Gapless phasons and transverse sound-wave asymmetries in K_2SeO_4

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Based on an inelastic neutron scattering experiment, it has recently been suggested that phasons in the incommensurate phase of K_2SeO_4 possess a gap of approximately 50 GHz [M. Quilichini and R. Currat, Solid State Commun. **48**, 1011 (1983)]. We have found that these data may also be explained using a coupled phason sound-wave theory, for which the phasons are gapless. We then use the coupled excitation theory to predict certain velocity and attenuation asymmetries for transverse sound waves under interchange of their propagation and polarization directions. Lastly, we find that, in the long-wavelength limit, the attenuation asymmetry present for a system with gapless phasons is absent for a system possessing a phason gap.

I. INTRODUCTION

The incommensurate phase of potassium selenate has been extensively studied over the last decade, and Axe *et al.*[1] have recently reviewed much of this work. Of particular interest is the inelastic neutron scattering experiment of Quilichini and Currat[2] which was consistent with a phason branch with a gap of approximately 50 GHz. To be specific, their data analysis was dependent upon an overdamped phase excitation. The intensity of scattered neutrons is directly proportional to a response function $S(\mathbf{k},\omega)$, and Quilichini and Currat employed the damped oscillator response function given by

$$S^0(\mathbf{k},\omega)=\frac{\Gamma}{[\omega^2-\omega_\phi^2(\mathbf{k})]^2+(\omega\Gamma)^2} . \tag{1}$$

At 120 K, their data led to the interpretation that for $\mathbf{k}$ parallel to $\mathbf{a}^*$

$$\omega_\phi(k)=\omega_0+vk \tag{2}$$

with a gap frequency of $\omega_0=0.05$ THz, a phason velocity of $v=266$ m/s, and a phason damping constant of $\Gamma=0.31$ THz, independent of k. They estimated an error of $\pm10\%$ for Γ. Inoue and Ishibashi[3] have found Raman scattering results which seem to be consistent[1] quantitatively with the results expressed by Eqs. (1) and (2).

Axe *et al.*[1] have discussed these results and suggest that the gap may be explained by the following considerations. On the one hand, higher harmonics describing the modulation pattern may be present at temperatures near the paraelectric-incommensurate transition ($T_i=129$ K, $T_c=93$ K). Then, in the continuum description of the modulation pattern, the soliton or domain-wall picture[4] may be applicable. However, analysis of the discreteness of such systems[4,5] has suggested that the domain walls can be pinned, in contrast to the continuum solutions for which they are unpinned. For example, it can happen that the local wall energy is much greater than the wall-wall interaction energy, and then the phason gap can correspond to a domain wall oscillating about some equilibrium position. On the other hand, a distribution of impurities in the crystal can pin the modulation pattern, directly leading to a gap.

Besides the presence of a phason gap, a possible explanation of the observed lineshapes concerns the coupling of the phason with other excitations in the crystal. The authors of Ref. 2 do note that the phason couples to the longitudinal acoustic phonon (for $\mathbf{k}$ parallel to $\mathbf{a}^*$), but have discounted the possibility that this can explain their experimental results. As we shall detail below, our analysis uses precisely this coupling to demonstrate the compatibility of their data with a coupled phason and sound-wave system.

The present authors have recently formulated a macroscopic theory[6] of the long-wavelength excitations of incommensurate solids for which $\omega\rightarrow0$ as $k\rightarrow0$. Central to this theory is a coupling of the phason and center-of-mass motions and a consequence of this coupling is that the phason-phason response function will be different from that given by Eq. (1). In Sec. II we show that the resulting response function, when applied to K_2SeO_4 for $\mathbf{k}$ parallel to $\mathbf{a}^*$, is in excellent agreement with $S^0(\mathbf{k},\omega)$, even when no phason gap is included. Thus the coupled excitation theory of Ref. 6 provides an alternate explanation to the data of Ref. 2.

One of the predictions of the theory of Ref. 6 concerns transverse sound-wave asymmetries. For a given transverse sound-wave normal mode, with certain propagation and polarization directions, it may happen that another sound-wave normal mode exists for these directions being interchanged. According to Ref. 6, in the long-wavelength limit these modes, in general, will have different attenuations. In Sec. III we examine some of the sound-wave solutions for incommensurate K_2SeO_4 that result from the coupled excitation theory. We display the transition from the low- to the high-frequency regime for a system with typical sound-wave and phason velocity and damping parameters, and then display the solutions that are obtained for the incommensurate phase of K_2SeO_4 in both of these limits.

The presence of transverse sound-wave asymmetries in incommensurate solids was first observed in $BaMnF_4$ by

Fritz,[7] and later observed in $RbH_3(SeO_3)_2$ by Esayan *et al.*[8] An explanation of these observations has been proposed by Scott[9] and is based on nonequilibrium defects being present in certain incommensurate crystals. Dvorak and Esayan[10] have used a Landau-type theory to discuss the $RbH_3(SeO_3)_2$ experiment and predict that the results may be explained via dispersive sound velocities. In Sec. III we shall show that our theory, over a small range of wave vectors, also produces dispersive sound velocities, although in the high-frequency regime the sound waves reobtain a nondispersive character. Finally, Poulet and Pick[11] have produced an elegant theory of coupled phasons and acoustic phonons based on a Landau-type theory. However, unlike our theory, their treatment of this coupling ignores the damping of these motions. In the high-frequency regime we shall show that our theory and that of Poulet and Pick predict identical sound wave asymmetries. This is to be expected since at high frequencies the damping of the propagating modes will not effect their speeds.

Next, in Sec. IV an extension of the theory of Ref. 6 is made to systems possessing a phason gap. It is found that the long-wavelength attenuation asymmetry, under interchange of the directions of propagation and polarization, present for systems with gapless phasons, is removed for systems possessing a phason gap. Then, for the incommensurate phase of K_2SeO_4, we propose a possible test for determining whether or not the 50 GHz phason gap suggested by the authors of Ref. 2 is indeed present in the incommensurate phase of K_2SeO_4. Finally, in Sec. V we summarize the application of the coupled phason and sound-wave theory to K_2SeO_4.

II. RESPONSE FUNCTION FOR COUPLED EXCITATIONS

In order to derive the necessary phason-phason response function, we require the Lagrangian density and dissipation function introduced in Ref. 6 that are appropriate for K_2SeO_4. Thus we need (i) the form of the relative phase-displacement field and (ii) the nonzero tensor coefficients which enter into these functions which are consistent with the incommensurate crystal's point-group symmetry. The relative phase-displacement field, viz. the vector field describing the motion of the modulation pattern with respect to the underlying crystal lattice, has only one component for K_2SeO_4. We shall denote it by U_x where we follow the convention[2] that the (fundamental) incommensurate-modulation wave vector is given by $\mathbf{q}_i=\frac{1}{3}(1-\delta)\mathbf{a}^*$. Furthermore, Janner and Janssen[12] have determined the superspace group of the incommensurate phase of K_2SeO_4 and have found the associated point-group symmetry to be orthorhombic *mmm*. Thus following the notation of Ref. 6, the Lagrangian density appropriate to the incommensurate phase of K_2SeO_4 is given by

$$\begin{aligned}\mathcal{L}=&\tfrac{1}{2}\rho\dot{u}_i\dot{u}_i+\tfrac{1}{2}\rho_{xx}\dot{U}_x^2-\tfrac{1}{2}\lambda_{xxxx}u_{(xx)}^2-\tfrac{1}{2}\lambda_{yyyy}u_{(yy)}^2-\tfrac{1}{2}\lambda_{zzzz}u_{(zz)}^2-\lambda_{yyzz}u_{(yy)}u_{(zz)}\\&-\lambda_{zzxx}u_{(zz)}u_{(xx)}-\lambda_{xxyy}u_{(xx)}u_{(zz)}-2\lambda_{xyxy}u_{(xy)}^2-2\lambda_{zxzx}u_{(zx)}^2-2\lambda_{yzyz}u_{(yz)}^2-\tfrac{1}{2}\Lambda_{xxxx}U_{x,x}^2\\&-\tfrac{1}{2}\Lambda_{xyxy}U_{x,y}^2-\tfrac{1}{2}\Lambda_{xzxz}U_{x,z}^2-\Pi_{xxxx}u_{(xx)}U_{x,x}-2\Pi_{xyxy}u_{(xy)}U_{x,y}-2\Pi_{zxxz}u_{(zx)}U_{x,z}\ .\end{aligned}\tag{3}$$

Also, the dissipation function is given by

$$R=\tfrac{1}{2}\zeta_{ijkl}\dot{u}_{(ij)}\dot{u}_{(kl)}+\frac{\rho_{xx}}{2\tau}\dot{U}_x^2\ ,\tag{4}$$

where the components of the tensor ζ_{ijkl} that are nonzero are the same as those of the λ_{ijkl} tensor which appear in Eq. (3). In the above equations, u_i for $i=x,y,z$ represents the components of the center-of-mass displacement field, $u_{(ij)}$ the linearized symmetric strain tensor, $U_{x,i}\equiv\partial U_x/\partial x_i$ the components of the relative phase-displacement field gradient, the summation convention is used and the overdot represents differentiation with respect to time. Note that the ξ_{ijx} terms in Eq. (32) of Ref. 6 vanish due to the *mmm* orthorhombic point-group symmetry. Also note that this point-group symmetry requires that no linear piezoelectric effects be present.[6]

If $\mathbf{k}$ is parallel to $\mathbf{a}^*$ (in our coordinate system this implies $\mathbf{k}=k\hat{\mathbf{e}}_x$, $\hat{\mathbf{e}}_x$ being a unit vector in the x direction), one then has that the phason-phason response function is given by

$$S^1(k,\omega)=\frac{1}{\omega}\mathrm{Im}\chi_{U,U}(k,\omega)\ ,\tag{5}$$

where

$$\begin{aligned}\chi_{U,U}(k,\omega)=&-(\omega^2-v_l^2k^2+id_l\omega k^2)\\&\times\{(\omega^2-v_l^2k^2+id_l\omega k^2)[\omega^2-v_p^2k^2+i\Gamma(k)\omega]\\&-(\Pi k^2)^2\}^{-1}\ .\end{aligned}\tag{6}$$

The function defined in Eq. (5), like that of Eq. (1), is directly proportional to the intensity of neutrons scattered by the phason modes, where $\mathbf{k}=k\hat{\mathbf{e}}_x$ is the wave vector of these excitations relative to the incommensurate wave vector $\mathbf{q}_i$. The constants in these equations are defined by

$$\begin{aligned}v_l&\equiv\left[\frac{\lambda_{xxxx}}{\rho}\right]^{1/2},\\d_l&\equiv\frac{\zeta_{xxxx}}{\rho}\ ,\\v_p&\equiv\left[\frac{\Lambda_{xxxx}}{\rho_{xx}}\right]^{1/2},\\\Pi&\equiv\frac{\Pi_{xxxx}}{(\rho\rho_{xx})^{1/2}}\ .\end{aligned}\tag{7}$$

Note that we have accounted for the dispersion of the phason damping constant by including an additional term in the dissipation function expressed by Eq. (4), viz.

$$R' = \rho_{xx} d_p \dot{U}^2_{x,x} \tag{8}$$

such that in Eq. (6) we have

$$\Gamma(k) = \tau^{-1} + d_p k^2 . \tag{9}$$

In order to apply Eq. (5) to K_2SeO_4 we have used the following numerical values for the longitudinal sound wave. From the ultrasonic data of Rehwald *et al.*[13] we take C_{11} $(=\lambda_{xxxx})=53$ GPa, $\rho=3066$ kg m^{-3}, and thus $v_l=4160$ m/s. No data is available for the attenuation of this mode. However, from the transverse acoustic peaks of Fig. 2 of Ref. 2 one may estimate[14] that d_l is of the order of 10 THz Å^2; we note that choosing any value of d_l between 0 and 100 THz Å^2 leaves our results unaffected.

Figure 1 displays S^0 and S^1 for $k=0.04$, 0.06, and 0.08 Å^{-1} over the range of frequencies $0.025 \le \omega \le 0.3$ THz (thus ignoring the central-peak contribution) for the following parameter values: $v_p=346$ m/s, $\Pi=30$ THz^2 Å^2, $\tau^{-1}=0.235$ THz, and $d_p=14$ THz Å^2. Thus $\Gamma(k)$ varies between 0.26 and 0.32 THz in approximate agreement with the value of 0.31 THz $\pm 10\%$ estimated by Quilichini and Currat.[2] The curves in Fig. 1 are scaled such that their maximum value over this frequency range is 1.

Clearly, the response functions are very nearly equal. Thus as an alternative to the conclusion of Ref. 2, we find that the coupling of the longitudinal acoustic phonon to a gapless phason can account for the observed lineshape anomalies. In fact, if one desires to know whether or not the phasons in K_2SeO_4 are gapless, one must rely on further experimental studies. In Secs. III and IV we discuss how studies of the sound waves could provide this information.

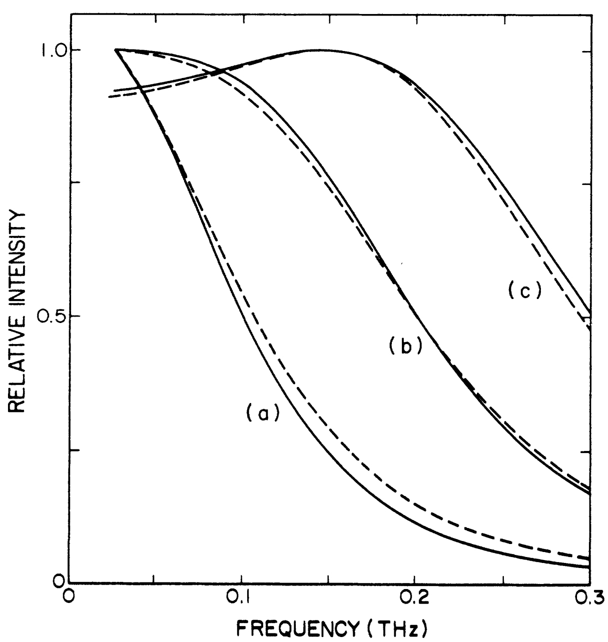

FIG. 1. The response functions given by $S^0(k,\omega)$ in Eqs. (1) and (2) (solid lines) and $S^1(k,\omega)$ in Eqs. (5) and (6) (dashed lines) for (a) $k=0.04$ Å^{-1}, (b) $k=0.06$ Å^{-1}, and (c) $k=0.08$ Å^{-1}, scaled to a maximum value of 1 over this frequency range.

As a final point, we present the long-wavelength phason eigenfrequencies which are predicted from both the gapless phason theory of Eqs. (3) and (4) and the response function of Quilichini and Currat. For the coupled excitation theory we find that as $k \to 0$ the phason eigenfrequency behaves according to

$$\omega = -i\tau \left[v_p^2 - \frac{\Pi^2}{v_l^2} \right] k^2 , \tag{10a}$$

or

$$\omega = -iDk^2 . \tag{10b}$$

Using the values obtained in this section, we find $D=49$ THz Å^2. However, in the presence of a gap Eq. (1) shows that at $k=0$ the phason eigenfrequency behaves as

$$\omega = \frac{-i\Gamma}{2} + (-\Gamma^2 + 4\omega_0^2)^{1/2} . \tag{11a}$$

Using the values given in Ref. 2 we have that Eq. (11a) becomes

$$\omega = -\frac{i}{T} , \tag{11b}$$

where $T^{-1}=10$ GHz. Thus the gapless phason theory predicts a diffusive mode at long wavelengths, while the presence of a gap produces a fast relaxing mode.

III. TRANSVERSE SOUND-WAVE ASYMMETRIES

Before we solve for the sound-wave normal modes, we wish to demonstrate the low- and high-frequency regimes[15] that can result for the eigenfrequencies of sound waves in incommensurate systems, based on the theory of Ref. 6. Suppose that a coupled phason sound-wave system has eigenfrequencies corresponding to the poles of $\chi_{U,U}(k,\omega)$, defined in Eq. (6). Then, for $v_l=1000$ m/s, $v_p=500$ m/s, $d_l=8$ THz Å^2 and $\Gamma(k)=0.06$ THz, independent of k, we plot the resulting ω_R/k values, where ω_R is the real part of the sound-wave eigenfrequency, for $\Pi=16$, 32, and 48 (THz Å)^2, in Fig. 2. We shall refer to the low-frequency regime as that for which

$$\frac{\omega_R}{k} \approx \lim_{k\to 0} \frac{\omega_R}{k} . \tag{12}$$

Figure 2 shows that when $k \ll 0.0005$ Å^{-1}, Eq. (12) is satisfied. Also, from this figure we see that at shorter wavelengths, viz. for the wave vector being greater than 0.02 Å^{-1}, this ratio approaches a constant value that is larger than that implicit in Eq. (12). (It is important to note that the increase in the sound velocity occurs because the bare speed of sound, viz. v_l, is greater than the bare phason speed, viz. v_p. If we considered a system for which $v_p > v_l$ we would find that the speed of the sound-

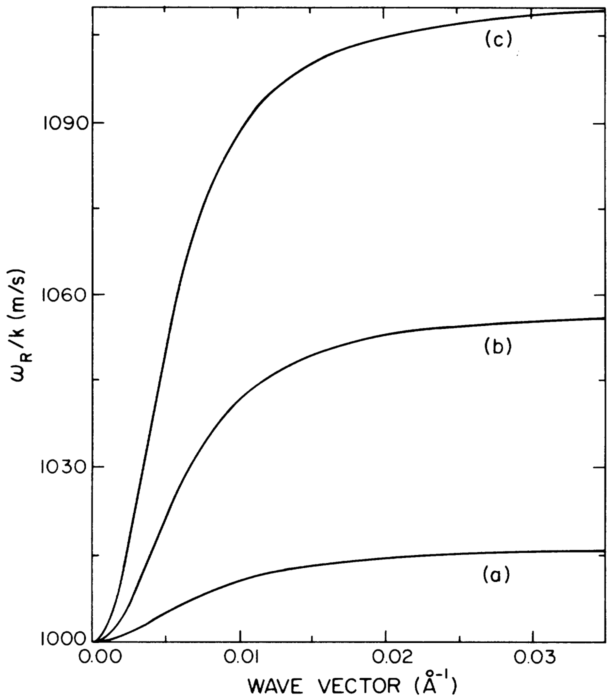

FIG. 2. The real part of the sound-wave eigenfrequency divided by k, found from the poles of Eq. (6) where $v_l = 1000$ m/s, $v_p = 500$ m/s, $d_l = 8$ THz $\mathrm{\AA}^2$, and $\Gamma(k) = 0.06$ THz, for $\Pi = 16$ (a), 32 (b), and 48 (c) (THz $\mathrm{\AA}$)^2. If $\Pi = 0$ this ratio would remain at the constant value of 1000 m/s ($= v_l$).

wave mode decreased for increasing k.) For the three Π values, we see that at shorter wavelengths the ratio of ω_R/k is greater than v_l by 1.6, 5.6, and 10.9 %, respectively. We shall refer to this regime as the high-frequency regime. Furthermore, we shall use the wave vector k_c to represent the cross-over region.

Now we examine the sound-wave solutions corresponding to particular transverse modes of the incommensurate phase of K_2SeO_4. We assume plane-wave solutions characterized by wave vector $\mathbf{k} = k\hat{\boldsymbol{\mu}}$, $\hat{\boldsymbol{\mu}}$ being a unit vector. Firstly, we consider the $\mathbf{k} = k\hat{\mathbf{e}}_x$ transverse sound wave whose eigenfrequency is found from the solution of

$$\rho \ddot{u}_y = \lambda_{xyxy} u_{y,xx} + \zeta_{xyxy} \dot{u}_{y,xx} \; . \tag{13}$$

This mode is polarized along the y direction. Then, for $\mathbf{k} = k\hat{\mathbf{e}}_y$, we consider a transverse sound wave polarized along the x direction which is found when one solves the coupled equations of motion given by

$$\rho \ddot{u}_x = \lambda_{xyxy} u_{x,yy} + \Pi_{xyxy} U_{x,yy} + \zeta_{xyxy} \dot{u}_{x,yy} \; , \tag{14a}$$

$$\rho_{xx} \ddot{U}_x = \Lambda_{xyxy} U_{x,yy} + \Pi_{xyxy} u_{x,yy} - \frac{\rho_{xx}}{\tau} \dot{U}_x \; . \tag{14b}$$

For a given k we choose to denote the eigenfrequencies found from Eqs. (13) and (14) by $\omega(\hat{\boldsymbol{\mu}}, \hat{\mathbf{v}})$, where $\hat{\mathbf{v}}$ is a unit vector in the direction of the polarization. The form of these solutions may be expressed as a power series in k:

$$\omega(\hat{\boldsymbol{\mu}}, \hat{\mathbf{v}}) = \pm v(\hat{\boldsymbol{\mu}}, \hat{\mathbf{v}})k - iD(\hat{\boldsymbol{\mu}}, \hat{\mathbf{v}})k^2 + O(k^3) \; . \tag{15}$$

In the analytic results given below, we shall ignore the third and higher order terms in k.

The $\omega(\hat{\mathbf{e}}_x, \hat{\mathbf{e}}_y)$ solution is obtained from Eq. (13). We find

$$v(\hat{\mathbf{e}}_x, \hat{\mathbf{e}}_y) = \left[\frac{\lambda_{xyxy}}{\rho} \right]^{1/2} , \tag{16}$$

$$D(\hat{\mathbf{e}}_x, \hat{\mathbf{e}}_y) = \frac{1}{2} \left[\frac{\zeta_{xyxy}}{\rho} \right] , \tag{17}$$

provided that

$$k^2 \ll \frac{4\rho\lambda_{xyxy}}{\zeta_{xyxy}^2} \; . \tag{18}$$

Since the center-of-mass displacement field is uncoupled from the relative phase-displacement field, as shown in Eq. (13), one does not speak of two frequency regimes for this mode. Instead, Eq. (18) mathematically states the propagating character of this long-wavelength mode.

Solutions for $\omega(\hat{\mathbf{e}}_y, \hat{\mathbf{e}}_x)$, however, do involve coupled excitations, as seen from Eqs. (14a) and (14b). In the low-frequency regime we find that

$$v(\hat{\mathbf{e}}_y, \hat{\mathbf{e}}_x) = v(\hat{\mathbf{e}}_x, \hat{\mathbf{e}}_y) \; , \tag{19}$$

$$D(\hat{\mathbf{e}}_y, \hat{\mathbf{e}}_x) = D(\hat{\mathbf{e}}_x, \hat{\mathbf{e}}_y) + \frac{1}{2} \left[\frac{\tau \Pi_{xyxy}^2}{\lambda_{xyxy}} \right] , \tag{20}$$

results that may also be obtained directly from Eqs. (31) and (32) of Ref. 6. The high-frequency regime results may be expressed in terms of

$$v_p \equiv \left[\frac{\Lambda_{xyxy}}{\rho_{xx}} \right]^{1/2} , \tag{21a}$$

$$\Pi \equiv \frac{\Pi_{xyxy}}{(\rho\rho_{xx})^{1/2}} , \tag{21b}$$

where we shall use a weak-coupling approximation, viz. $|v^2(\hat{\mathbf{e}}_x, \hat{\mathbf{e}}_y) - v_p^2| \gg \Pi$. For the speed of this mode we find

$$v^2(\hat{\mathbf{e}}_y, \hat{\mathbf{e}}_x) = v^2(\hat{\mathbf{e}}_x, \hat{\mathbf{e}}_y) + \frac{\Pi^2}{[v^2(\hat{\mathbf{e}}_x, \hat{\mathbf{e}}_y) - v_p^2]} , \tag{22}$$

from which it follows that if the transverse speed of sound $v(\hat{\mathbf{e}}_x, \hat{\mathbf{e}}_y)$ is greater than the bare phason speed, viz. v_p, then $v(\hat{\mathbf{e}}_y, \hat{\mathbf{e}}_x) > v(\hat{\mathbf{e}}_x, \hat{\mathbf{e}}_y)$. Note that Eq. (22) differs fundamentally from the theory of Dvorak and Esayan[10] [that was applied to the incommensurate phase of $RbH_3(SeO_3)_2$] in that the sound-velocity asymmetry expressed by Eq. (22) is independent of k in the high-frequency regime. Also note that Eq. (22) is identical to the result that follows from Eq. (4-10) of Poulet and Pick[11] (for their coupling parameter $h^1_{\alpha\beta,\gamma}$ satisfying $h^1_{xy,y} \propto \Pi$) when the coupling to the amplitude mode is ignored. For the attenuation of this mode we find

$$D(\hat{\mathbf{e}}_y, \hat{\mathbf{e}}_x) = D(\hat{\mathbf{e}}_x, \hat{\mathbf{e}}_y) \left[1 - \frac{\Pi^2}{[v^2(\hat{\mathbf{e}}_x, \hat{\mathbf{e}}_y) - v_p^2]^2} \right] . \tag{23}$$

Thus we see that this asymmetry is reversed in comparison to that expressed by Eq. (20).

Note that $\omega(\hat{\mathbf{e}}_x,\hat{\mathbf{e}}_z)$ and $\omega(\hat{\mathbf{e}}_z,\hat{\mathbf{e}}_x)$ are also eigenfrequencies of normal modes and thus all of the above relations apply if (i) $\hat{\mathbf{e}}_y$ is replaced by $\hat{\mathbf{e}}_z$, and (ii) the appropriate tensor coefficients are interchanged (e.g., $\Pi_{xyxy}\rightarrow\Pi_{zxxz}$).

Our theory thus predicts the following relations: For $k \ll k_c$,

$$v(\hat{\mathbf{e}}_y,\hat{\mathbf{e}}_x)=v(\hat{\mathbf{e}}_x,\hat{\mathbf{e}}_y)\ , \tag{24a}$$

$$D(\hat{\mathbf{e}}_y,\hat{\mathbf{e}}_x)>D(\hat{\mathbf{e}}_x,\hat{\mathbf{e}}_y)\ , \tag{24b}$$

$$v(\hat{\mathbf{e}}_z,\hat{\mathbf{e}}_x)=v(\hat{\mathbf{e}}_x,\hat{\mathbf{e}}_z)\ , \tag{24c}$$

$$D(\hat{\mathbf{e}}_z,\hat{\mathbf{e}}_x)>D(\hat{\mathbf{e}}_x,\hat{\mathbf{e}}_z)\ , \tag{24d}$$

and for $k_c \ll k$,

$$v(\hat{\mathbf{e}}_y,\hat{\mathbf{e}}_x)\gtrless v(\hat{\mathbf{e}}_x,\hat{\mathbf{e}}_y)\ , \tag{25a}$$

$$D(\hat{\mathbf{e}}_y,\hat{\mathbf{e}}_x)<D(\hat{\mathbf{e}}_x,\hat{\mathbf{e}}_y)\ , \tag{25b}$$

$$v(\hat{\mathbf{e}}_z,\hat{\mathbf{e}}_x)\gtrless v(\hat{\mathbf{e}}_x,\hat{\mathbf{e}}_z)\ , \tag{25c}$$

$$D(\hat{\mathbf{e}}_z,\hat{\mathbf{e}}_x)<D(\hat{\mathbf{e}}_x,\hat{\mathbf{e}}_z)\ , \tag{25d}$$

where the greater than or less than inequality of Eq. (25a) pertains to $v(\hat{\mathbf{e}}_x,\hat{\mathbf{e}}_y)>v_p$ or $v(\hat{\mathbf{e}}_x,\hat{\mathbf{e}}_y)<v_p$, respectively. Similiarly for Eq. (25c). Also note that k_c for the $\omega(\hat{\mathbf{e}}_y,\hat{\mathbf{e}}_x)$ mode is not necessarily the same as that of the $\omega(\hat{\mathbf{e}}_z,\hat{\mathbf{e}}_x)$ mode.

Unfortunately, the data of Ref. 2 does not allow for the numerical estimation of ρ_{xx}, Λ_{xyxy}, Λ_{xzxz}, Π_{xyxy}, and Π_{zxxz}. A number of experiments for **k** parallel to both **b*** and **c*** would be required. Thus we cannot predict whether or not these asymmetries are large enough to be seen. These parameters are also required in order to estimate the k_c values and thus we cannot predict for what wavelengths one could possibly see the high-frequency regime results. However, if these asymmetries are observed the validity of applying the gapless phason theory of Ref. 6 to K_2SeO_4 [as expressed by Eqs. (3) and (4)] will be strongly supported.

IV. SOUND-WAVE EIGENFREQUENCIES FOR PHASONS WITH A GAP

The results of Sec. III were derived for an incommensurate phase for which no phason gap is present. However, all real crystals have some impurities, as well as other imperfections, and thus according to the present theoretical view all real incommensurate crystals will have a small but nonzero phason gap. In this section we address the following question: how will the presence of a phason gap affect the asymmetries that are predicted in Eqs. (24) and (25)?

The inclusion of a gap in the phason spectrum has been discussed by a number of authors.[5,16–19] Here we extend the coupled excitation theory of Ref. 6 and incorporate a phason gap. In an incommensurate system for which the modulation pattern is pinned, we assume a rigid translation of the relative phason-displacement field by a small distance U produces a restoring force directly proportional to U. Thus in the elastic-energy density we must include a term like $\frac{1}{2}C\omega_0^2U^2$, where ω_0 is the phason gap and C is a constant. Restricting our attention to contributions to the Lagrangian density that are the lowest order in k, we find that the most general expression for an incommensurate crystal with a phason gap (that is consistent with the rotational invariance of $\mathcal{L}$) is given by

$$\mathcal{L}=\tfrac{1}{2}\rho\dot{u}_i\dot{u}_i+\tfrac{1}{2}\rho_{ab}\dot{U}_a\dot{U}_b -\tfrac{1}{2}\lambda_{ijkl}u_{(ij)}u_{(kl)}-\tfrac{1}{2}\tilde{\rho}_{ab}\omega_0^2U_aU_b-\kappa_{ija}u_{(ij)}U_a\ , \tag{26}$$

where

$$\rho_{ab}=\rho_{ba}\ , \tag{27a}$$

$$\lambda_{ijkl}=\lambda_{klij}=\lambda_{jikl}=\lambda_{ijlk}\ , \tag{27b}$$

$$\tilde{\rho}_{ab}=\tilde{\rho}_{ba}\ , \tag{27c}$$

$$\kappa_{ija}=\kappa_{jia}\ , \tag{27d}$$

with $i,j,k,l=x,y,z$, a,b label the components of the relative phase-displacement field and the summation convention is used. The dissipation function is the same as that of Ref. 6, viz.

$$R=\tfrac{1}{2}\zeta_{ijkl}\dot{u}_{(ij)}\dot{u}_{(kl)}+\tfrac{1}{2}\eta_{ab}\dot{U}_a\dot{U}_b+\xi_{ija}\dot{u}_{(ij)}\dot{U}_a\ , \tag{28}$$

where

$$\zeta_{ijkl}=\zeta_{klij}=\zeta_{jikl}=\zeta_{ijlk}\ , \tag{29a}$$

$$\eta_{ab}=\eta_{ba}\ , \tag{29b}$$

$$\xi_{ija}=\xi_{jia}\ . \tag{29c}$$

The resulting equations of motion are

$$\rho\ddot{u}_i=\lambda_{ijkl}u_{l,jk}+\kappa_{ija}U_{a,j}+\zeta_{ijkl}\dot{u}_{l,jk}+\xi_{ija}\dot{U}_{a,j}\ , \tag{30a}$$

$$\rho_{ab}\ddot{U}_b=\tilde{\rho}_{ab}\omega_0^2U_b+\kappa_{ija}u_{i,j}-\eta_{ab}\dot{U}_b-\xi_{ija}\dot{u}_{i,j}\ . \tag{30b}$$

We now solve for the sound-wave eigenfrequencies by assuming $\omega=O(k)$. Then, the contribution of the relative phase-displacement field to the sound-wave eigenvectors is determined by Eq. (30b), and for $k\rightarrow 0$ we find

$$U_a=-\frac{\sigma_{ab}\kappa_{ijb}}{\omega_0^2}\frac{\partial u_i}{\partial x_j}\ , \tag{31}$$

where the matrix σ is defined by

$$\sigma_{ab}\rho_{bc}=\delta_{ac}\ . \tag{32}$$

Equation (31) is valid when ω is such that

$$\tilde{\rho}_{ab}\omega_0^2\gg\rho_{ab}\omega^2+i\eta_{ab}\omega \tag{33}$$

is satisfied.

From Eq. (31) we see that $U\sim O(k)u$. Then, substitution of Eq. (31) into Eq. (30a) yields sound-wave eigenvalues for $k\rightarrow 0$. We find [see Eq. (15)]

$$v(\hat{\boldsymbol{\mu}},\hat{\boldsymbol{\nu}})=(\rho^{-1}\lambda'_{ijkl}\nu_i\mu_j\mu_k\nu_l)^{1/2}\ , \tag{34a}$$

$$D(\hat{\boldsymbol{\mu}},\hat{\boldsymbol{\nu}})=\tfrac{1}{2}\rho^{-1}\zeta'_{ijkl}\nu_i\mu_j\mu_k\nu_l\ , \tag{34b}$$

where the renormalized stiffness and viscosity tensors are given by

$$\lambda'_{ijkl}=\lambda'_{jilk}$$

$$=\lambda_{ijkl}-\frac{\kappa_{ija}\kappa_{klb}\sigma_{ab}}{\omega_0^2}\ , \tag{35a}$$

$$\zeta'_{ijkl}=\zeta'_{jilk}$$

$$=\zeta_{ijkl}-\frac{\kappa_{ija}\xi_{klb}\sigma_{ab}}{\omega_0^2}\ . \tag{35b}$$

From Eqs. (34) and (35) we see that $v(\hat{\mu},\hat{v})=v(\hat{v},\hat{\mu})$ and $D(\hat{\mu},\hat{v})=D(\hat{v},\hat{\mu})$. This last relation is in direct contradiction to the gapless phason theory of Ref. 6, where it was found that in the long-wavelength limit, in general, $D(\hat{\mu},\hat{v})\neq D(\hat{v},\hat{\mu})$.

From Eq. (33) one may show that Eqs. (34) and (35) may be applied to the incommensurate phase of K_2SeO_4, in which a phason gap is present, provided that $\omega \ll \min(\omega_0,\omega_0^2/\Gamma)$. Using the values from Ref. 2 this restriction implies that $\omega \ll 0.01$ THz. Thus we propose that if an experimental probe for which $\omega \leq 0.001$ THz observes the attenuation asymmetries expressed by Eqs. (24b) and/or (24d), the 50 GHz phason gap suggested in Ref. 2 cannot be present in the incommensurate phase of K_2SeO_4.

V. SUMMARY

We have shown that the phason gap suggested in Ref. 2 is not essential in analyzing their lineshape anomalies. Instead, the phason-phason response function derived from the coupled excitation theory of Ref. 6 provides an alternate explanation.

The gapless-phason coupled excitation theory predicts certain sound-wave asymmetries. To be specific, under interchange of the directions of propagation and polarization we find attenuation asymmetries in the low-frequency regime, viz. those expressed in Eq. (24), as well as sound-velocity and attenuation asymmetries in the high-frequency regime, viz., those expressed by Eq. (25).

Finally, we showed how a phason gap may be included into the coupled excitation theory of Ref. 6. Then, at sufficiently long wavelengths the attenuation asymmetry is predicted to be absent. Thus if a low-frequency probe (viz. energies less than 1 GHz) does observe the asymmetries expressed in Eq. (24), a 50-GHz phason gap cannot be present in the incommensurate phase of K_2SeO_4.

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[1]J. D. Axe, M. Iizumi, and G. Shirane, in *Incommensurate Phases in Dielectrics,* edited by R. Blinc and A. P. Levanyuk (North-Holland, Amsterdam, 1986), Vol. II.

[2]M. Quilichini and R. Currat, Solid State Commun. **48**, 1011 (1983).

[3]K. Inoue and Y. Ishibashi, J. Phys. Soc. Jpn. **52**, 556 (1983).

[4]P. Bak, Rep. Prog. Phys. **45**, 587 (1982).

[5]D. A. Bruce, J. Phys. C **13**, 4615 (1980).

[6]R. J. Gooding and M. B. Walker, Phys. Rev. B **35**, 6831 (1987).

[7]I. J. Fritz, Phys. Lett. **51A**, 219 (1975).

[8]S. Kh. Esayan, V. V. Lemanov, M. Mamatkulov, and L. A. Shuvalov, Sov. Phys.—Crystal. **26**, 619 (1981).

[9]J. F. Scott, Ferroelectrics **66**, 11 (1986).

[10]V. Dvorak and S. Kh. Esayan, Solid State Commun. **44**, 901 (1982).

[11]H. Poulet and R. M. Pick, J. Phys. C **14**, 2675 (1981).

[12]A. Janner and T. Janssen, Acta Crystallogr. Sect. A **36**, 399 (1980).

[13]W. Rehwald, A. Vonlanthen, J. K. Kruger, R. Wallerius, and H. G. Unruh, J. Phys. C **13**, 3823 (1980).

[14]We used an uncoupled response function, viz. Eq. (1) with $\omega_\phi=v_t k$ and $\Gamma=d_l k^2$, to estimate d_l.

[15]The low- and high-frequency regimes have also been discussed for the excitations of the incommensurate phase of quartz: M. B. Walker and R. J. Gooding, Phys. Rev. B **32**, 7412 (1985).

[16]S. Aubry, in *Solitons in Condensed Matter Physics,* edited by A. R. Bishop and T. Schneider (Springer-Verlag, Berlin, 1979).

[17]A. D. Bruce, R. A. Cowley, and A. F. Murray, J. Phys. C **11**, 3591 (1978).

[18]A. D. Bruce and R. A. Cowley, J. Phys. C **11**, 3609 (1978).

[19]R. Zeyher, Ferroelectrics **66**, 217 (1986).