Chirality-induced Phonon Dispersion in a Noncentrosymmetric Micropolar Crystal

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Features of the phonon spectrum of a chiral crystal are examined within the micropolar elasticity theory. This formalism accounts for not only translational micromotions of a medium but also rotational ones. It is found that there appears the phonon band splitting depending on the left/right-circular polarization in a purely phonon sector without invoking any outside subsystem. The phonon spectrum reveals parity breaking while preserving time-reversal symmetry, i.e. it possesses true chirality. We find that hybridization of the micro-rotational and translational modes gives rise to the acoustic phonon branch with a “roton” minimum reminiscent of the elementary excitations in the superfluid helium-4. We argue that a mechanism of this phenomena is in line with Nozières’ reinterpretation \[ J. \text{Low Temp. Phys.} \textbf{137}, 45 (2004) \] of the rotons as a manifestation of an incipient crystallization instability. We discuss a close analogy between the translational and rotational micromotions in the micropolar elastic medium and the Bogoliubov quasiparticles and gapful density fluctuations in $^4$He.

Introduction.— In hierarchy of electric, magnetic and mechanical degrees of freedom and attendant interrelationships namely mechanical and elastic properties of solids are most sensitive to structure. This is why the mechanical response in solids has recently been the target of a new branch of electronics, referred to as straintronics\textsuperscript{[1]}. Connection of structural chirality with static and dynamical properties offers a key to understand functionality of chiral systems\textsuperscript{[2]}. A quintessential example of the chirality-controlled phenomena is the optical activity\textsuperscript{[3]}, where propagation of circularly polarized light through a chiral material depends on its handedness. As for elastic degrees of freedom, a mechanical counterpart of optical activity, so-called acoustical activity, has been attracting revived attention\textsuperscript{[4]}. This phenomena was first predicted by Portigal and Burstein\textsuperscript{[5]} and direct observation thereof was subsequently provided for the α−quartz crystal belonging to an enantiomorphic space group\textsuperscript{[6]}.

The optical activity, as a paradigm for chirality, is an example of true chirality according to Laurence Barrons’ definition\textsuperscript{[2]}, i.e., true chirality is exhibited by systems, where parity $P$ is broken but not time reversal $T$ combined with any proper rotation $R$. This modified definition is quite natural in the sense that the dynamical meaning of chirality is a coupling of rotation and translation, i.e. it is an equivalent of helicity. The optical activity is also known to result from a nonlocal response of a crystal to a light wave, when there are first-order spatial dispersion contributions to the dielectric constant determined via the optical gyrotropic tensor. This situation is somewhat similar to a role of Dzyaloshinskii-Moriya interaction which leads to a linear Lifshitz invariant in free energy of a chiral helimagnet\textsuperscript{[7,8]}.

The acoustical activity\textsuperscript{[5]} is also related with first-order spatial dispersion contributions to the elastic constants. However, chirality effects are beyond the conventional elasticity theory\textsuperscript{[9]}, which considers only a local translation of points and the force stress (force per unit area) but completely ignores a local rotation of these points and the concomitant couple stress (a torque per unit area). As a consequence, the elastic four-rank tensor, $c_{ijkl}$, while reflecting crystal symmetry, can be neither pseudotensor nor axial one.

These missing effects, which may be viewed as a particular manifestation of nonlocality, are addressed in the micropolar elasticity theory\textsuperscript{[10,11]}. This theory accommodates size effects in elastic behaviour, which are expected to appear as the consequence of the largest structural elements in solids, and allowed for consideration of chirality effects in the elastic media. The latter have recently become topics for study in metamaterials\textsuperscript{[12]} and in quasi two-dimensional monolayers of noncentrosymmetric tungsten diselenide crystals\textsuperscript{[13]}. It must be recognized, however, that applications of the micropolar elasticity theory to noncentrosymmetric materials have been mainly limited to mechanical engineering problems, including investigations of static deformations in bones and synthetic composites containing twisted fibers\textsuperscript{[14]}, an analysis of static and wave properties of tetrachiral lightweight lattices\textsuperscript{[15]}.

So far, only few attempts have been made to calculate dispersion curves of the micropolar elastic waves in crystals. We note in this regard the pioneering research undertaken by Pouget at al. for the compound KNO\textsubscript{3}\textsuperscript{[16]}. Motivated by the inelastic neutron scattering data\textsuperscript{[17]}, they presented a lattice theory of ferroelectric
crystals with molecular groups, which accounts for their microrotations added to the usual displacements. In the long-wavelength limit this approach coincides with the micropolar theory of elasticity. Dispersion curves demonstrate mixing of the longitudinal and transverse acoustic modes with the rotational one, but the spectrum remains symmetric against the sign of crystal momentum.

Then, the natural question arises as to whether a phonon spectrum in a chiral crystal exhibits non-reciprocity effects in propagation of micropolar elastic waves. This is the question addressed in this Letter. A salient feature of the nonreciprocity is a polarization-dependent splitting of phonon bands similar to the one for electronic bands due to spin-orbit coupling[18]. In contrast to the phonon magneto-chiral effect[19, 20], for which the splitting is achieved through coupling with nonreciprocal magnons, our aim is to find the result for a purely phononic sector without the involvement of any subsystem outside.

Another issue discussed in this Letter is how the structural chirality relates to the phonon angular momentum and spin. In this regard, we mention the recent studies of chiral phonons in monolayers of hexagonal[21] and kagome lattices[22]. In these systems, the phonon eigenmodes at high-symmetry points of the Brillouin zone (BZ) inherit the threefold rotational symmetry of the lattice which allows to label these phonon eigenmodes with pseudoangular momentum. It includes both orbital and spin parts, the latter coincides with the phonon chirality characterized by the circular polarization of phonons. In contrast to this scenario in which chirality is assigned only to special BZ points, we examine an effect of the structural chirality on phonon dispersion over the whole Brillouin zone. The chiral helimagnet CrNb$_3$S$_6$ serves to illustrate our results, one of which is striking similarity of microrotations embedded in the micropolar elasticity theory with roton excitations in helium-4.

**General theory.**—We outline a general scheme of treatment of elementary excitations in the context of the micropolar elasticity theory. All technical details may be found in Supplemental Material[22]. In this theory, the field of translational displacements, $\mathbf{u}(\mathbf{r})$, is supplemented by the field of microrotations, $\varphi(\mathbf{r})$, and the both are attributed to a microelement located at the position $\mathbf{r}$ (see Fig. 1). Two linear strain tensors $\varepsilon_{kl} = \partial_l u_k - \varepsilon_{klm} \varphi_m$ (Cosserat’s deformation tensor) and $\gamma_{kl} = \partial_l \varphi_k$ (wryness tensor), form measures of microdeformations. Here and throughout, $\varepsilon_{klm}$ is the Levi-Civita symbol and Einstein summation convention is used.

The strain energy density is given by the quadratic form[10, 11], $U = \frac{1}{2} A_{klmn} \varepsilon_{klm} \varepsilon_{mn} + \frac{1}{2} B_{klmn} \gamma_{klm} \gamma_{mn} + C_{klmn} \varepsilon_{klm} \gamma_{mn}$. The four-rank tensors $A$ and $B$ are both polar tensors and exist irrespective of inversion symmetry. They possess the property $A_{klmn} = A_{mnkl}$ and $B_{klmn} = B_{mnkl}$, which has an internal symmetry $[(V^4)^2]$ in Jahn’s symbol[24]. The pseudotensor $C$ has an internal symmetry $V^4$ and exists only in crystals without inversion symmetry. It provides interaction between the polar displacements and the axial microrotations. This ‘translation-rotation’ coupling is the essential feature of chirality as has been highlighted in the Introduction. A number of independent elements of the $A_{klmn}$, $B_{klmn}$ and $C_{klmn}$ conditioned by permutations of the indices amounts to 45 and 81, respectively. A further account of the point group symmetry of a given crystal reduces their number. In addition, the tensor components must maintain positive definiteness of the internal energy, $U > 0$, for all strains to guarantee stability of the ground state against elementary excitations (phonons). Using the lattice theory[10], whereby atomic interactions
are modelled by springs, typical magnitudes of the tensor components amount at most to $A_{klmn} \sim 10^{10} \text{N} \cdot \text{m}^{-2}$, $B_{klmn} \sim 10^{-10} \text{N}$ and $C_{klmn} \sim 10^9 \text{N} \cdot \text{m}^{-1}$.

Variation of $U$ with respect to $\varepsilon_{kl}$ leads to the linear constitutive equation for the stress tensor, $t_{kl} = \delta U / \delta \varepsilon_{kl} = A_{klmn} \varepsilon_{mn} + C_{klmn} \gamma_{mn}$. Similarly, one obtain the couple stress tensor $m_{kl} = \delta U / \delta \gamma_{kl} = C_{mntk} \varepsilon_{mn} + B_{klmn} \gamma_{mn}$. Balance laws for momentum and momen-tum-of-momentum yield the equations of motion (EOM) for the translational microdisplacements, $\rho \ddot{u}_i = \partial_k t_{ki}$, and for the microrotations, $\rho j_{ik} \ddot{\varphi}_k = \partial_k m_{ik} + \epsilon_{jmn} t_{mn}$, respectively. Here, $\rho$ is the mass density and $j_{ik}$ is the microinertia tensor. Being written for the strain fields only, the EOM read as,

$$
\rho \ddot{u}_i = A_{klmn} \partial_k \partial_m u_n - \epsilon_{mn} A_{klmn} \partial_k \varphi_r + C_{klmn} \partial_k \partial_n \varphi_r \\
\rho j_{ik} \ddot{\varphi}_k = C_{klmn} \partial_k \partial_m u_n - \epsilon_{mn} C_{klmn} \partial_k \varphi_r + B_{klmn} \partial_k \partial_n \varphi_r \\
+ \epsilon_{jmn} A_{mnpl} \partial_p u_q - \epsilon_{mn} A_{mnpl} \partial_p \varphi_r - \epsilon_{mn} C_{mnpl} \partial_q \varphi_r.
$$

(1)

(2)

Assuming propagating plane wave solutions, Eqs. (1,2) provide desired dispersion relations.

**Chiral phonon dispersions for the point group 622.** To demonstrate how the chiral coupling gives rise to new peculiarities of phonon dispersion, we consider, as an example, the layered compound CrNb$_3$S$_6$ which has the non-centrosymmetric hexagonal space group $P6_322$. In this material, the Cr atoms are intercalated between the sandwich layers S-Nb-S of the disulfide NbS$_2$ and surrounded by the six S atoms in an octahedral geometry. The CrS$_6$ octahedra are not linked to each other. The distance $d$(Cr-S) $= 2.393 \text{Å}$ is less than $\sim 0.1 \text{Å}$ the distance $d$(Nb-S) $= 2.47 \pm 0.05 \text{Å}$.

Assuming that modes due to relative displacements of the Cr and S ions are not excited, the CrS$_6$ may be modeled as a rigid structural unit. The values $j_{11} = 0.5 \times 10^{-19} \text{m}^2$ and $j_{33} = 1.0 \times 10^{-19} \text{m}^2$ are used for estimations whenever it is necessary.

The mass density is taken as $\rho = 5.029 \text{ g/cm}^3$. Below, we consider plane-waves propagating along the a-crystalline [001] axis, i.e. the chiral axis.

Dispersion of the micropolar waves is obtained in the following way. To describe transverse modes, it is appropriate to introduce the circular basis, $u_\pm = u_1 \pm i u_2$, and $\varphi_\pm = \varphi_1 \mp i \varphi_2$, where $+/-$ corresponds to the left/right circularly polarized microdeformation fields. For the 622 point group, the polar tensors $A$, $B$ have 8 independent elements, while the axial one $C$ does 10 (see, for instance, Tables D.20 and D.22 in Ref. [24]). Then, the EOMs for the decoupled transverse modes may be written as,

$$
\rho \ddot{u}_\pm = A_{55} \partial_\pm^2 u_\pm + C_{74} \partial_\pm^2 \varphi_\pm \mp i (A_{47} - A_{55}) \partial_\pm \varphi_\pm,
$$

(3)

$$
\rho j_{ik} \ddot{\varphi}_k = C_{74} \partial_\pm^2 u_\pm \mp i (A_{47} - A_{55}) \partial_\pm u_\pm + B_{34} \partial_\pm^2 \varphi_\pm

+ 2i (C_{44} - C_{74}) \partial_\pm \varphi_\pm - (A_{44} - 2A_{47} + A_{55}) \varphi_\pm.
$$

(4)

where $j_+ = j_- = j_1$. Here, the tensor components are represented in the Voigt notation [24]. The most important point is the appearance of the linear gradient terms, $\partial_\pm u_\pm$ and $\partial_\pm \varphi_\pm$, which cause polarization-dependent velocities. Hybridization of these circularly polarized $u_\pm$ and $\varphi_\pm$-modes gives rise to acoustic and optical branches of the spectrum of propagating transverse waves in the micropolar medium. It is to be noted that each of the modes, $\varphi_+$ or $\varphi_-$, is non-reciprocal in real space, but they form a pair invariant under time-reversal symmetry. This may be inferred from Eqs. (3,4) which break parity $P$ but preserve time-reversal symmetry $T$. This situation means that phonons in the micropolar chiral crystal exhibit true chirality in contrast to the phonon [14, 20] or electrical [30] magneto-choiral effects, where both $P$ and $T$ are simultaneously broken. On that understanding, we call these excitations truly chiral phonons. This polarization dependent splitting of the transverse phonon branches is analogous to the Rashba splitting of electronic bands, in which $P$ is also broken but not $T$. Non-reciprocity of the truly chiral phonons originates from the $(C_{44} - C_{74})$-coupling term in the EOMs (3,4) and vanishes for $C_{44} = C_{74}$. This chiral coupling between microtranslations and microdeformations appears in the energy functional as $(C_{44} \varepsilon_{\alpha 3} + C_{74} \varepsilon_{3\alpha}) \gamma_{\alpha 3}$, $\alpha = 1, 2$.

Next, we focus our attention to the longitudinal branches. The equations that govern their propagation

![FIG. 2: Phonon dispersion curves for the chiral micropolar crystal: the longitudinal acoustic (LA) and optical (LO) branches (green dotted), the transverse left-handed acoustic (TA-left) and optical (TO-left) branches (blue solid), the transverse right-handed acoustic (TA-right) and optical (TO-right) branches (red solid). Numerical values of the tensor components are chosen as $A_{33} = 0.4 \cdot 10^{10} \text{ N/m}^2$, $A_{65} = 4.9 \cdot 10^{10} \text{ N/m}^2$, $A_{69} = 4.7 \cdot 10^{10} \text{ N/m}^2$, $B_{33} = 1.5 \cdot 10^{-10} \text{ N/cm}$, $C_{33} = 0.3 \text{ N/m}$ for the longitudinal modes and $A_{44} = 0.21 \cdot 10^{10} \text{ N/m}^2$, $A_{55} = 0.215 \cdot 10^{10} \text{ N/m}^2$, $A_{47} = 0.195 \cdot 10^{10} \text{ N/m}^2$, $B_{44} = 1.0 \cdot 10^{-10} \text{ N}$, $C_{44} = 0.44 \text{ N/m}$, $C_{74} = 0.36 \text{ N/m}$ for the transverse modes, respectively.](image)
are written as
\begin{align}
\rho \ddot{u}_3 &= A_{33} \partial^2_t u_3 + C_{33} \partial^2_k \varphi_3, \\
\rho \ddot{\varphi}_3 &= C_{33} \partial^2_t u_3 + B_{33} \partial^2_k \varphi_3 - 2(A_{66} - A_{69}) \varphi_3.
\end{align}

Note the absence of linear gradient terms that entails simple hybridization of the $u_3$ and $\varphi_3$ modes.

Phonon dispersion relations may be obtained inserting plane waves propagating along the chiral ($\Delta$) modes. The associated dispersion relations into Eqs. (3-6). The results are summarized in Fig. 2. There are six branches in total, namely, we have the longitudinal acoustic (LA)/optical (LO), the transverse left-handed acoustic (TA-left)/optical (TO-left), and the transverse right-handed acoustic (TA-right)/optical (TO-right) modes. The associated dispersion relations take the form
\begin{equation}
\left[ \omega^{(O/A)}_\alpha (k) \right]^2 = \frac{1}{2\rho j_\alpha} \left[ b_\alpha + j_\alpha a_\alpha \pm \sqrt{(b_\alpha - j_\alpha a_\alpha)^2 + 4j_\alpha \Delta^2_\alpha} \right].
\end{equation}

Here, the index $\alpha$ labels either the transverse-left/right (+/−) circular or longitudinal (“3”) polarization of the phonon branches. The upper and lower ± signs in the r.h.s. of Eq. (7) correspond to the optical (O) and acoustic (A) branches, respectively. The $k$-dependent parameters ($a_\alpha$, $b_\alpha$, and $\Delta_\alpha$) are given by $a_\pm = A_{55}k^2$, $b_\pm = B_{44}k^2 \mp 2(C_{44} - C_{74})k + (A_{44} + A_{55} - 2A_{47})$, $\Delta_\pm = C_{74}k^2 \mp (A_{47} - A_{55})k$, and $a_3 = A_{33}k^2$, $b_3 = B_{33}k^2 + 2(A_{66} - A_{69})$, $\Delta_3 = C_{33}k^2$. In the long wavelength limit, $k \to 0$, the frequencies of the acoustic branches are proportional to the wavenumber, while the frequencies of the optical branches tend to finite values, $\omega^{(O)}_\alpha (0) = \sqrt{(A_{44} + A_{55} - 2A_{47})/\rho j_\alpha}$ and $\omega^{(O)}_3 (0) = \sqrt{(A_{66} - A_{69})/\rho j_\alpha}$.

Similarity to roton spectrum. – Figure 3 contains only the TA-right handed phonon mode specially selected from all branches shown in Fig. 2. We observe that hybridization of the rotational and translational modes gives rise to the lowest phonon (TA-right or TA-left) branch which exhibits a “roton minimum” around $k_m \sim (C_{44} - C_{74})/B_{44}$, reminiscent of the excitation spectrum in superfluid $^4$He. The minimum occurs around a scale inversely proportional to the unit cell length and reflects hybridization of the rotational and translational degrees of freedom of the microelement $\text{CrS}_6$.

To elucidate reasons behind an appearance of the roton-like minimum, we trace how translational and rotational modes contribute to the hybridized excitations. We find weights of these modes specified by the dimensionless coefficients $c_{u,\alpha} = u_\alpha/\sqrt{j_\alpha}$ and $c_{\varphi,\alpha} = \varphi_\alpha$, whose explicit expressions are reproduced in Supplemental Material. Their $k$-dependence is illustrated by Fig. 4 for the TA-right and LA branches. It is evident that the $u_+\varphi_+$ hybridization causes resonant enhancement of the rotational degrees of freedom ($\varphi_+$) when approaching the roton minimum. In contrast, the ratio between the coefficients $c_{u,3}$ and $c_{\varphi,3}$ is reversed near the crossing of the corresponding dispersion curves, which are modified into the hybridized LA and LO phonon modes.

There is actually an interesting parallel between the TA chiral phonons and roton excitations in superfluid $^4$He. Originally, the rotons had been interpreted as a signature of some local vorticity. According to Feynman’s view, a maximum in the static form factor $S_q$, which signals a short range crystalline order, gives a roton minimum in the excitation spectrum through the relation $\omega_q = q^2/2mS_q$. Later, Nozières proposed an alternate scenario, where the rotons should be viewed as an incipient soft mode associated with a crystallization instability. According to this view, the Bogoliubov quasiparticles hybridize with soft density fluctuations to cause a resultant roton minimum. The picture can be formulated by assuming two coupled excitations where the first correspond to the Bogoliubov quasiparticles with the spectrum $\varepsilon^B_q = \left[ \xi^2_q + 2\xi_q N_0 U \right]^{1/2}$ and the second do to the density fluctuation mode, which is characterized by the single mode frequency $\Omega_q$. Here, $\xi_q$ is the
boson kinetic energy, $N_0$ is a condensate fraction, and $U$ is a direct repulsion between bosons. The spectrum of the hybridized quasiparticles has the form similar to Eq. (7),

$$E_q^2 = \frac{1}{2} \left( \Omega_q^2 + (\varepsilon_q^B)^2 \right) \pm \frac{1}{2} \left[ \left( \Omega_q^2 - (\varepsilon_q^B)^2 \right)^2 + 32 \Omega_q \alpha_q \right]^{1/2}, \quad (8)$$

where $\alpha_q$ is a strength of coupling between the bosons and density fluctuations. Direct comparison between Eqs. (7) and (8) eloquently illustrates the one-to-one correspondence between the translational displacement, $u(r)$, and the microrotation, $\varphi(r)$, on one hand and the Bogoliubov quasiparticles and density fluctuations on the other $[a_\pm(k)$ and $b_\pm(k)$ respectively correspond to $\varepsilon_q^B$ and $\Omega_q]$.

However, the roton minimum of the superfluid helium arises in the spectrum of the longitudinal sound wave of the normal component, while in the micropolar crystal this effect is observed in propagation of the transversal elastic waves, but not for the longitudinal ones. This difference may materialize arguments propounded by Landau and Feynman that the rotons are related to local vorticity [31]. The micro-rotation does warrant the name roton as spinning motion of the microelement.

There is further similarity between the rotons in $^4$He and the chiral phonons. The boson condensate lowers the roton minimum but the latter will remain finite due to the effect of condensate depletion [32, 33]. For the chiral phonons, a finite value of the roton minimum may be inferred from the requirement that the crystal must be stable against propagating elastic waves. Indeed, the roton minimum appears owing to the coupled translation-rotation fluctuations, whose contribution to the energy $U$ amounts to $\frac{1}{2} \sum_{\alpha=1,2,3} \eta_\alpha^T \hat{M} \eta_\alpha > 0$ with

$$\hat{M} = \begin{pmatrix} A_{44} & A_{47} & C_{44} \\ A_{47} & A_{55} & C_{74} \\ C_{44} & C_{74} & B_{44} \end{pmatrix}, \quad (9)$$

and $\eta_\alpha^T = (\varepsilon_{\alpha 3}, \varepsilon_{3\alpha}, \gamma_{\alpha 3})$. The stability condition (positive definiteness of the energy functional) imposes restrictions on the allowed components of the $A$, $B$, and $C$ tensors, in particular $A_{55} > 0$ and $\det \hat{M} > 0$. On the other hand, from the excitation spectrum [7] it can be determined that a disruptive effect of the soft mode instability is absent provided $A_{355} \det \hat{M} > 0$ [24]. This implies that the roton minimum will remain finite (never touch zero). An area of stability within the $C_{44}$-$C_{74}$ plane (these constants responsible for parity breaking) is plotted in Fig. 5 together with a region where the roton minimum emerges.

**Phonon angular momentum.**—Once the spectrum of the micropolar waves is known, one is capable to determine naturally orbital and spin parts of the angular momentum associated with elastic deformations. By specifying the basis of the left-handed $(L)$ and right-handed $(R)$ circularly polarized transverse modes as $|R_a\rangle = |L_a\rangle^* = \frac{1}{\sqrt{2}} (1, i, 0, 0, 0, 0)^T$, and $|R_\varphi\rangle = |L_\varphi\rangle^* = \frac{1}{\sqrt{2}} (0, 0, 0, 1, i, 0)^T$ the solution $|v\rangle = (c_{a1}, c_{a2}, c_{a3}, c_{\varphi1}, c_{\varphi2}, c_{\varphi3})$ of Eqs. (30) may be decomposed as $|v\rangle = \sum_{\alpha=u,\varphi} (R_\alpha |v\rangle |R_\alpha\rangle + \langle L_\alpha |v\rangle |L_\alpha\rangle)$. Then, the generator of rotations of the phonon polarization plane around the $z$-axis, or phonon spin [21], is

$$\hat{S}^z = h \sum_{\alpha=u,\varphi} (|R_\alpha\rangle \langle L_\alpha| - |L_\alpha\rangle \langle R_\alpha|),$$

so that

$$\langle v|\hat{S}^z|v\rangle = \frac{h}{2} \sum_{f=u,\varphi} \left( \langle c_{f,-}|^2 - \langle c_{f,+}|^2 \right) \quad (10)$$

is fulfilled. Obviously, the phonon circular polarization is quantized, i.e. it can take only the $\pm h$ values for the “$+$”-branches, respectively. Unlike chiral phonons in the theories [21, 22], the phonon spin is introduced for any $k$-point of the Brillouin zone. Neglecting the microrotations $\varphi$, the definition coincides with the spin part of the phonon angular momentum given in Ref. [34].

$$L_s = h \sum_k \epsilon_k \left( a_{k-}^\dagger a_{k+} - a_{k+}^\dagger a_{k-} \right),$$

where the angular momenta $h$ of the individual $(k\pm)$-phonons add up either parallel or anti-parallel to their wave vectors, $\epsilon_k = k/|k|$. Classical interpretation of the resultant $L_s$ corresponds to small-radius circular shear displacements of points around their equilibrium positions [Fig. 1(b)].

With regard to the orbital part of the phonon angular momentum, its definition, $L_o = \rho j_{\pm} \varphi_{\pm}$, follows directly from the appropriate conservation law of the micropolar theory [10]. This implies propagation of plane waves of the angular momentum density.
Roton and acoustic activity.—Nonreciprocity of the chiral phonons results in consequential acoustic activity, most notably in the vicinity of the roton minimum. By definition, the effect occurs when incident transverse acoustic waves, which are linearly polarized, propagate in the crystal along the $z$-axis. It is characterized by the rotation angle, $\phi = \frac{1}{2} \omega (1/v_+ - 1/v_-)$, at a distance $l$ from the incident surface. The phase velocities $v_\pm$ of the circularly polarized transverse modes may be calculated from the dispersion relations (7) to give

$$\frac{\phi}{l} = \rho \omega^3 \frac{(A_{47} - A_{55})}{(A_{44}A_{55} - A_{47}^2)} \times [A_{55}C_{44} + A_{44}C_{74} - A_{47} (C_{44} + C_{74})] .$$

Comparison with EOMs [8] shows that apart from the intercross coupling $A_{55} - A_{47}$ between the micro-translations and microrotations, the inherent gyrotropy $C_{44} - C_{74}$ of the microrotations is an essential element of acoustic activity. Unlike the micropolar theory, the conventional approach [5, 6] relates the latter to non-local interaction between stress and strain; this is reflected in the first-order dispersion in expansion of the elastic coefficients $c_{ij}(k, \omega)$. As a consequence, a difference in phase velocities of circularly polarized waves appears at any $\mathbf{k}$ vector.

Concluding remarks.—We demonstrate polarization-dependent splitting of phonon bands in a chiral crystal, using the micropolar elasticity theory for CrNb$_3$S$_6$. Our main results may be summarized as follows. (I) The splitting is reached solely within the phonon sector of elementary excitations, and it is maintained by coupling between the transverse translational and rotational modes of the micropolar medium. (II) Transverse acoustic branches of the hybridized phonon spectrum exhibit a "roton" minimum reminiscent of elementary excitations in the superfluid helium-4. We argue that the translational and rotational degrees of freedom of the chiral phonon system correspond to the Bogoliubov quasiparticles and massive density fluctuations, respectively, in the superfluid helium-4. In addition, we discuss acoustic activity originated from nonreciprocity of the phonon spectrum.

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GENERAL FORMULATION

In this Supplementary Material we present technical details related to the micropolar elasticity theory. In Sec. I, we summarize a general formulation. In Sec. II, a concrete form of equations of motion is derived for the 622 point group. In Sec. III, the stability condition and a finite gap of the roton minimum are discussed.

The constitutive tensors obey the symmetry relations,

\[ \epsilon_{ijkl} = -\epsilon_{jikl}, \]

and the wryness (microrotation) tensor

\[ \gamma_{ijkl} = \partial_i \varphi_k, \]

where \( \epsilon_{ijkl} \) is the totally antisymmetric tensor (Levi-Civita symbol) and the Einstein summation convention is used.

The strain energy density related to these microdeformations is given by

\[ U = \frac{1}{2} A_{klmn} \epsilon_{kl} \epsilon_{mn} + \frac{1}{2} B_{klmn} \gamma_{kl} \gamma_{mn} + C_{klmn} \epsilon_{kl} \gamma_{mn}. \]

The constitutive tensors obey the symmetry relations, \( A_{klmn} = A_{mnkl} \) and \( B_{klmn} = B_{mnkl} \), and possess the internal symmetry \( (|V|^2)^2 \) in the notation of the Jahn’s symbols.

Inasmuch as the microrotation vector \( \varphi \) is an axial vector and the wryness tensor \( \gamma_{kl} \) is an axial tensor, the \( C_{klmn} \) is the forth-rank pseudotensor with the internal symmetry \( V^4 \).

Here, we briefly mention point group symmetry transformations described by a matrix \( \hat{G} = (g_{ij}) \) with an orthogonal basis. The polar tensor \( P \) of the rank \( r \) obeys

\[ P_{ijkl...} = g_{ij} g_{gm} g_{kn} ... P_{lmn...}. \]

The inversion operation, \( g_{11} = g_{22} = g_{33} = -1 \), plays a special role and gives

\[ P_{ijkl...} = (-1)^r P_{lmn...}. \]

Therefore, \( P \) possesses inversion symmetry for an even \( r \).

Similarly, if \( Q \) is an axial tensor of the rank \( r \), then, under the inversion operation,

\[ Q_{ijk...} = (\det \hat{G}) (-1)^r Q_{lmn...} = (-1)^{r+1} Q_{lmn...}. \]

Therefore, \( Q \) possesses inversion symmetry for an odd \( r \).

Taking variation of \( U \) with respect to \( \epsilon_{kl} \), we have the constitutive equation,

\[ t_{kl} = \frac{\partial U}{\partial \epsilon_{kl}} = A_{klmn} \epsilon_{mn} + C_{klmn} \gamma_{mn}, \]

where \( t_{kl} \) is the stress tensor.

Similarly, one obtains the couple stress tensor

\[ m_{kl} = \frac{\partial U}{\partial \gamma_{lk}} = B_{lkmn} \gamma_{mn} + C_{mnkl} \epsilon_{mn}. \]

Now consider an elastic body occupying a region of volume \( V \) bounded by the body surface \( \partial V \). The torque exerted on an element of the surface area reads a

\[ dS = \hat{n} dS \]

with the exterior normal \( \hat{n} \) to \( \partial V \).

The total torque acting on this surface microelement includes the orbital torque, \( \mathbf{x} \times \mathbf{T} \), and the spin torque, \( \mathbf{M} \), where \( \mathbf{T} \) denotes a force acting on \( dS \). Then, we have

\[ (\mathbf{x} \times \mathbf{T} + \mathbf{M}) \alpha dS = \epsilon_{\alpha\beta\gamma} x_{\beta} T_{\gamma} dS + M_{\alpha} dS. \]

The vectors \( \mathbf{T} \) and \( \mathbf{M} \) may be rewritten via the force stress and coupled stress tensors as

\[ T_{\gamma} = t_{\delta\gamma} \hat{n}_{\delta}, \quad M_{\alpha} = m_{\delta\alpha} \hat{n}_{\delta}. \]

We shall restrict our discussion to a case when there is neither external volume force nor volume torque. Then, the rate of change of the angular momentum results from the orbital and spin torques acting on the microelement surface. This consideration leads to the balance equation,

\[ \frac{d}{dt} \int_{\partial V} \left( \epsilon_{\alpha\beta\gamma} x_{\beta} T_{\gamma} + \epsilon_{\alpha\beta\gamma} \hat{\varphi}_{\beta} \right) n_{\delta} dS = \int_{\partial V} \left( \epsilon_{\alpha\beta\gamma} x_{\beta} \hat{\varphi}_{\gamma} + m_{\delta\alpha} \hat{n}_{\delta} \right) n_{\delta} dS. \]
where $\rho$ is the mass density and $j_{\alpha\beta}$ is the microinertia tensor.

Applying the Gauss theorem to the r.h.s. of Eq. (12) one obtains

$$ \int_V \rho (\epsilon_{\alpha\beta\gamma} x_\beta \dot{v}_\gamma + j_{\alpha\beta} \ddot{\varphi}_\beta) \, dV = \int_V \partial_\delta (\epsilon_{\alpha\beta\gamma} x_\beta \delta_\gamma + m_{\delta\alpha}) \, dV, \quad (13) $$

what may be converted to

$$ \int_V \epsilon_{\alpha\beta\gamma} x_\beta (\rho \dot{\delta}_\gamma - \delta_\beta \delta_\gamma) \, dV = - \int_V (\rho j_{\alpha\beta\gamma} \ddot{\varphi}_\beta - \epsilon_{\alpha\beta\gamma} x_\beta - \delta_\beta \delta_\gamma) \, dV. \quad (14) $$

Since $V$ and $x_\beta$ are chosen arbitrarily, the integrands of both sides in Eq. (14) must be zero. This immediately yields the equations of motion

$$ \rho \dddot{u}_t = \partial_k t_{kt}, \quad (15) $$

$$ \rho j_{ik\gamma} \ddot{\varphi}_k = \partial_k m_{kt} + \epsilon_{lmn} m_{mn}, \quad (16) $$

for the microdisplacements and microrotations, respectively.

Inserting Eqs. (7,8) into Eqs. (15,16) we get

$$ \rho \dddot{u}_t = A_{klmn} \partial_k \partial_m u_n - \epsilon_{mn} A_{klmn} \partial_k \varphi_r + C_{klmn} \partial_k \partial_n \varphi_r, \quad (17) $$

and

$$ \rho j_{ik\gamma} \dddot{\varphi}_k = C_{mnik} \partial_k \partial_m u_n - \epsilon_{mn} C_{mnik} \partial_k \varphi_r + B_{mnik} \partial_k \partial_m \varphi_r + \epsilon_{mn} A_{mnipq} \partial_{pq} \varphi_r + \epsilon_{mn} C_{mnipq} \partial_{pq} \varphi_r. \quad (18) $$

These two equations constitute the basis of the linear micropolar elasticity theory and may be applicable to any micropolar medium.

Furthermore, we focus on the phonon modes propagating along the chiral axis, namely, the ‘$3$’-axis. It means that spatial derivatives in the above equations are taken with respect to this coordinate only. Consequently, Eqs. (14,18) can be recast in the form

$$ \rho \dddot{u}_1 = A_{313n} \partial_3^2 u_n - \epsilon_{mn} A_{31mn} \partial_3 \varphi_r + C_{31mn} \partial_3^2 \varphi_r, \quad (19) $$

$$ \rho \dddot{u}_2 = A_{323n} \partial_3^2 u_n - \epsilon_{mn} A_{32mn} \partial_3 \varphi_r + C_{32mn} \partial_3^2 \varphi_r, \quad (20) $$

$$ \rho \dddot{u}_3 = A_{333n} \partial_3^2 u_n - \epsilon_{mn} A_{33mn} \partial_3 \varphi_r + C_{33mn} \partial_3^2 \varphi_r, \quad (21) $$

$$ \rho j_{1k\gamma} \ddot{\varphi}_k = C_{3n13} \partial_3^2 u_n - \epsilon_{mn} C_{mn13} \partial_3 \varphi_r + B_{3n13} \partial_3^2 \varphi_r + \epsilon_{mn} A_{mn3q} \partial_3 \varphi_r - \epsilon_{mn} A_{mpq} \partial_{pq} \varphi_r + \epsilon_{mn} C_{mpq} \partial_{pq} \varphi_r, \quad (22) $$

$$ \rho j_{2k\gamma} \ddot{\varphi}_k = C_{3n23} \partial_3^2 u_n - \epsilon_{mn} C_{mn23} \partial_3 \varphi_r + B_{3n23} \partial_3^2 \varphi_r + \epsilon_{mn} A_{mn3q} \partial_3 \varphi_r - \epsilon_{mn} A_{mpq} \partial_{pq} \varphi_r + \epsilon_{mn} C_{mpq} \partial_{pq} \varphi_r, \quad (23) $$

$$ \rho j_{3k\gamma} \ddot{\varphi}_k = C_{3n33} \partial_3^2 u_n - \epsilon_{mn} C_{mn33} \partial_3 \varphi_r + B_{3n33} \partial_3^2 \varphi_r + \epsilon_{mn} A_{mn3q} \partial_3 \varphi_r - 3 \epsilon_{mn} A_{mpq} \partial_{pq} \varphi_r + \epsilon_{mn} C_{mpq} \partial_{pq} \varphi_r. \quad (24) $$

### THE POINT GROUP 622

**Tensor components**

The point group symmetry of a given crystal imposes severe restrictions on the tensors $A$, $B$ and $C$. For example, in the case of the 622 point group, Table 20 of Ref. [9] gives $A$ and $B$ in the form summarized in Table I. Apparently, there are only 8 independent elements: $A_{12}$, $A_{13}$, $A_{33}$, $A_{44}$, $A_{55}$, $A_{66}$, $A_{47}$, and $A_{69}$.

**Table I:** Nonzero tensor components of $A$ and $B$ for the point group 622. There is the constraint $A_{11} = A_{12} + A_{66} + A_{69}$.

| 11 | 22 | 33 | 23 | 31 | 12 | 32 | 13 | 21 |
|----|----|----|----|----|----|----|----|----|
| 11 | $A_{11}$ | $A_{12}$ | $A_{13}$ | 0 | 0 | 0 | 0 | 0 |
| 22 | $A_{12}$ | $A_{11}$ | $A_{13}$ | 0 | 0 | 0 | 0 | 0 |
| 33 | $A_{13}$ | $A_{13}$ | $A_{33}$ | 0 | 0 | 0 | 0 | 0 |
| 23 | 0 | 0 | 0 | $A_{44}$ | 0 | 0 | $A_{47}$ | 0 |
| 31 | 0 | 0 | 0 | 0 | $A_{55}$ | 0 | 0 | $A_{47}$ |
| 12 | 0 | 0 | 0 | 0 | 0 | $A_{66}$ | 0 | 0 |
| 32 | 0 | 0 | 0 | $A_{66}$ | 0 | 0 | $A_{55}$ | 0 |
| 13 | 0 | 0 | 0 | 0 | $A_{47}$ | 0 | 0 | $A_{44}$ |
| 21 | 0 | 0 | 0 | 0 | 0 | 0 | $A_{47}$ | 0 |

Table 22 of Ref. [9] gives $C$ in the form summarized in Table II. It is readily seen that a number of independent elements equals to 10: $C_{12}$, $C_{13}$, $C_{31}$, $C_{33}$, $C_{44}$, $C_{47}$, $C_{55}$, $C_{74}$, $C_{66}$, $C_{69}$.
PHONON DISPERSIONS

Transverse branches

Making use data from Tables I and II an explicit form of the equations of motion comes from Eqs. (17, 18). The microinertia tensor can be written in the diagonal form \( j = \text{diag}(j_1, j_1, j_3) \) by taking into account the almost 3\( m \) (\( D_{3d} \)) point group symmetry of the CrS\(_6\) block. This leads to

\[
\rho \ddot{u}_1 = A_{55} \partial^2_u u_1 - (A_{55} - A_{47}) \partial_3 \varphi_2 + C_{74} \partial^2_3 \varphi_1, \quad (25)
\]

\[
\rho \ddot{u}_2 = A_{55} \partial^2_u u_2 - (A_{47} - A_{55}) \partial_3 \varphi_1 + C_{74} \partial^2_3 \varphi_2, \quad (26)
\]

\[
\rho \ddot{u}_3 = A_{33} \partial^2_u u_3 + C_{33} \partial^2_3 \varphi_3, \quad (27)
\]

\[
\rho j_1 \dot{\varphi}_1 = C_{74} \partial^2_3 u_1 + 2 (C_{44} - C_{74}) \partial_3 \varphi_2 + B_{44} \partial^2_3 \varphi_1 + (A_{47} - A_{55}) \partial_3 u_2 - (A_{44} - 2 A_{47} + A_{55}) \varphi_1, \quad (28)
\]

\[
\rho j_1 \dot{\varphi}_2 = C_{74} \partial^2_3 u_2 - 2 (C_{44} - C_{74}) \partial_3 \varphi_1 + B_{44} \partial^2_3 \varphi_2 - (A_{47} - A_{55}) \partial_3 u_1 - (A_{44} - 2 A_{47} + A_{55}) \varphi_2, \quad (29)
\]

\[
\rho j_3 \dot{\varphi}_3 = C_{33} \partial^2_3 u_3 + B_{33} \partial^2_3 \varphi_3 - 2 (A_{66} - A_{69}) \varphi_3. \quad (30)
\]

To simplify a solution of the equations of motion, the linearly polarized fields are related by

\[
u \pm = u_1 \pm i u_2, \quad (31)
\]

\[
\varphi \pm = \varphi_1 \pm i \varphi_2, \quad (32)
\]

that are circularly polarized and counterrotating in the (1, 2) plane. The sign (+) corresponds to the left (right) circular polarization.

In the chiral basis the EOMs for the transverse modes are

\[
\rho \ddot{u}_\pm = A_{55} \partial^2 u_\pm + C_{74} \partial^2_3 \varphi_\pm + i (A_{47} - A_{55}) \partial_3 \varphi_\pm, \quad (33)
\]

\[
\rho j_\pm \dot{\varphi}_\pm = C_{74} \partial^2_3 u_\pm \mp i (A_{47} - A_{55}) \partial_3 u_\pm + B_{44} \partial^2_3 \varphi_\pm \mp 2i (C_{44} - C_{74}) \partial_3 \varphi_\pm - (A_{44} - 2 A_{47} + A_{55}) \varphi_\pm, \quad (34)
\]

where \( j_+ = j_1 = j_3 \).

To get a dispersion relation, we use the plane-wave solutions

\[
u_\alpha(x_3, t) = U_\alpha e^{-i(\omega t - k x_3)}, \quad \varphi_\alpha(x_3, t) = \Phi_\alpha e^{-i(\omega t - k x_3)}, \quad (35)
\]

where the index \( \alpha \) labels +, − and 3.

The EOMs for the transverse modes \( (\alpha = \pm) \) are written in the matrix form as

\[
\begin{pmatrix}
\rho \omega_\alpha^2 - a_\alpha & -\Delta_\alpha \\
-\Delta_\alpha & \rho j_\alpha \omega_\alpha^2 - b_\alpha
\end{pmatrix}
\begin{pmatrix}
U_\alpha \\
\Phi_\alpha
\end{pmatrix} = 0. \quad (36)
\]

Here, the \( k \)-dependent coefficients are introduced

\[
a_\pm = A_{55} k^2, \quad (37)
\]

\[
b_\pm = B_{44} k^2 + 2 (C_{44} - C_{74}) k
\]

\[
+ (A_{44} + A_{55} - 2 A_{47}), \quad (38)
\]

\[
\Delta_\pm = C_{74} k^2 \pm (A_{55} - A_{47}) k. \quad (39)
\]

It is immediately recognized that the crystal has two branches for each polarization \( \alpha \):

(I) the acoustic branch

\[
\omega_\alpha^{(A)}(k)^2 = \frac{1}{2 \rho j_\alpha} \left[ b_\alpha + j_\alpha a_\alpha - \sqrt{(b_\alpha - j_\alpha a_\alpha)^2 + 4 j_\alpha \Delta_\alpha^2} \right]; \quad (40)
\]

(II) the optical branch

\[
\omega_\alpha^{(O)}(k)^2 = \frac{1}{2 \rho j_\alpha} \left[ b_\alpha + j_\alpha a_\alpha + \sqrt{(b_\alpha - j_\alpha a_\alpha)^2 + 4 j_\alpha \Delta_\alpha^2} \right]. \quad (41)
\]

The acoustic branch starts off at zero frequency, while the optical branch starts off at some finite value

\[
\omega_\pm^{(A)}(0)^2 = 0, \quad \omega_\pm^{(O)}(0)^2 = \frac{1}{2 \rho j_\pm} (A_{44} + A_{55} - 2 A_{47}). \quad (42)
\]

Longitudinal branches

It is to be noted that the transverse and longitudinal modes are totally decouple. The EOMs for the longitudinal modes read

\[
\rho \ddot{u}_3 = A_{33} \partial^2 u_3 + C_{33} \partial^2_3 \varphi_3, \quad (43)
\]

\[
\rho j_3 \dot{\varphi}_3 = C_{33} \partial^2_3 u_3 + B_{33} \partial^2_3 \varphi_3 - 2 (A_{66} - A_{69}) \varphi_3. \quad (44)
\]

By repeating the procedure presented above one obtains the acoustic and optical branches given by Eqs. (40, 41), where \( \alpha = 3 \) and

\[
a_3 = A_{33} k^2, \quad (45)
\]

\[
b_3 = B_{33} k^2 + 2 (A_{66} - A_{69}), \quad (46)
\]

\[
\Delta_3 = C_{33} k^2. \quad (47)
\]

At \( k = 0 \) we have

\[
\omega_3^{(A)}(0)^2 = 0, \quad \omega_3^{(O)}(0)^2 = \frac{2}{\rho j_3} (A_{66} - A_{69}). \quad (48)
\]
Weights of the transverse $u$ and $\varphi$ modes

The EOMs (36) can be recast in the equivalent form

\[
\begin{pmatrix}
j_{\alpha}^{1/2}(\rho\omega_{\alpha}^2 - a_{\alpha}) & -\Delta_{\alpha} \\
-j_{\alpha}^{1/2}\Delta_{\alpha} & (\rho j_{\alpha}\omega_{\alpha}^2 - b_{\alpha})
\end{pmatrix}
\times
\begin{pmatrix}
j_{\alpha}^{-1/2}U_{\alpha} \\
\Phi_{\alpha}
\end{pmatrix} = 0,
\]

(49)

where both $j_{\alpha}^{-1/2}U_{\alpha}$ and $\Phi_{\alpha}$ are dimensionless, since $j_{\alpha}^{1/2}$ has a length scale of order of the microelement size.

Inserting in this equation the relation

\[
\rho\omega_{\alpha}^2 - a_{\alpha} = \frac{1}{2j_{\alpha}} \left[b_{\alpha} - j_{\alpha}a_{\alpha} \mp \sqrt{(b_{\alpha} - j_{\alpha}a_{\alpha})^2 + 4j_{\alpha}\Delta_{\alpha}^2}\right],
\]

we obtain

\[
j_{\alpha}^{-1/2}|U_{\alpha}| = \left[\frac{1}{2} + \frac{j_{\alpha}(a_{\alpha} - \rho\omega_{\alpha}^2)^2}{2\Delta_{\alpha}^2}\right]^{-1/2}
= \left[\frac{1}{2} + \frac{\sqrt{(b_{\alpha} - j_{\alpha}a_{\alpha})^2 + 4j_{\alpha}\Delta_{\alpha}^2}}{8j_{\alpha}\Delta_{\alpha}^2}\right]^{-1/2},
\]

(51)

where the upper/lower sign corresponds to the acoustic/optical branch.

Analogously, we get

\[
|\Phi_{\alpha}| = \left[\frac{1}{2} + \frac{\Delta_{\alpha}^2}{2j_{\alpha}(a_{\alpha} - \rho\omega_{\alpha}^2)^2}\right]^{-1/2}
= \left[\frac{1}{2} + \frac{2j_{\alpha}\Delta_{\alpha}^2}{(b_{\alpha} - j_{\alpha}a_{\alpha} \mp \sqrt{(b_{\alpha} - j_{\alpha}a_{\alpha})^2 + 4j_{\alpha}\Delta_{\alpha}^2})^2}\right]^{-1/2},
\]

(52)

where the normalization condition

\[
\frac{j_{\alpha}^{-1/2}U_{\alpha}}{\sqrt{2}}^2 + |\Phi_{\alpha}|^2 = 1
\]

(53)

is accounted for.

In Fig. S1 we show the dispersion curves of the TA-right branch and corresponding weights. It is clearly seen that the roton minimum is correlated to the depletion of the $u+$ mode and complementary evolution of the $\varphi+$ mode.

![FIG. S1: Phonon dispersion curves for the chiral micropolar crystal: the longitudinal acoustic (LA) and optical (LO) branches (green dotted), the transverse left-handed acoustic (TA-left) and optical (TO-left) branches (blue solid), the transverse right-handed acoustic (TA-right) and optical (TO-right) branches (red solid). The wavenumber dependence of the weights of translational and rotational modes in the hybridized TA-right (red solid) and LA (green dotted) branches. $A_{44} = 0.2 - 10^{-15}$ N/m$^2$, $A_{55} = 0.2 - 10^{-15}$ N/m$^2$, $A_{47} = 0.11 - 10^{-10}$ N/m$^3$, $B_{44} = 0.92115 - 10^{-10}$ N, and $C_{74} = 0.35N/m$. $C_{44} = 0.35$, (2) 0.36, (3) 0.37, (4) 0.38, (5) 0.39, (6) 0.398, and (7) 0.40 in the unit of N/m.](image)

**STABILITY CONDITION AND ROTON MINIMUM**

Below we formulate the condition of ground state stability against phonon excitations. For this purpose, we write down the energy (48) by retaining only those tensor components which appear in the equations of motion for
the stability condition reads
\[ U = \frac{1}{2} (A_{66} + A_{69}) (\varepsilon_1^4 + \varepsilon_2^4) + \frac{1}{2} A_{66} (\varepsilon_1^2 + \varepsilon_2^2) + A_{69} \varepsilon_1 \varepsilon_2 + \frac{1}{2} A_{33} \varepsilon_3^2 + C_{33} \varepsilon_3 \gamma_3 + \frac{1}{2} B_{33} \gamma_3^2 \\
+ \frac{1}{2} A_{44} (\varepsilon_2^2 + \varepsilon_3^2) + A_{47} (\varepsilon_2 \varepsilon_3 + \varepsilon_3 \varepsilon_1) + C_{44} (\varepsilon_2 \gamma_3 + \varepsilon_3 \gamma_1) + C_{44} (\varepsilon_2 \gamma_3 + \varepsilon_3 \gamma_1) + C_{44} (\varepsilon_2 \gamma_3 + \varepsilon_3 \gamma_1) \\
+ \frac{1}{2} B_{44} (\gamma_2^2 + \gamma_3^2), \] (54)

where we used
\[ A_{11} = A_{66} + A_{69}, \quad C_{11} = C_{66} + C_{69}. \] (55)

Introducing the vectors
\[ \eta_1 = (\varepsilon_1, \varepsilon_2)^T, \quad \eta_2 = (\varepsilon_1, \varepsilon_2)^T, \quad \eta_3 = (\varepsilon_3, \gamma_3)^T, \quad \eta_4 = (\varepsilon_2, \varepsilon_3, \gamma_2)^T, \quad \eta_5 = (\varepsilon_3, \varepsilon_1, \gamma_1)^T, \] (56)

the stability condition reads
\[ U = \frac{1}{2} \sum_{i=1}^{5} \eta_i^T \tilde{M} \eta_i > 0, \] (57)

where the matrices are
\[ \tilde{M}_1 = \begin{pmatrix} A_{66} + A_{69} & 0 \\ 0 & A_{66} + A_{69} \end{pmatrix}, \] (58)
\[ \tilde{M}_2 = \begin{pmatrix} A_{66} & A_{69} \\ A_{69} & A_{66} \end{pmatrix}, \] (59)
\[ \tilde{M}_3 = \begin{pmatrix} A_{33} & C_{33} \\ C_{33} & B_{33} \end{pmatrix}, \] (60)
\[ \tilde{M}_4 = \tilde{M}_5 = \tilde{M} = \begin{pmatrix} A_{44} & A_{47} & C_{44} \\ A_{47} & A_{55} & C_{47} \\ C_{44} & C_{47} & B_{44} \end{pmatrix}. \] (61)

This condition ensures that all eigenvalues of the \( \tilde{M} \)-matrices are positive. The terms that contain \( \tilde{M}_4 = \tilde{M}_5 = \tilde{M} \) are of particular importance. As will be seen below, it is because the requirement
\[ \frac{1}{2} \sum_{\alpha=1,2} \eta_\alpha^T \tilde{M} \eta_\alpha > 0 \] (62)

with \( \eta_\alpha^T = (\varepsilon_{\alpha 3}, \varepsilon_{3 \alpha}, \gamma_{\alpha 3}) \), the roton minimum never reaches zero.

**Impossibility of zeroing of the roton minimum**

From the dispersion relation (40) it can be readily established that the roton minimum touches zero provided \( a_\alpha b_\alpha = \Delta_\alpha^2 \), where \( \alpha = \pm \). This condition maintains
\[ (A_{55} B_{44} - C_{44}^2) k^2 \mp 2 (A_{55} C_{44} - A_{47} C_{74}) k + A_{44} A_{55} - A_{47}^2 = 0. \] (63)

This equation has no nontrivial solutions for \( k \) if its discriminant \( D \) is negative. It is straightforward to show that
\[ D/4 = - A_{55} \det \tilde{M} < 0. \] (64)

Now remains only to note that both \( A_{55} \) and \( \det \tilde{M} \) are positive due to the stability condition (62) and, as such, a "roton" instability of the crystalline order never occurs.

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