D magnet than $CsNiCl_3$.[18]

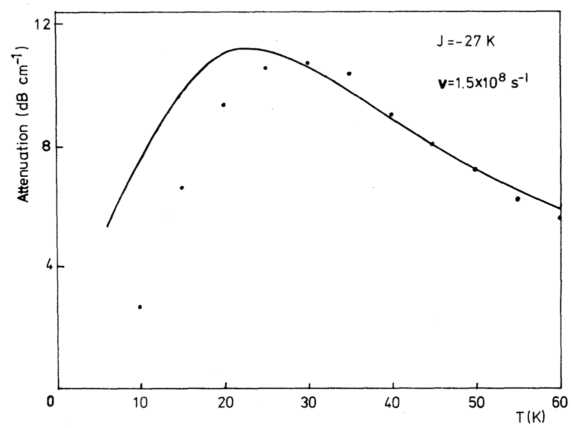

FIG. 1. Ultrasonic attenuation due to the spin ordering as a function of temperature. The solid line depicts the theoretical attenuation. The dots are experimental points for $CsNiCl_3$ taken from Almond and Rayne (Ref. 13). Phonon-phonon interactions become important at 70 K.

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