Triangular Pair Density Wave in Confined Superfluid ³He

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Recent advances in experiment and theory suggest that superfluid ³He under planar confinement may form a pair density wave (PDW) whereby superfluid and crystalline orders coexist. While a natural candidate for this phase is a unidirectional stripe phase predicted by Vorontsov and Sauls in 2007, recent nuclear magnetic resonance measurements of the superfluid order parameter rather suggest a twodimensional PDW with noncollinear wave vectors, of possibly square or hexagonal symmetry. In this Letter, we present a general mechanism by which a PDW with the symmetry of a triangular lattice can be stabilized, based on a superfluid generalization of Landau's theory of the liquid-solid transition. A softmode instability at a finite wave vector within the translationally invariant planar-distorted B phase triggers a transition from uniform superfluid to PDW that is first order due to a cubic term generally present in the PDW free-energy functional. This cubic term also lifts the degeneracy of possible PDW states in favor of those for which wave vectors add to zero in triangles, which in two dimensions uniquely selects the triangular lattice.

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Introduction.—³He arguably best epitomizes the paradigm of emergence in condensed matter physics. Atomically one of the simplest isotopes in the periodic table of elements, ³He, nonetheless, gives rise to a rich variety of paired superfluid phases at low temperatures. In bulk and absent magnetic fields, only two superfluid phases are thermodynamically stable [1]: the B phase with isotropic quasiparticle gap appears at zero pressure, and an additional A phase with nodal quasiparticles is stabilized at high pressure. In contrast to ⁴He atoms or *s*-wave Cooper pairs, the *p*-wave Cooper pairs in ³He couple strongly to geometric perturbations on account of their spatial anisotropy, leading to the possibility of new superfluid phases under confinement.

While superfluid ³He in planar confinement was first studied decades ago [2,3], recent developments have led to a resurgence of interest in the subject. For ³He confined to a slab of thickness D on the order of the superfluid coherence length ξ_0 , Ginzburg-Landau (GL) [4] and quasiclassical [5–8] theories predict that the A phase may appear at zero pressure, and that the B phase gives way to the planardistorted B phase (PDB phase) with a gap that differs in directions parallel and normal to the confinement plane. Owing to advances in microfabrication techniques, those early predictions have recently been verified experimentally [9–13]. Since, for moderate confinement, the PDB phase can be viewed as a three-dimensional (3D) topological superfluid [14–17], such ³He films would also provide an ideal platform for the detection of Majorana fermions [18–21].

In addition to the A and PDB phases, which are homogeneous in the confinement plane, Vorontsov and Sauls predicted, in 2007, that confinement could lead to an additional superfluid phase with spontaneously broken translation symmetry—the stripe phase [22–24]. The stripe phase is a unidirectional pair density wave (PDW) [25] analogous to the Larkin-Ovchinnikov state [26–28] that can also be understood as a periodic arrangement of domain walls [29,30] in the PDB phase order parameter. While such domain walls are not energetically favorable in bulk ³He, they reduce the amount of surface pair breaking relative to a homogeneous phase under planar confinement [31].

Recent experiments have sought evidence of the stripe phase via nuclear magnetic resonance (NMR) measurements of the superfluid order parameter [12] and fourthsound measurements of the superfluid density [13]. The latter experiment suggested that a new phase sandwiched between the A and PDB phases appears under sufficient confinement, although the precise nature of this phase and of the transitions surrounding it remains to be elucidated. Reference [12], likewise, found evidence of a new phase in the vicinity of the A-PDB transition, but the NMR signatures of this phase were inconsistent with the stripe phase. Specifically, the observation of a kink in the NMR frequency shift at a critical tipping angle β^* ruled out the stripe phase, which should exhibit no such kink [23], but the measured value of β^* did not match that expected for the translation-invariant PDB phase. Rather, the authors of Ref. [12] reconciled those observations by proposing a "polka-dot phase" [32]: a two-dimensional (2D) PDW, of possibly hexagonal or square symmetry. The possibility of a 2D PDW as an alternative to the 1D stripe phase was recognized by Vorontsov and Sauls [22,33], but not explored quantitatively [34]. A preliminary GL analysis for a 2D PDW with the symmetry of a square lattice, mentioned in Ref. [12], found that such a phase was only metastable, with a free energy higher than that of the stripe phase.

To resolve this conundrum from a theoretical standpoint, we provide, in this Letter, a general physical argumentsummarized in this paragraph and later substantiated by explicit calculations-according to which a PDW in confined ³He ought, indeed, to be two dimensional and, also, to possess the hexagonal symmetry of a triangular lattice. Our theory can be viewed as a generalization of Landau's theory of weak crystallization [36] to superfluids. In Landau's theory, the Fourier component n_q of an equilibrium deviation $\delta n(\mathbf{r}) = n(\mathbf{r}) - n_0$ of a classical fluid's density $n(\mathbf{r})$ from uniform density n_0 serves as the order parameter for the liquid-solid transition. As the transition is approached from the liquid side, density fluctuations become strongly peaked in reciprocal space on a surface of momenta $|\mathbf{q}| = Q$ which is spherical on account of the fluid's unbroken rotational symmetry. Although this naively induces an instability toward crystallization for a continuously degenerate set of wave vectors [37], the Landau free-energy functional contains a cubic term $\sim \sum_{\pmb{q}_1, \pmb{q}_2, \pmb{q}_3} n_{\pmb{q}_1} n_{\pmb{q}_2} n_{\pmb{q}_3} \delta_{\pmb{q}_1 + \pmb{q}_2 + \pmb{q}_3, 0}$ which lifts this degeneracy, simultaneously rendering the transition first order and favoring crystalline lattices for which wave vectors add up to zero in triangles. A first attempt to transpose these ideas to a PDW or "superfluid crystallization" transition in ³He meets the objection that cubic terms in the GL functional for a superfluid are forbidden by U(1) gauge symmetry. However, the latter functional is appropriate for a superfluid transition out of the normal state. As we show below, the Landau functional for a PDW transition within a superfluid state, in which gauge symmetry is already broken, can, indeed, contain a cubic term. This term is analogous to that of the liquid-solid transition, with n_a replaced by the Fourier components of the superfluid order parameter. Combined with the softening at a finite wave vector $|\mathbf{q}| = O$ of a particular collective mode in the confined superfluid [38], the cubic term drives a first-order transition from the homogeneous superfluid to a PDW whose Bravais lattice structure, being two dimensional, is necessarily triangular.

Fluctuations in the PDB phase.—Our starting point is the GL free-energy functional for 3 He [1]

$$F = \int d^3 r [K_1 \partial_k A_{\mu j} \partial_k A^*_{\mu j} + K_2 \partial_j A_{\mu j} \partial_k A^*_{\mu k} + K_3 \partial_k A_{\mu j} \partial_j A^*_{\mu k} + \alpha \operatorname{tr} A A^{\dagger} + \beta_1 |\operatorname{tr} A A^T|^2 + \beta_2 (\operatorname{tr} A A^{\dagger})^2 + \beta_3 \operatorname{tr} A A^T (A A^T)^* + \beta_4 \operatorname{tr} (A A^{\dagger})^2 + \beta_5 \operatorname{tr} A A^{\dagger} (A A^{\dagger})^*],$$
(1)

where $A_{\mu j}$ is the 3 × 3 superfluid order parameter with μ spin and *j* orbital indices, and α , β_1 , ..., β_5 , and K_1 , K_2 , K_3 are phenomenological parameters. We will employ values of these parameters given by the weak-coupling approximation [1], but comment on the effect of strong-coupling corrections [23,39] at the end. Assuming planar confinement with specular interfaces in the *z* direction, the order parameter in the PDB phase is [8]

$$\bar{A}_{\mu j}(z) = \begin{pmatrix} \Delta_{\parallel}(z) & 0 & 0\\ 0 & \Delta_{\parallel}(z) & 0\\ 0 & 0 & \Delta_{\perp}(z) \end{pmatrix}, \qquad (2)$$

which incorporates the planar phase [8] with $\Delta_{\perp}(z) = 0$ as a special case. The order parameter is uniform in the confinement (*xy*) plane and invariant under simultaneous $SO(2)_{L_z+S_z}$ rotations of the orbital and spin coordinates about the *z* axis. To search for a PDW instability in the PDB phase, we write $A_{\mu i}$ as

$$A_{\mu j}(\boldsymbol{r}_{\parallel}, z) = \bar{A}_{\mu j}(z) + \sum_{\boldsymbol{q}} \phi_{\mu j}(\boldsymbol{q}, z) e^{i \boldsymbol{q} \cdot \boldsymbol{r}_{\parallel}}, \qquad (3)$$

where $\mathbf{r}_{\parallel} = (x, y)$ and $\mathbf{q} = (q_x, q_y)$. Our strategy is to construct a Landau functional for the fluctuation ϕ by treating it as a small correction to the PDB order parameter (2) and expanding (1) to quartic order in ϕ . Subtracting the free energy $F[\bar{A}]$ of the PDB phase, and dividing by the sample area, the resulting PDW free-energy-density functional is of the form

$$f_{\rm PDW}[\phi] = f^{(2)}[\phi] + f^{(3)}[\phi] + f^{(4)}[\phi], \qquad (4)$$

where the superscripts refer to the order of expansion in ϕ , and linear terms are absent since \overline{A} is a stationary point of *F*.

First, we focus on the quadratic term $f^{(2)}$, which contains contributions from both the quadratic and quartic terms in (1), and can be written as

$$f^{(2)}[\phi] = \sum_{q} \int_{0}^{D} dz \, \phi_{\mu j}^{*}(q, z) \hat{C}_{\mu j, \nu k}(q, z) \phi_{\nu k}(q, z), \qquad (5)$$

where *D* is the sample thickness, and \hat{C} is a *q*-dependent Hermitian differential operator that can be written as the sum of two terms, $\hat{C}(\mathbf{0}, z) + \delta \hat{C}(\mathbf{q}, z)$. $\hat{C}(\mathbf{0}, z)$ contains "kinetic" terms proportional to ∂_z^2 as well as *z*-dependent "potential" terms quadratic in the equilibrium order parameters $\Delta_{\parallel}(z)$ and $\Delta_{\perp}(z)$ [38]. $\delta \hat{C}(\mathbf{q}, z)$ contains explicitly *q*-dependent terms arising from the derivative terms in (1).

In the spirit of a Landau expansion, first, we use Eq. (5) to determine the normal modes of the system and isolate the particular mode that becomes critical at the PDW transition. These normal modes are the eigenvectors $\Phi_{\boldsymbol{q}}^{(j)}(z)$ of $\hat{C}(\boldsymbol{q}, z)$ with eigenvalues $\lambda^{(j)}(\boldsymbol{q})$, which we compute numerically [40]. Expanding the fluctuation ϕ in (5) in terms of those normal modes, we have

$$f^{(2)}[\phi] = \sum_{q} \sum_{j} \lambda^{(j)}(q) |u_{q}^{(j)}|^{2},$$
(6)

where $u_q^{(j)}$ is the amplitude of the fluctuation in the normal mode $\Phi_q^{(j)}(z)$. In Fig. 1(a), we plot the lowest 25 eigenvalues, which, by rotational invariance in the PDB phase, depend only on the magnitude q of the wave vector q. Omitting spin and orbital indices, the fluctuation $\phi(\mathbf{r}) = \sum_q \phi(\mathbf{q}, z) e^{i\mathbf{q}\cdot\mathbf{r}_{\parallel}}$ can be decomposed into real $\phi^+(\mathbf{r})$ and imaginary $\phi^-(\mathbf{r})$ parts which do not mix at quadratic order because of time-reversal symmetry in the PDB phase; the normal modes can then be separated into real and imaginary modes according to this decomposition. At $\mathbf{q} = 0$, the normal modes carry an additional $SO(2)_{L_z+S_z}$ angular momentum quantum number $m = 0, \pm 1, \pm 2$ [38]; a nonzero \mathbf{q} acts as a vector perturbation which mixes $\mathbf{q} = 0$



FIG. 1. (a) Eigenvalues $\lambda^{(j)}(\boldsymbol{q})$ in units of the normal-state density of states N(0) vs momentum $\boldsymbol{q} = |\boldsymbol{q}|$ for real (black) and imaginary (blue) normal modes in the PDB phase; their square root (b),(c) is proportional to collective mode frequencies. Parameters are chosen as D = 300 nm, P = 10 bar, and T = 0.914 mK; other parameters in the PDB phase give qualitatively similar results.

eigenmodes with different angular momenta. By plotting the square root $\sqrt{\lambda^{(j)}(q)}$ of the normal mode eigenvalues, which is proportional to bosonic collective mode frequencies $\omega_j(q)$ [38], we find one imaginary [Fig. 1(b)] and three real [Fig. 1(c)] linearly dispersing Goldstone modes. The former corresponds to the usual U(1) phase (phonon) mode of neutral superfluids, and the latter are associated with the $SO(3)_S \times SO(2)_{L_z} \rightarrow SO(2)_{L_z+S_z}$ breaking of spin and orbital symmetries peculiar to the PDB phase [40].

Mode softening.-In addition to the gapless Goldstone modes, the PDB phase supports gapped collective modes which, should they soften as external parameters such as temperature T, pressure P, or confinement D are varied, can lead to additional symmetry-breaking instabilities within the superfluid state. Such mode softening was observed in Ref. [38], where upon tuning the sample thickness across a certain critical value, the frequency $\omega_{i_{*}}(\boldsymbol{q})$ of a particular real collective mode $j = j_*$ was found to touch zero at a finite wave vector |q| = Q and subsequently become purely imaginary. The ensuing instability was then argued to lead to a 1D stripe phase with wave vector Q. In our GL description (6), this softening corresponds to the (real) eigenvalue $\lambda^{(j_*)}(q)$ for a particular real normal mode $\Phi_{a}^{(j_{*})}(z)$ continuously crossing from positive to negative on a ring of momenta |q| = Q (Fig. 2), and identifies this mode as the critical mode for the PDW transition. The instability region is bounded by two mode softening temperatures T_1^* and T_2^* that straddle the A-PDB transition line [Fig. 3(a)]; given our expansion (3), T_2^* corresponds, here, to an instability of the planar (P) phase, which is degenerate with the A phase at weak coupling [8]. Thus, discarding the noncritical modes in the vicinity of the transition, we can approximate



FIG. 2. Black lines: eigenvalues $\lambda^{(j)}(q)$ vs momentum q = |q| for real normal modes in the vicinity of the softening instability at $T = T_1^*$: (a) before softening (T = 1.318 mK, r > 0); (b) at softening $(T = T_1^* = 1.336 \text{ mK}, r = 0)$; (c) after softening (T = 1.380 mK, r < 0). Here, *r* is the tuning parameter for the instability, appearing in the approximate form $\lambda^{(j_*)}(q) \approx r + \kappa (q^2 - Q^2)^2$ of the critical mode eigenvalue (dashed red line). Parameters are chosen as D = 300 nm and P = 10 bar, and we find $Q \approx \pi/(\sqrt{3}D)$ as in Ref. [23].



FIG. 3. (a) Phase diagram of a D = 300 nm slab, with modesoftening temperatures T_1^* and T_2^* flanking the would-be A-PDB transition line T_{AB} . The instability on the PDB side (T_1^*) is preempted by a first-order transition at T_{PDW} , which (b) is approximately 4% lower than T_1^* relative to the width of the instability region.

$$f^{(2)}[\phi] \approx \sum_{q} [r + \kappa (q^2 - Q^2)^2] |u_q|^2,$$
(7)

where $u_q \equiv u_q^{(j_*)}$ is the critical mode amplitude, which obeys $u_q^* = u_{-q}$ since $\phi^+(\mathbf{r})$ and its normal-mode decomposition are real. We have also replaced the exact normal mode eigenvalue $\lambda^{(j_*)}(\mathbf{q})$ by an approximate functional form which captures its key qualitative features near the instability (Fig. 2). The parameter \mathbf{r} changes sign across the instability, and κ controls the velocity of the linearly dispersing mode $\omega_{j_*}(\mathbf{q}) \propto \sqrt{\kappa}Q||\mathbf{q}| - Q|$ that obtains at criticality ($\mathbf{r} = 0$) near $|\mathbf{q}| = Q$. As we now discuss, this further approximation—while not strictly necessary allows for a simplified analytical treatment of the PDW transition that makes its analogy to the liquid-solid transition manifest [43].

Landau theory of the PDW transition.—So far, our analysis has identified the critical normal-mode amplitude u_q —or equivalently its position-space inverse Fourier transform $u(r_{\parallel}) = \sum_{q} e^{iq \cdot r_{\parallel}} u_q$, which is a real function as the appropriate order parameter for the PDW transition. A well-behaved Landau functional for u must include terms beyond quadratic order, i.e., self-interaction terms which arise from the terms cubic and quartic in ϕ in the freeenergy-density functional (4). To compute these terms, we substitute in $f^{(3)}[\phi]$ and $f^{(4)}[\phi]$ the approximate mode expansion $\phi \approx \sum_{q} u_{q} \Phi_{q}^{(j_{*})}(z) e^{iq \cdot r_{\parallel}}$ that discards the noncritical modes $\Phi_{q}^{(j)}(z)$ with $j \neq j_{*}$. One obtains $f^{(k)}[\phi] \approx$ $\sum_{q_1,...,q_k} \Gamma^{(k)}(q_1,...,q_k) u_{q_1} \cdots u_{q_k} \delta_{\sum_{j=1}^k q_j,0}$, where the k-point vertex $\Gamma^{(k)}$ is, in general, momentum dependent [40], and the corresponding interaction nonlocal in position space. In the spirit of a gradient expansion, we approximate this nonlocal interaction by a local interaction $f^{(k)}[\phi] \propto \int d^2 r_{\parallel} u^k(\mathbf{r}_{\parallel})$ obtained by setting to zero all momenta in $\Gamma^{(k)}$. The cubic and quartic terms in (4) become

$$f^{(3)}[\phi] \approx -w \sum_{q_1, q_2, q_3} u_{q_1} u_{q_2} u_{q_3} \delta_{q_1 + q_2 + q_3, 0}, \tag{8}$$

$$f^{(4)}[\phi] \approx \eta \sum_{q_1, q_2, q_3, q_4} u_{q_1} u_{q_2} u_{q_3} u_{q_4} \delta_{q_1 + q_2 + q_3 + q_4, 0}, \qquad (9)$$

where $w \propto \Gamma^{(3)}(0, ..., 0)$ and $\eta \propto \Gamma^{(4)}(0, ..., 0)$. We find that $\eta > 0$ regardless of temperature, pressure, and confinement [40], thus, $f_{\rm PDW}$ in (4) is properly bounded from below.

Equations (7), (8), and (9) define a well-behaved Landau functional for the PDW transition in confined ³He, which is the first main result of this Letter. This Landau functional being mathematically identical to that for the crystallization transition in classical statistical mechanics [36,43], we simply transpose well-known results for the latter to the PDW transition. Restricting our analysis to momenta q with fixed magnitude |q| = Q, since, at r = 0, only modes with such momenta soften, we identity the PDW phase as that in which order-parameter configurations $u_q \neq 0$ globally minimize f_{PDW} . Provided $w \neq 0$, the continuous degeneracy of such configurations is partially lifted by the cubic term (8), which favors a PDW with noncollinear wave vectors such that $q_1 + q_2 + q_3 = 0$, i.e., that add up to zero in triangles. In the planar phase at T_2^* , we find w = 0 and the degeneracy remains unresolved at the mean-field level [40]. In the PDB phase at T_1^* , however, we find $w \neq 0$ at all pressures and the noncollinear constraint applies. In 2D, this necessarily implies a PDW whose reciprocal lattice is triangular [43], corresponding to a triangular Bravais lattice with lattice constant $a = 4\pi/(\sqrt{3}Q)$ in position space. This conclusion is the second main result of this Letter.

To be explicit, we set the PDW order parameter u_q to a constant u for $q \in \{\pm G_1, \pm G_2, \pm G_3\}$ and to zero otherwise, where we can take $G_1 = Q(1,0)$, $G_2 = Q(-\frac{1}{2}, \frac{\sqrt{3}}{2})$, and $G_3 = -G_1 - G_2$ without loss of generality $(f_{\text{PDW}}$ is invariant under a global rotation). The Landau functional (4) then reduces to a simple function of u

$$f_{\rm PDW}(u) = 6ru^2 - 12wu^3 + 216\eta u^4, \tag{10}$$

whose phase diagram is well understood [43]. Assuming fixed pressure and sample thickness for illustration, we can write $r = b(T_1^* - T)$ in the vicinity of the PDW transition from the PDB side, where b > 0. Because of the cubic term, the mode softening instability at T_1^* is preempted by a first-order transition at a lower temperature $T_{PDW} < T_1^*$



FIG. 4. (a) Irreducible *z*-dependent amplitudes of the PDW order parameter; these multiply invariants of the triangular spin-orbital point group that involve r_{\parallel} -dependent basis functions $\Psi(r_{\parallel})$ for the (b) A_1 , (c) E_1 , and (d) E_2 irreducible representations of D_6 .

$$T_{\rm PDW} = T_1^* - \frac{w^2}{36\eta b},$$
 (11)

which is the true PDW transition temperature. We find the difference $T_1^* - T_{PDW}$ is approximately 4% of the width $T_2^* - T_1^*$ of the instability region for a wide range of pressures and sample thicknesses [Fig. 3(b)]. T_1^* is then understood as the limit of metastability of the uniform (PDB) phase inside the PDW phase. Conversely, the PDW phase exists as a metastable phase for $T^{**} < T < T_{PDW}$ where $T^{**} = T_{PDW} - w^2/(288\eta b)$.

Triangular lattice PDW.—Finally, we turn to the detailed structure of the PDW order parameter. In the PDW phase, the continuous $SO(2)_{L_z+S_z}$ spin-orbital rotation symmetry is spontaneously broken to a discrete $C_6^{L_z+S_z}$ subgroup of joint spin-orbital rotations. The PDW order parameter $\phi = u \sum_q \Phi_q^{(j_*)}(z) e^{iq \cdot r_{\parallel}}$, where the sum is now restricted to the six wave vectors $q \in \{\pm G_1, \pm G_2, \pm G_3\}$, is, in fact, invariant under the full spin-orbital point group $D_6^{L_z+S_z}$

$$\mathcal{D}(g)\phi(g^{-1}\boldsymbol{r}_{\parallel},z)\mathcal{D}^{-1}(g) = \phi(\boldsymbol{r}_{\parallel},z), g \in D_{6}, \quad (12)$$

where the representation matrices $\mathcal{D}(g)$ act on both orbital and spin indices. Using the theory of invariants [44], ϕ can be expressed as $\phi = u \sum_{j,k} \phi^{(j,k)}(z) X^{(j,k)}(\mathbf{r}_{\parallel})$ where $X^{(j,k)}(\mathbf{r}_{\parallel})$ denotes the *k*th $D_6^{L_z+S_z}$ invariant associated with the irreducible representation *j* of D_6 , and $\phi^{(j,k)}(z)$ the corresponding nonuniversal amplitude encapsulating the *z* dependence of the PDW order parameter [40]. Only five such amplitudes are nonzero [Fig. 4(a), also, Fig. (S1) in Ref. [40]]; the corresponding *X* invariants involve basis functions for the irreducible A_1 , E_1 , E_2 representations [Figs. 4(b)–4(d)], to be understood as triangular lattice harmonics with angular momentum $\ell = 0, 1, 2$, respectively. The *z* dependence of the amplitudes in Fig. 4(a) is such that ϕ is also invariant under reflection in the *z* direction: $M_z \phi(\mathbf{r}_{\parallel}, -z) M_z^{-1} = \phi(\mathbf{r}_{\parallel}, z)$, where $M_z =$ diag(1, 1, -1) acts on both orbital and spin indices. *Conclusion.*—In summary, we have proposed a general mechanism whereby a 2D PDW with hexagonal symmetry is stabilized in confined ³He: the Landau functional for a PDW transition within the uniform superfluid generically contains a cubic term which, upon approach to a crystallization instability, leads to a first-order transition to a PDW with noncollinear wave vectors forming a triangular lattice. We demonstrated that, in the weak-coupling approximation, this mechanism is operative for a wide range of sample thicknesses and pressures near the crystallization instability within the PDB phase.

In our weak-coupling treatment, the coefficient of the cubic term was found to vanish at the PDW instability of the planar phase (T_2^*) . In reality, strong-coupling effects stabilize the A phase over the planar phase in this part of the phase diagram, and Eq. (2) is the wrong expansion point. The construction of a PDW Landau functional using the A phase as a starting point and incorporating strong-coupling corrections would be necessary to address the question whether the A-PDW transition remains continuous or also becomes first order. Additionally, direct numerical minimization of the 3D GL functional using a $D_6^{L_z+S_z}$ -invariant PDW ansatz beyond the single-harmonic approximation utilized here would be desirable for more quantitative comparisons with experiment.

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