I. INTRODUCTION

The remarkable realization of synthetic spin-orbit coupling in quantum gases is opening new perspectives in the study of many-body phenomena with ultracold atoms. To date, a specific type of spin-orbit coupling, which is generated by a pair of Raman laser beams, has been experimentally achieved in Bose-Einstein condensate (BEC) of $^{87}$Rb and degenerate Fermi gases of $^{40}$K and $^{6}$Li. For bosonic atoms, Raman coupling could give rise to novel superfluid phases due to the dramatic modification of the single-particle dispersion. One particular interesting phase is the stripe (STR) BEC, which simultaneously breaks the $U(1)$ symmetry and the translational invariance symmetry. In current experiments of Rb gases, due to the slight difference between inter- and intra-species interactions, the stripe phase is expected to exist only in the weak coupling regime. At a critical Raman strength, a first order transition between the STR phase and plane-wave (PW) phase takes place. Since the PW condensate is almost fully-polarized in this regime, the STR-PW transition can be also regarded as a consequence of the competition between the crystalline order and the ferromagnetic order.

So far, the spatial density modulation in the stripe phase has not been directly observed experimentally. The major difficulties are the small contrast of the stripes and the limited resolution of in-situ imaging. Nevertheless, a miscible-immiscible transition has been identified at a value of Raman strength, which is close to the theoretical predicted critical point of the STR-PW transition. Very recently, the temperature dependence of the phase boundary is experimentally determined through the statistical analysis of magnetization measurements.

Previously, most theoretical works focus on the ground state properties and quantum dynamics of the STR phase and the PW phase, very few studies pay attention to the thermal effects at finite temperature. In particular, to our knowledge, the influence of thermal fluctuations on the STR-PW transition has not been addressed in the literature. In this work, we develop a perturbation approach to resolve this problem in weak Raman coupling regime. By expanding the equation of state in terms of the Raman strength up to the second order, we find a temperature dependent STR-PW phase boundary, which is in qualitative agreement with recent experimental observations.

II. PERTURBATION FORMALISM

Consider a two-component Bose gas coupled by a pair of counter propagating Raman beams along $x$ direction, the effective Hamiltonian is given by (set $\hbar = k_B = 1$)

$$\hat{H} = \hat{H}_0 + \Omega \hat{R} + \hat{H}_{\text{int}},$$

$$\hat{H}_0 = \int d\mathbf{r} \hat{\psi}^\dagger \left( -\frac{\hbar^2}{2m} \nabla^2 - \hbar \sigma_z \right) \hat{\psi},$$

$$\hat{R} = \frac{1}{2} \int d\mathbf{r} \hat{\psi}^\dagger \left( \sigma_x e^{-i2k_x x} + \text{H.c.} \right) \hat{\psi},$$

$$\hat{H}_{\text{int}} = \frac{1}{2} \sum_{\sigma,\sigma'} \int d\mathbf{r} \hat{\psi}_\sigma^\dagger \hat{\psi}_{\sigma'} \hat{\chi}_{\sigma\sigma'} \hat{\psi}_{\sigma'} \hat{\psi}_{\sigma}$$

where $\hat{\psi}_\sigma$ is the field operator for the pseudo-spin $\sigma$, $\hat{\psi}_\sigma^\dagger = (\hat{\psi}_{\uparrow\uparrow}^\dagger, \hat{\psi}_{\downarrow\downarrow}^\dagger)$, $k_x$ is the recoil momentum of the laser beams, $\Omega$ is the Raman coupling strength, $\hbar$ is the effective Zeeman field fixed by the Raman detuning, $\sigma_{\alpha\beta}$ are the contact interaction parameters with $g_{\uparrow\downarrow} = g_{\downarrow\uparrow}$, $\sigma_{\alpha,y,z}$ are Pauli operators, $\tilde{\sigma}_z = \frac{1}{2}(\sigma_z \pm i\sigma_y)$, and $\mathbf{I}$ is the identity matrix.

When the Raman coupling is weak, one can treat $\Omega \hat{R}$ as a perturbation and expand free energy in terms of $\Omega$. In the linear response regime, the expansion can be truncated at second order, and free energy at temperature $T$ is given by

$$F(\Omega) = F + \Omega \langle \hat{R} \rangle - \frac{1}{2} \chi \Omega^2.$$ 

Here, all the thermodynamic quantities on the right hand side of Eq. (5) are for the equilibrium state without Raman coupling at same temperature: $F$ is free energy, $\langle \hat{R} \rangle$ is the mean value of the Raman field.
is the ensemble average value of $\hat{R}$, and $\chi$ is referred to as Raman susceptibility with a formalistic expression

$$\chi = 2 \sum_{\ell \neq \ell'} \rho_\ell \frac{|\langle \Phi_{\ell'} | \hat{R} | \Phi_\ell \rangle|^2}{E_{\ell'} - E_\ell},$$

where $|\Phi_\ell \rangle$ is the $\ell$-th eigen-state of the Hamiltonian $\hat{H}_0 + \hat{H}_{\text{int}}$, $E_\ell$ is the corresponding eigen-energy, $\rho_\ell = e^{-E_\ell/T}/Z$, and $Z$ is the partition function. At $T = 0$, Eq. (5) and (6) just reduce to the usual second order perturbation formula in quantum mechanics.

The expansion of free energy in Eq. (5) can be further simplified by recognizing that the average value of $\hat{R}$ always vanishes in absence of Raman coupling, i.e.,

$$\langle \hat{R} \rangle = 0.$$

This is because the equilibrium states in the case of $\Omega = 0$ persist the translational invariance symmetry, while the operator $\hat{R}$ does not commute with total momentum. An alternative argument bases on the fact that free energy of the STR/PW phase should not depend the sign of $\Omega$, hence the linear term in the expansion must vanish.

We note that the effective Hamiltonian [1] is written in the laboratory frame. In contrast, the rotating frame is frequently used in previous studies [5, 6, 8–11], where a unitary transformation $\hat{U} = e^{i\hat{h}_{\text{rot}} \tau_3}$ is performed. While the rotating frame is useful in many cases, the laboratory frame is much more convenient to our problem, because all the quantities in the perturbation theory only concern the states without Raman coupling. One can be readily checked that the value of Raman susceptibility $\chi$ is actually independent of the frame [13].

A key ingredient in the perturbation theory is that the equilibrium states evolve smoothly when Raman coupling is switched on. To shed more light on this point, it is helpful to recall the condensate wave-function at $\Omega = 0$. Consider the symmetric case with $g_{\uparrow\uparrow} = g_{\downarrow\downarrow} = g$ and $h = 0$. In the rotating frame, the STR phase and the PW phase can be described by a variational wave-function [6],

$$\psi = \sqrt{n} \left[ c_+ \left( \frac{\cos \eta}{\sin \eta} \right) \cos \left( \frac{\theta}{2} \right) - \sin \left( \frac{\theta}{2} \right) \frac{\sin \eta}{\cos \eta} \right] + c_- \left[ \sin \left( \frac{\theta}{2} \right) \right]$$

with $n = N/V$ the total density of atoms. In the STR phase, $|c_+|^2 = |c_-|^2 = 1/2$; in the PW phase, one of the coefficients $c_\pm$ is zero. For a given Raman strength $\Omega$, variation parameters $\eta$ and $p_0$ have been determined in Ref [6]. As $\Omega \to 0$, one finds $p_0 \to k_r$ and $\eta \to 0$ in both the STR phase and the PW phase, therefore, the condensate wave-functions in the laboratory frame ($\varphi = \hat{U}^{-1} \varphi$) reduce to

$$\varphi_{\text{STR}} \overset{\Omega \to 0}{\longrightarrow} \sqrt{n} \left( e^{i\theta_i} e^{i\theta_j} \right), \quad (8)$$

$$\varphi_{\text{PW}} \overset{\Omega \to 0}{\longrightarrow} \sqrt{n} \left( e^{i\theta_i} \right) \text{ or } \sqrt{n} \left( e^{i\theta_j} \right), \quad (9)$$

with $\theta_i$ the phase of $\sigma$-component. As it is well known, the unpolarized BEC and ferromagnetic BEC described by Eq. (3) and (4) are two possible ground states in a spin-$1/2$ Bose gas without interspecies coupling. When the Raman coupling is gradually switched on, these two phases continuously evolve into the STR phase and the PW phase respectively. At finite temperature, although equilibrium states are not characterized by the condensate wave-function alone, similar connections are still expected.

In the following, we apply the perturbation formalism to study the transition between the STR phase and the PW phase. For this purpose, we focus on the case with $g_{\uparrow\downarrow} = g_{\downarrow\uparrow} = g$ and $h = 0$, where the Hamiltonian possesses a $\mathbb{Z}_2$ symmetry [13]. And all the interactions are assumed to be repulsive. This simplified Hamiltonian is a minimal model to understand the ground state phase diagram [3, 6], and it is also relevant to the available experiments with rubidium atoms [2, 7]. The extension to more complicated situations is straightforward.

### III. THERMODYNAMICS IN ABSENCE OF RAMAN COUPLING

In this section, we study the thermodynamics of two-component Bose gases without Raman coupling. For the convenience of later discussions, we use $E_r = k_r^2/(2m)$ as energy unit in the numerical calculation. The density of atoms $n$ and recoil momentum $k_r$ are set by the typical values in experiments [2, 7].

#### A. Popov approximation

In a BEC state, condensate and non-condensed atoms can be treated separately. In our case, the field operator can be written as

$$\hat{\psi}_\sigma = \varphi + \delta \hat{\psi}_\sigma = e^{i\theta_\sigma} \left( \sqrt{n_{0\sigma}} + \frac{1}{\sqrt{n}} \sum_p \hat{\psi}_{p\sigma} e^{ipx} \right)$$

where $\varphi = \langle \hat{\psi} \rangle$ is the condensate wave-function of $\sigma$-component, $n_{0\sigma}$ is the condensate density, and $\sum_p$ denotes the summation excluding zero momentum. Non-condensed atoms, which are usually negligible in ground state, may play an important role at higher temperature due to thermal fluctuations. In weakly interacting gases, the interactions between non-condensed atoms can be treated in the mean-field manner. A widely used mean-field prescription is Popov approximation [15], which recovers Bogoliubov theory in low temperature limit and reduces Hartree-Fock theory when condensate vanishes above $T_c$. In Popov approximation, the grand-canonical Hamiltonian of a spin-$1/2$ Bose gas is given by

$$\hat{K} = K^{(0)} + \hat{K}^{(2)}$$
with $K^{(0)} = V[−\mu n_0 + g(\frac{1}{2} n_{tt}^2 + \frac{1}{2} n_{0t}^2) - \delta n^2_t - \delta n^2_t) + g_{\uparrow \downarrow}(n_{0t} n_{0t} - \delta n^2_t - \delta n^2_t)]$ and

$$\hat{K}(2) = \sum p \sum_{\sigma=\uparrow, \downarrow} \left[ (\xi_p + 2g_{\sigma} n_{\sigma} + g_{\uparrow \downarrow} n_{\sigma})\psi_p^{\dagger} \psi_{\sigma}^{\dagger} \right.$$

$$+ \frac{1}{2}(g_0 n_{\sigma} \psi_p^{\dagger} \psi_{-\sigma} + g_{\uparrow \downarrow} \sqrt{n_{0t} n_{0\delta}} \psi_p^{\dagger} \psi_{-\sigma} + \text{H.c.}) + g_{\uparrow \downarrow}(\sqrt{n_{0t} n_{0\delta}} + \delta s) \psi_p^{\dagger} \psi_{-\sigma} \left. \right] .$$

(11)

Here, $\delta n_\sigma = \frac{1}{V} \sum_p (\psi_p^{\dagger} \psi_{\sigma}^{\dagger})$ and $n_\sigma = n_{0\sigma} + \delta n_\sigma$ are the non-condensate density and total density of $\sigma$-component respectively, $\delta s = \frac{1}{V} \sum p (\psi_p^{\dagger} \psi_{p\uparrow}^{\dagger})$ is the spin-flipping mean-field parameter, $n_{\sigma} = n_{0\sigma} + n_{\sigma\downarrow}$ is total condensate density, $\mu$ is chemical potential, $\xi_p = \epsilon_p - \mu = p^2 / (2m) - \mu$, and $\sigma$ denotes the spin opposite to $\sigma$.

The condensate wave-function should be determined by energy minimization. The stationary conditions $\langle \partial K / \partial \varphi_\sigma \rangle = 0$ can be explicitly written as

$$\mu \varphi = L \varphi ,$$

(12)

with $\varphi = (\varphi_\uparrow, \varphi_\downarrow)$ and

$$L = \left( \begin{array}{cc} g(n_{0\uparrow} + 2n_{\uparrow}) + g_{\uparrow \downarrow} n_{\downarrow} & g_{\uparrow \downarrow} n_{\downarrow} e^{(\theta_{\downarrow} - \theta_{\uparrow})} - g_{\uparrow \downarrow} n_{\downarrow} e^{(\theta_{\downarrow} - \theta_{\uparrow})} \\ g_{\uparrow \downarrow} n_{\downarrow} e^{(\theta_{\downarrow} - \theta_{\uparrow})} & g(n_{0\downarrow} + 2n_{\uparrow}) + g_{\uparrow \downarrow} n_{\uparrow} \end{array} \right) .$$

At low temperature, where non-condensate atoms can be safely ignored, Eq. (12) reduces to the time-independent Gross-Pitaevskii equation. Once the condensate wavefunction is determined, the quadratic Hamiltonian can be readily solved via Bogoliubov transformation.

\section*{B. Equation of state}

There are two possible equilibrium states satisfying the stationary equation (12). One is the spin-balanced phase, where both condensate and non-condensate atoms are unpolarized, i.e., $n_{0\uparrow} = n_{0\downarrow}$ and $n_{\uparrow \downarrow} = n_{\downarrow \uparrow}$. Through a standard diagonalization procedure, we obtain free energy of the spin-balanced phase as

$$F_B = E_{B0} + T \sum_{p} \sum_{\sigma = \uparrow, \downarrow} \ln (1 - e^{-\omega_p \sigma / T}) ,$$

(13)

where $E_{B0} = V[\frac{1}{2}(g_{+} n_{+}^2 + g_{-} n_{-}^2) + g_{\uparrow \downarrow} n_0 + g_{\uparrow \downarrow} \delta n_0] + \frac{1}{2} \sum_p \omega_p \left( \omega_p - 2\epsilon_p n_{\sigma} n_{0\delta} + \omega_p + \omega_p \right) / (4\epsilon_p)$

with $\delta n = \delta n_0 + \delta n_\downarrow$, and $\omega_p$ are excitation spectrum of quasi-particles,

$$\omega_p = \sqrt{\epsilon_p (\epsilon_p + g_n n_0)} ,$$

(14)

$$\omega_{p+} = \sqrt{\epsilon_p (\epsilon_p + g_n n_0)} ,$$

(15)

where $g_{\pm} \equiv g \pm g_{\uparrow \downarrow}$. The mean-field parameters $\delta n$ and $\delta s$ should be self-consistently determined from

$$\delta n = \frac{2}{V} \sum_{p} \sum_{\sigma = \pm} \left( \omega_p^{2} + \frac{1}{2} \omega_p \right) f_p ,$$

(16)

$$\delta s = \frac{1}{V} \sum_{p} \sum_{\sigma = \pm} \alpha \left( \omega_p^{2} + \frac{1}{2} \omega_p \right) f_p ,$$

(17)

where $f_p = 1 / (e^{\omega_p T} - 1)$, $u_{p, \sigma}$ and $v_{p, \sigma}$ are coefficients of Bogoliubov transformation with $v_{p, \sigma}^2 / \omega_p^{2} = 1 / (4 \epsilon_p)$. $\omega_p$ are neglegted the terms associated with quantum depletions at zero temperature. This treatment is well justified in dilute gases. Obviously, only a negative $\delta s$ is allowed in the self-consistency equations.

Intuitively, the spin-balanced phase is favored in the case $g_- > 0$, where the intra-species repulsion is stronger. For $g_- < 0$, the excitation branch $\omega_{p-}$ suffers an dynamic instability in the low temperature limit, and the spin-balanced phase is not available until temperature beyond a threshold value. The threshold temperature $T_B$ is determined by the stability condition

$$g_n n_0 - 2g_{\uparrow \downarrow} \delta s \geq 0 ,$$

(18)

when the equality is satisfied. Above $T_B$, the spin-balanced phase becomes a (meta-)stable state corresponding to a (local) minimum in free energy landscape.

Another possible equilibrium state is the spin-polarized phase, which spontaneously breaks $Z_2$ symmetry. In this phase, condensate is fully polarized, and spin-up and spin-down are decoupled ($\delta s = 0$). Free energy of the spin-polarized phase (assume $M_0 = +1$) is given by

$$F_p = E_{P0} + T \sum_{p} \sum_{\sigma = \uparrow, \downarrow} \ln (1 - e^{-\omega_p \sigma / T}) ,$$

(19)

where $E_{P0} = V[\frac{1}{2} (g_{+} n_{+}^2 + g_{-} n_{-}^2) + g_{\uparrow \downarrow} n_0 + g_{\uparrow \downarrow} \delta n_0 +$$

$$(g_{-} g_{\uparrow \downarrow}) n_0^2 / (4\epsilon_p)]$ with $\delta n_0 = \delta n_\downarrow - \delta n_\uparrow$, and $\omega_p$ are the excitation spectrum of quasi-particles

$$\omega_p = \sqrt{\epsilon_p (\epsilon_p + g_n n_0)} ,$$

(20)

$$\omega_{p\uparrow} = \sqrt{\epsilon_p (\epsilon_p + g_n n_0)} ,$$

(21)

The mean-field parameters $\delta n_\sigma$ should be determined by the self-consistency equations

$$\delta n = \frac{1}{V} \sum_{p} \left( \omega_p^{2} + \frac{1}{2} \omega_p \right) f_p \sigma ,$$

(22)

$$\delta n_\uparrow M = \frac{1}{V} \sum_{p} \left( \omega_p^{2} + \frac{1}{2} \omega_p \right) f_p \sigma -$$

(23)

where $f_p = 1 / (e^{\omega_p T} - 1)$, and $u_{p, \sigma} = v_{p, \sigma} = \frac{1}{2} (\epsilon_p + gn_0) \omega_p / 2$.
For $g_- > 0$, the excitation branch $\omega_{p^\dagger}$ suffers an energetic instability in the low temperature limit, and the spin-polarized phase is only available when the stability condition
\begin{equation}
g_- n_0 + (2g - g_+ ) \delta n \delta M \leq 0
\end{equation}
is satisfied. The equality of above condition determines a threshold temperature $T_p$. Above $T_p$, $\omega_{p^\dagger}$ is gapped, and the spin-polarized phase becomes a (meta-)stable equilibrium state. For $g_- < q$, $T_p$ is indeed very low.

In Fig. 1, we numerically compare the free energy of the spin-balanced phase and the spin-polarized phase at finite temperature. For $g_- > 0$, the spin-balanced phase is the only possible equilibrium state when temperature below the threshold value $T_p$. Although the spin-polarized phase becomes available at higher temperature, a free-energy-crossing is never observed, hence the system prefers the spin-balanced phase up to the condensation temperature $T_c$. On the other hand, for $g_- < 0$, $F_p < F_B$ holds in the entire temperature region $T_B < T < T_c$, which means the spin-polarized phase is energetically favored in this case.

Above results about equation of state provide strong constraints for the phase diagram in presence of Raman coupling. Since there is a finite free energy difference between the spin-balanced phase and the spin-polarized phase, an infinitesimal Raman coupling could not induce any phase transition. Therefore, for $g_- > 0$, the STR-PW transition in the weak Raman coupling limit is impossible unless temperature approaches to $T_c$, i.e., the scenario of the phase diagram as shown in Fig. 1(b) can be completely ruled out \cite{10}. Similarly, for $g_- < 0$, the STR phase at finite temperature can be also excluded in the weak Raman coupling regime [see Fig. 1(c)].

C. Raman susceptibility

According to Kubo’s formula, the fluctuation $\delta \langle \hat{R} \rangle$ generated by the Raman perturbation is described by the dynamic response function \cite{17},
\begin{equation}
\chi_R(\omega) = i \int_0^\infty dt \langle [\hat{R}(t), \hat{R}] \rangle e^{i \omega t},
\end{equation}
where the time-dependent operator $\hat{R}(t)$ is defined in Heisenberg picture as usual. Using Lehmann representation, one can immediately recognize that the Raman susceptibility in Eq. (25) is just the static response, $\chi = \chi_R(\omega = 0)$.

As a leading order approximation, we ignore the dynamics of non-condensate mean-fields and write the time-dependent field operator as
\begin{equation}
\hat{\psi}_\sigma(t) = e^{i(\theta_{\sigma} - \mu t)} [\sqrt{n_{0\sigma}} + \frac{1}{\sqrt{N}} \sum_p e^{i K t} \hat{\psi}_{p\sigma} e^{-i K t} e^{ipr}],
\end{equation}
where $K$ is the static Popov Hamiltonian given by Eq. (11). Since $K$ is diagonal in quasi-particles representation, $\chi_R$ can be easily derived. After a straightforward algebra, we obtain Raman susceptibility in the spin-balanced phase and the spin-polarized phase as
where \( k_r = (k_1, 0, 0) \), and \( \sum^n \) denotes a summation with the constraint \( p \neq 0, 2k_r \). At low temperature, the contribution from thermal atoms is negligible, Raman susceptibility can be written in analytical forms,

\[
\chi_B = \frac{N}{16E_r} + \frac{N}{16E_r + 8g_+ n},
\]

\[
\chi_P = \frac{N}{8E_r - 2g_- n},
\]

where we have replace \( N_0 \) by the total number of atoms. In the weak interacting limit, both Eq. (26) and (27) approach to the noninteracting result (see Appendix A).

In Fig. 2, we plot Raman susceptibility \( \chi_B \) and \( \chi_P \) as a function of temperature. In spite of a non-monotonic temperature dependent behavior, \( \chi_B \) is always smaller than \( \chi_P \) for \( T < T_c \). This fact implies free energy of the spin-polarized phase will decrease faster when Raman coupling switched on [see Eq. (5)]. Therefore, for \( g_- > 0 \), a transition between the STR phase and the PW phase is expected at a critical Raman coupling strength.

\[
\chi_B = \frac{N}{16E_r} + \frac{N}{16E_r + 8g_+ n},
\]

\[
\chi_P = \frac{N}{8E_r - 2g_- n},
\]

\[
\chi = \chi_B + \chi_P
\]

where \( \chi = \chi_B + \chi_P \) is a parameter of the system.

IV. TRANSITION BETWEEN STRIPE PHASE AND PLANE-WAVE PHASE

With the equation of state and Raman susceptibility obtained previously, we can determine the phase diagram in presence of Raman coupling via the perturbation approach. According to Eq. (5), free energy of the STR phase and the PW phase are given by

\[
F_{\text{STR}}(\Omega) = F_B - \frac{1}{2} \chi_B \Omega^2,
\]

\[
F_{\text{PW}}(\Omega) = F_P - \frac{1}{2} \chi_P \Omega^2.
\]

For the case \( g_- > 0 \), \( F_B < F_P \), hence system is in the STR phase when Raman coupling is weak enough. As \( \Omega \) increases, a first order transition takes place when the condition \( F_{\text{STR}}(\Omega_c) = F_{\text{PW}}(\Omega_c) \) is satisfied, and critical Raman strength can be explicitly written as

\[\Omega_c^2 = 2 \frac{F_B - F_P}{\chi_B - \chi_P}.\]

For \( \Omega > \Omega_c \), the PW phase is energetically favored. Since the spin-polarized phase is not available at very low temperature, the phase transition can be only addressed above \( T_{\text{th}} \), which is a limitation of the perturbation approach.

In Fig. 3, the phase boundary obtained from Eq. (32) is plotted for various densities. As temperature increases, the phase boundary bends toward the stripe phase side in most temperature region, which implies the PW phase is more robust than STR phase in presence of thermal fluctuations. Although the calculation is performed in uniform case, our theoretical results qualitatively agree with the recent experimental measurement in Rb gases [7]. At higher temperature close to \( T_c \) [15], the critical Raman strength shows a suspicious non-monotonic behavior. Since both \( F_B - F_P \) and \( \chi_B - \chi_P \) vanish at \( T_c \), the value of \( \Omega_c \) is sensitive to the temperature dependence details of each quantities. In fact, if we use Hartree-Fock approximation to compute \( F \) and \( \chi \) (see Appendix B), the phase boundary shows a quite different behavior in the vicinity of \( T_c \). As it is well known, mean-field theories usually produce artificial results near \( T_c \) [19], thus the STR-PW transition in this narrow region is not conclusive.

At \( \Omega_c \), we also numerically check the inhomogeneous state with a spacial separation between the STR phase and the PW phase. The density jump across the interface is found to be extremely small, hence the phase separation is almost invisible in a uniform system. Previously, the variational study at zero temperature came to a similar conclusion [8].
V. DISCUSSION AND CONCLUSION

The perturbation approach developed in this work is expected to be reliable when Raman coupling is weak enough. To provide an estimation of applicable regime, we examine the equation of state in a noninteracting Bose gas with Raman coupling. As shown in Appendix A, the expansion of free energy in Eq. (5) is very accurate for \( \Omega \lesssim 0.2 E_r \), and the contribution from higher orders can be safely ignored in this regime. A similar situation can be expected in a weakly interacting system.

The equation of state obtained via the perturbation approach can be tested by future experimental measurements and quantum Monte Carlo simulations. We note that Raman susceptibility \( \chi \) is not only a useful quantity from theoretical viewpoint but also measurable via two-photon Bragg spectroscopy. By varying the detuning of the Bragg lasers, dynamic structure factor in the density and spin channels can be measured separately \cite{2020arXiv200403736}. In the spin channel, the \( f \)-sum rule for the dynamic structure factor is modified by Raman coupling \cite{2019arXiv190501815}. From the commutation relation \( \int d\omega S_M(q, \omega) = N \left( \frac{q^2}{2m} - 2\Omega R \right) \), we derive

\[
\int d\omega S_M(q, \omega) = N \left( \frac{q^2}{2m} - 2\Omega R \right),
\]

where \( S_M(q, \omega) = \sum_{\ell, \ell'} e^{-E_{\ell}/T} |\langle \Phi_{\ell'} | \hat{\sigma}_q^z | \Phi_\ell \rangle|^2 \delta(\omega - E_{\ell'} + E_\ell) \) is the spin dynamic structure factor, \( \hat{\sigma}_q^z = \int \hat{\psi}^\dagger \hat{\sigma}_z \hat{\psi} e^{i q \cdot r} \) is spin fluctuation operator, and \( R \) is the measured value of \( R \) in presence of Raman coupling. The \( f \)-sum rule in Eq. (33) is model-independent and holds for both bosons and fermions. This exact relation provides a practical way to deduce the quantity \( R \) through the measurement of dynamic structure factor. Once \( R \) is archived, Raman susceptibility can be readily obtained from a linear fitting: for weak Raman coupling, \( \chi \) is proportional to \( \Omega \) with the simple relation \( R = -\chi \Omega \).

In present work, we only consider the uniform situation. In a harmonic trap, condensate and thermal atoms construct an inhomogeneous shell structure. To determine the density profile in trap, both the equation of state and the knowledge of condensate fraction are required. However, the condensate fraction at finite \( \Omega \) can not be directly obtained via the perturbation approach. At this stage, the phase diagram of a trapped system is still an open question, and we leave this issue to future study.

In summary, a perturbation theory of Raman coupled Bose gases is developed. The transition between the STR phase and the PW phase is investigated in the uniform case, and the phase boundary is determined at finite temperature. Our theoretical results qualitatively agree with the recent measurements in Rb gases, and the equation of state studied here may be useful to future experiments.

\section*{Appendix A: Noninteracting Bose Gas with Raman coupling}

In a non-interacting Bose gas with Raman coupling, the energy spectrum has two degenerate minima for \( \Omega < 4E_r \), and there are many possibilities for condensate to occupy these two minima. Nevertheless, the thermodynamic properties does not depend on the configuration of condensate, and free energy is given by

\[
F(\Omega) = \mu N + \sum_{p} \sum_{\alpha = \pm} [\ln(1 - e^{-\xi_{p,\alpha}})],
\]

with \( \xi_{p,\pm} = \epsilon_p + E_r \pm \sqrt{(p_x k_r/m)^2 + \Omega^2/4 - \mu} \). Below \( T_c \), \( \mu \) equals to the lowest energy of single particle dispersion, for \( \Omega < 4E_r, \mu = -\Omega^2/(16E_r) \).

In Fig. 4, we numerically compare the actual free energy with the perturbation formula \( F_{\text{pert}} = F(\Omega = 0) - \frac{1}{2} \chi \Omega^2 \), where \( \chi \) is the noninteracting Raman susceptibility

\[
\chi = \frac{N_0}{8E_r} + \sum_p \frac{m}{2k_r(k_r - p_x)} \frac{1}{e^{\epsilon_p/T} - 1}.
\]

As one can see, for weak Raman coupling, the deviation of \( F_{\text{pert}} \) from the actual value is extremely small, which justifies the expansion in Eq. (5) being a very good approximation. In the inset of Fig. 4, the noninteracting Raman susceptibility is plotted as a function of temperature up to \( T_c \).

\begin{figure}[h]
\centering
\includegraphics[width=0.5\textwidth]{fig4.png}
\caption{(color online). Comparison of actual free energy with the perturbation value \( F_{\text{pert}} \) in a noninteracting Bose gas at various temperature. Inset: noninteracting Raman susceptibility as a function of temperature (\( n = 0.5k^3 \)).}
\end{figure}

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Appendix B: Hartree-Fock approximation

The grand-canonical Hamiltonian in Hartree-Fock (HF) approximation can be readily obtained from Eq. (11) by omitting the anomalous quadratic terms. In HF theory, the formulas for free energy and Raman susceptibility remain the same as in Popov theory except the following replacements: in the spin-balanced phase, \( v_{p,\alpha} \to 0, \omega_{p+} \to \epsilon_p + \frac{1}{2} g_p n_0 \) and \( \omega_{p-} = \epsilon_p + 2 g_{p+} s + \frac{1}{2} g_{p-} n_0 \); in the spin-polarized phase, \( v_p \to 0 \) and \( \omega_{p^+} \to \epsilon_p + g m_0 \).

In Fig. 5, the boundary between the STR phase and the PW phase is plotted with \( F_B - F_P \) and \( \chi_B - \chi_P \) calculated in HF approximation. At \( T_c \), the critical Raman strength \( \Omega_c \) approaches to zero, which is in contrast to the results of Popov theory (see Fig. 3). As mentioned before, both Popov theory and HF theory are not reliable in the vicinity of \( T_c \). At very low temperature, where thermal fluctuations are dominated by phonon, HF approximation is also not good due to the gapped excitation spectrum. Nevertheless, in most temperature region the phase boundary shows a similar trend in both theories.

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