

Quantum Phases for Finite-Temperature Gases of Bosonic Polar Molecules Shielded by Dual Microwaves

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We investigate the finite-temperature phase diagram of polar molecules shielded by dual microwave fields using the path integral Monte Carlo method combined with the worm algorithm. We determine the critical temperature T_c for Bose-Einstein condensations (BECs) and identify two distinct phases below T_c : the expanding gas (EG) phase and the self-bound gas (SBG) phase. We further analyze the temperature and interaction-strength dependence of the condensate and superfluid fractions. Notably, in contrast to dilute atomic BECs, the SBG phase displays a low condensate fraction and a high superfluid fraction, resembling the behavior of strongly correlated ^4He superfluids. These significant many-body correlations arise from the interplay between long-range dipole-dipole interactions and the short-range shielding potential. Furthermore, we demonstrate that the aspect ratio of the gas provides a characteristic geometric signature to accurately determine the EG-to-SBG transition, robust against external trapping potentials. Our findings provide unbiased and numerically exact results to guide upcoming experiments with polar molecules.

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I. INTRODUCTION

Recent advances in the field of ultracold polar molecules, enabled by innovative microwave-shielding techniques [1–6], have led to groundbreaking achievements. Notable examples include the realization of a degenerate Fermi gas of NaK molecules [7,8] and the creation of ultracold tetramers $(\text{NaK})_2$ [9,10] using a single microwave field, as well as the achievement of Bose-Einstein condensations (BECs) in NaCs gases [11] via dual microwave fields. These milestones have firmly established ultracold polar molecules as a versatile platform for exploring novel quantum phenomena [12–22], with a wide range of applications in quantum simulation [23–26], quantum computing [27–32], ultracold chemistry [33–35], and precision measurements [36–41].

The mechanisms underlying both single and dual microwave shielding techniques have been elucidated

through multi-channel calculations [42–44]. The effective potential between microwave shielded polar molecules (MSPMs), featuring a dipole-dipole interaction (DDI) at long range and a characteristic $1/r^6$ repulsion at short range [42,43], provides a simplified and quantitative description of the shielding mechanism. The effective potential exhibits an extensive shielding core, which induces strong many-body correlations. Recent theoretical studies [21,22,43] suggest that the conventional Gross-Pitaevskii equation, even when extended with Lee-Huang-Yang corrections, may be inadequate for describing strongly correlated MSPMs, particularly in the strong DDI regime. To incorporate many-body correlations, cluster expansion methods [21,43] and path integral ground state Monte Carlo simulations [22] are employed to investigate the zero-temperature properties of MSPMs. These methods have shown that the significant shielding core generated by microwave fields distinguishes MSPMs apart from traditional dipolar atoms [45,46]. In BECs of magnetic atoms, the condensate fraction is typically large. In contrast, for MSPMs, the condensate fraction can be tuned from a large value in the weak DDI regime to a small value in the strong DDI regime via microwave parameters [21,43]. Consequently, MSPMs provide a unique platform that continuously bridges the gap between weakly correlated atomic gases and strongly correlated gases, akin to the superfluid ^4He with condensate fractions ranging from 7% to 13% [47–51].

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Existing studies of MSPMs have primarily focused on zero-temperature systems [21,43] or on single microwave shielding [22]. In contrast, the finite-temperature many-body properties of polar molecules under dual microwave fields remain largely unexplored. Notably, the recent experiment [11] conducted at finite temperature demonstrated the stability of BECs in NaCs gases under dual microwave shielding. Given ongoing experimental efforts involving NaCs [11] and anticipated studies on other bosonic MSPMs such as NaRb [5], a deeper and more comprehensive understanding of the finite-temperature properties of MSPMs—beyond the realm of dipolar atomic BECs and ^4He physics—is urgently needed.

In this paper, we investigate the finite-temperature behavior of a trapped gas of bosonic MSPMs under realistic experimental conditions using the state-of-the-art path integral Monte Carlo method combined with the worm algorithm [52–54]. This highly efficient, unbiased, and numerically exact approach has been successfully applied to study a wide range of strongly correlated systems, including ultracold atomic gases [55], quantum magnets [56], and quantum gases with long-range interaction [57–59]. In the finite-temperature phase diagram, we determine the critical temperature T_c for BECs, which is enhanced (reduced) by the attractive (repulsive) interactions. Below T_c , we identify a phase transition between the expanding gas (EG) phase and the self-bound gas (SBG) phase, both of which exhibit significant many-body correlations characterized by antibunched density-density correlations [21,43]. We analyze two key quantities: the condensate fraction, governed by the interplay between long-range DDI and the short-range shielding potential, and the superfluid fraction, determined by low-lying phonon excitations. While both quantities increase as temperature decreases, their responses to changes in interaction strength differ fundamentally, in contrast to dilute atomic BECs. Notably, the SBG phase exhibits a low condensate fraction and a high superfluid fraction, resembling the behavior of strongly correlated ^4He superfluids [47–51]. Furthermore, we demonstrate that although radial confinement blurs the phase boundary measured by the gas volume, the aspect ratio of the gas provides a clear signature for accurately determining the EG-to-SBG transition, highlighting the intricate interplay between condensate fractions and DDI.

II. MODEL

We consider a trapped gas of N bosonic polar molecules under dual microwaves at temperature T . In second-quantized form, the total Hamiltonian of the system reads

$$H = \int d\mathbf{r} \left[\frac{\hbar^2}{2M} \nabla \hat{\psi}^\dagger(\mathbf{r}) \nabla \hat{\psi}(\mathbf{r}) + U(\mathbf{r}) \hat{\psi}^\dagger(\mathbf{r}) \hat{\psi}(\mathbf{r}) \right] + \frac{1}{2} \int d\mathbf{r} d\mathbf{r}' V(\mathbf{r} - \mathbf{r}') \hat{\psi}^\dagger(\mathbf{r}) \hat{\psi}^\dagger(\mathbf{r}') \hat{\psi}(\mathbf{r}') \hat{\psi}(\mathbf{r}), \quad (1)$$

where M is the molecular mass, $\hat{\psi}(\mathbf{r})$ is the field operator, and $U(\mathbf{r}) = M[\omega_\perp^2(x^2 + y^2) + \omega_z^2 z^2]/2$ is the trapping potential with $\omega_\perp$ and ω_z being the transverse and axial trap frequencies, respectively. The interaction potential between two MSPMs takes the form [43]

$$V(\mathbf{r}) = \frac{C_6}{r^6} \left[\sin^4 \theta + \frac{w_1}{w_2} \sin^2 \theta \cos^2 \theta + \frac{w_0}{w_2} (3 \cos^2 \theta - 1)^2 \right] + \frac{C_3}{r^3} (3 \cos^2 \theta - 1), \quad (2)$$

where $C_6 (> 0)$ is the strength of the shielding potential, θ is the polar angle of the relative coordinate $\mathbf{r}$, and $w_{m=0,1,2}$ quantify the contributions from the angular momentum partial waves $|Y_{2m}(\hat{\mathbf{r}})|^2$, respectively. The first term on the right-hand side of Eq. (2) is always positive, ensuring that the shielding potential is repulsive in all directions. Additionally, C_3 is the strength of the negated long-range DDI. We emphasize that all interaction parameters in Eq. (2) are determined by the permanent dipole moment d of the molecules, as well as by the Rabi frequencies (Ω_+ , Ω_π) and detunings (δ_+ , δ_π) of the σ^+ - and π -polarized microwave fields, where the detunings are defined as the molecular transition frequencies minus the microwave frequencies. As demonstrated in Ref. [43], the approximate analytic expressions for all interaction parameters yield scattering properties with high precision, validating their applicability to the study of many-body physics.

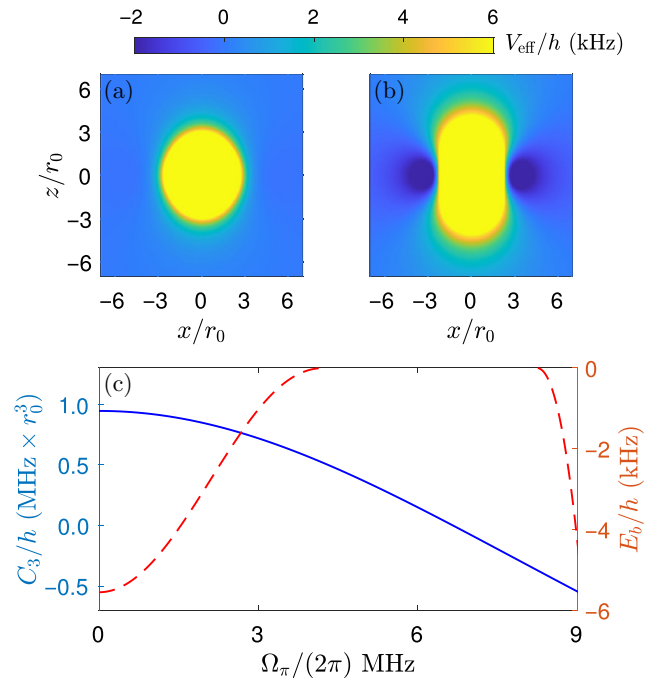


FIG. 1. Effective potentials in the x - z plane for $\Omega_\pi = 2\pi \times 6.5$ MHz (a) and $2\pi \times 5.8$ MHz (b), respectively. (c) Dipolar interaction strength and binding energy as functions of Ω_π .

As a concrete example, we consider NaCs molecules with a permanent dipole moment $d = 4.6$ Debye [60]. Following the experimental setup in Ref. [11], we assume the Rabi frequency and detuning of the σ^+ -polarized microwave field to be $\Omega_+ = 2\pi \times 7.9$ MHz and $\delta_+ = -2\pi \times 8$ MHz (blue detuning), respectively. Additionally, the detuning of the π -polarized microwave field is fixed at $\delta_\pi = -2\pi \times 10$ MHz (blue detuning), leaving Ω_π as the sole free parameter for tuning the inter-molecular potential. In Figs. 1(a) and 1(b), we present the cylindrically symmetric interaction in Eq. (2) in the x - z plane for two Rabi frequencies: $\Omega_\pi = 2\pi \times 6.5$ MHz and $2\pi \times 5.8$ MHz. As shown by the solid blue curve in Fig. 1(c), the effective dipolar interaction strength C_3 decreases monotonically with increasing Ω_π , and is canceled at $\Omega_\pi = 2\pi \times 6.66$ MHz. When Ω_π is reduced below this value, $C_3 > 0$, and the long-range attraction in the x - y plane is enhanced, as shown in Figs. 1(a) and 1(b). Conversely, increasing Ω_π beyond this point changes the sign of $C_3 < 0$, leading to attractive interactions along the z -direction and repulsive interactions in the x - y plane.

To identify a stable regime for exploring many-body physics at finite temperatures, we first compute the binding energy E_b of a tetramer composed of two molecules. We find that a tetramer bound state, with the wave function localized in the x - y plane (along the z -axis), emerges when a field-linked resonance occurs at $\Omega_\pi = 2\pi \times 4.22$ MHz ($\Omega_\pi = 2\pi \times 8.24$ MHz) [43]. The corresponding binding energy is shown by the dashed red curve in Fig. 1(c), where no bound state exists between $\Omega_\pi = 2\pi \times 4.22$ MHz and $2\pi \times 8.24$ MHz. Since three-body losses become significant near resonance, throughout this work, we focus on the range of the Rabi frequency $\Omega_\pi \in 2\pi \times [5.75, 6.5]$ MHz away from field-linked resonances, within which C_3 remains non-negative [11,61]. Notably, at $\Omega_\pi = 2\pi \times 5.8$ MHz the dipolar interaction becomes sufficiently strong. Therefore, we choose $\Omega_\pi = 2\pi \times 6.5$ MHz and $2\pi \times 5.8$ MHz as representative Rabi frequencies for weak and strong dipolar interactions, respectively. Finally, following the experimental conditions, the axial trap frequency is set to $\omega_z = 2\pi \times 59$ Hz. Unless otherwise specified, the radial frequency is chosen as $\omega_\perp = 2\pi \times 33.6$ Hz, such that it matches the geometric average of the radial trap frequencies in the experiment [11].

To investigate the finite-temperature many-body physics of this strongly correlated molecular gas under realistic experimental conditions, we employ the path integral Monte Carlo method based on the worm algorithm, circumventing the need for mean-field approximations or uncontrolled expansions.

III. PHASE DIAGRAM

In this section, we explore the many-body quantum phases in MSPMs at finite temperatures. Figure 2(a) displays the phase diagram in the Ω_π - T parameter plane

for $N = 200$, with two representative Rabi frequencies, $\Omega_\pi = 2\pi \times 6.5$ MHz and $2\pi \times 5.8$ MHz, marked by blue and red stars along the horizontal axis. The dashed line denotes the critical temperature T_c for BECs, determined using the criterion $f_c(T_c) \approx 5\%$. Here, $f_c(T) \equiv N_0(T)/N$ is the condensate fraction, where N_0 is the largest eigenvalue of the first-order correlation function $G_1(\mathbf{r}, \mathbf{r}') \equiv \langle \hat{\psi}^\dagger(\mathbf{r}) \hat{\psi}(\mathbf{r}') \rangle$ computed in the Appendix A. While the choice of the threshold condensate fraction for BECs is somewhat arbitrary, slight variations in this threshold only lead to quantitative changes in T_c . Since the BEC critical temperature T_c also depends on the external trap frequency, when determining T_c , we choose $\omega_\perp = 2\pi \times 33.6$ Hz. Notably, T_c increases monotonically as Ω_π decreases, a trend that will be explained in detail through an analysis of the inter-molecular potential in Sec. IV.

The solid line in Fig. 2(a) marks the boundary between the EG and SBG phases. Since the two-body interaction considered here does not support two-body bound states,

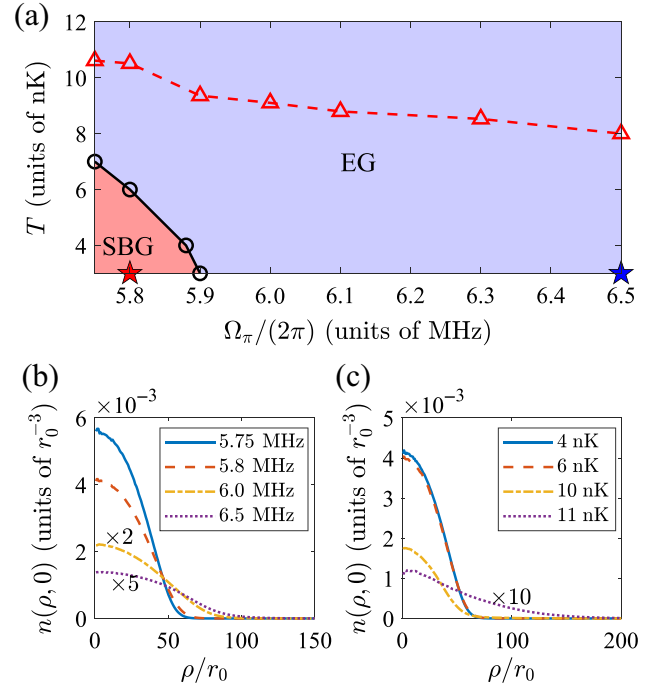


FIG. 2. (a) Phase diagram on the Ω_π - T plane. The dashed line denotes the critical temperature T_c for the BEC transition and the solid line indicates the critical temperature T_{sb} for the SBG phase. Two representative Rabi frequencies $\Omega_\pi = 2\pi \times 6.5$ MHz and $2\pi \times 5.8$ MHz are labeled by blue and red stars along the horizontal axis. T_c is determined at $f_c = 5\%$. T_{sb} is identified as the highest temperature at which the system remains self-bound in equilibrium without a radial trapping potential. (b) Radial density profiles at $T = 4$ nK for different Ω_π 's. (c) Radial density profiles at $\Omega_\pi = 2\pi \times 5.8$ MHz for different T 's. To improve visibility, some density profiles in (b) and (c) are scaled by appropriate factors. All results presented here are for a system of $N = 200$ molecules.

as shown in Fig. 1(c) and Ref. [43], the SBG phase arises purely from many-body effects. The solid line can also be interpreted as a critical temperature T_{sb} , such that for a given N and Ω_π , the SBG phase forms only when $T < T_{\text{sb}}$. We remark that the boundary T_{sb} does not depend on the radial confinement. Here, to accurately identify the SBG phase, we determine T_{sb} by turning off the radial trap entirely and checking whether the gas remains self-bound in thermal equilibrium.

Within the parameter regime $\Omega_\pi \in 2\pi \times [5.75, 6.5]$ MHz explored in our calculations, we find that $T_{\text{sb}} < T_c$, as the interaction strengths considered here are insufficient to stabilize the SBG phase at higher temperatures. However, as Ω_π is further decreased, enhancing the dipolar interaction, the self-bound transition temperature T_{sb} increases and may eventually surpass the BEC critical temperature T_c . In this case, as the temperature decreases, the system would first form SBG and then undergo the BEC transition. Nevertheless, for smaller Ω_π near the shape resonance at $2\pi \times 4.22$ MHz, the NaCs molecular gas becomes unstable due to significant three-body losses. The possibility of achieving $T_{\text{sb}} > T_c$ could be explored with other polar molecular species that remain stable in this strong attractive regime.

In Figs. 2(b) and 2(c), we compare the radial density profiles for different values of Ω_π at $T = 4$ nK and for different temperatures at fixed $\Omega_\pi = 2\pi \times 5.8$ MHz, where $r_0 = 10^3 a_B$ with a_B denoting the Bohr radius. The density distributions along the z -axis are provided in Appendix B. To ensure a consistent comparison between the EG and SBG phases, we present the density profiles with an external radial trap of frequency $\omega_\perp = 2\pi \times 33.6$ Hz. Although the radial size of the gas changes abruptly across the T_{sb} boundary in free space, this distinction is obscured in the presence of radial confinement. For example, as shown in Fig. 2(c), the trapped gas at $T = 10$ nK shrinks abruptly just below the BEC transition temperature of 10.5 nK, rather than at the EG-to-SBG transition around $T_{\text{sb}} \sim 6$ nK. In particular, the radial size of the trapped gas at $T = 10$ nK in the EG phase is comparable in magnitude to that in the SBG phase at 6 nK, indicating that the external potential blurs the boundary between the two phases when using the cloud size as a diagnostic. In Sec. V, we propose a robust method to determine the same transition temperature T_{sb} as in free space, even when the radial trap is present.

IV. THERMODYNAMIC TRANSITION

To gain deeper insight into the thermodynamic phase transition, we analyze the properties of the quantum phases in this section. We plot the temperature dependence of the condensate fraction (blue lines) for the representative Rabi frequencies $\Omega_\pi = 2\pi \times 5.8$ MHz and $2\pi \times 6.5$ MHz in Fig. 3(a). Remarkably, we observe that for $\Omega_\pi = 2\pi \times$

6.5 MHz, $f_c \approx 0.7$ and 0.9 at $T = 4$ nK and 2 nK, respectively, in agreement with the experimental measurement [11] and the cluster expansion result [43]. For comparison, we also plot f_c as a function of T (dashed black line) for a non-interacting gas of $N = 200$ particles in the same harmonic potential. Due to finite-size effects [62], the dashed black line deviates from the thermodynamic limit result, $1 - (T/T_c^{(0)})^3$, where the BEC critical temperature $T_c^{(0)} = 10.69$ nK is calculated [63] for free bosons in the trap with frequencies $\omega_\perp = 2\pi \times 33.6$ Hz and $\omega_z = 2\pi \times 59$ Hz. Notably, the critical temperature for $\Omega_\pi = 2\pi \times 5.8$ MHz ($2\pi \times 6.5$ MHz) is higher (lower) than that of the non-interacting gas. This behavior can be understood as follows: when the two-body interaction is attractive, it is energetically favorable for molecules to occupy the same quantum state, thereby facilitating condensation. Roughly speaking, the stronger the attraction,

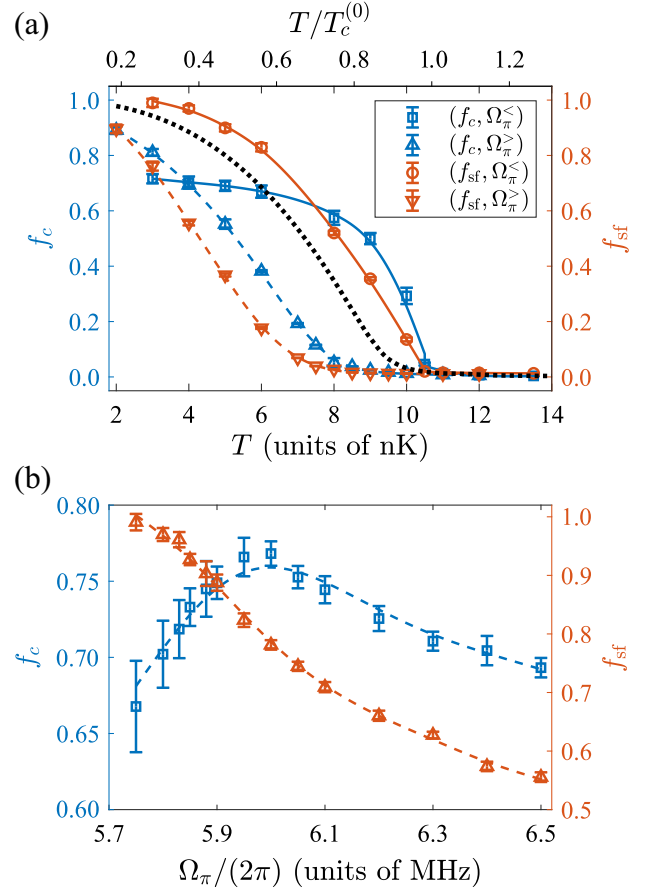


FIG. 3. (a) Condensate fraction f_c (blue, left axis) and superfluid fraction f_{sf} (red, right axis) as functions of T for $\Omega_\pi = \Omega_\pi^< \equiv 2\pi \times 5.8$ MHz (solid lines) and $\Omega_\pi^> \equiv 2\pi \times 6.5$ MHz (dashed lines). The dotted line denotes the condensate fraction as a function of T for a non-interacting gas. (b) Condensate fraction (left axis) and superfluid fraction (right axis) as functions of Ω_π at $T = 4$ nK. For both (a) and (b), the system consists of $N = 200$ molecules.

the higher the critical temperature. Conversely, when the inter-molecular potential is repulsive, the critical temperature decreases, explaining why the critical temperature for $\Omega_\pi = 2\pi \times 6.5$ MHz is lower than that of the non-interacting gas. This result is consistent with that observed in atomic BECs [64–68].

Another frequently used physical quantity closely relating to the BEC transition is the superfluid fraction $f_{\text{sf}} \equiv n_{\text{sf}}/n$, where n_{sf} and n are the superfluid and total densities of a homogeneous gas, respectively. For a finite system, f_{sf} can be evaluated by examining its response to an imposed rotation. This calculation is particularly convenient in the path integral Monte Carlo, as the total superfluid response along the axis of rotation is proportional to the square of the total projected area of the imaginary-time paths [69]. Physically, f_{sf} is primarily determined by the behavior of low-lying phonon modes, e.g., the sound velocity.

The red lines in Fig. 3(a) show the temperature dependence of the superfluid fraction for different values of Ω_π . As can be seen, f_{sf} monotonically increases to unity as T decreases. Notably, the onset of a nonzero f_{sf} coincides with that of f_c , indicating that, in three-dimensional systems, the formation of BECs implies superfluidity. Indeed, the appearance of BECs signifies a change in the low-energy dispersion from the quadratic dispersion of free particles to the linear dispersion of phonons, which results in a finite Landau critical velocity and, consequently, superfluidity. We note that in lower dimensions, e.g., in two-dimensional systems, superfluidity emerges below the Berezinskii-Kosterlitz-Thouless transition temperature [70,71].

Furthermore, we explore the dependence of the condensate and superfluid fractions on the interaction strength (Ω_π). Figure 3(b) shows f_c and f_{sf} as functions of Ω_π for $T = 4$ nK and $N = 200$. Unlike the temperature dependence observed in Fig. 3(a), here f_c and f_{sf} may exhibit contrasting behaviors with respect to Ω_π . Specifically, while f_{sf} increases monotonically as Ω_π decreases, f_c decreases in the small Ω_π regime. To understand this, we note that in the weak DDI regime corresponding to large Ω_π [cf. Fig. 1(c)], the gas has a lower density. Thus, decreasing Ω_π enhances the attraction, leading to a higher condensate fraction. However, in the strong dipolar interaction regime corresponding to small Ω_π [cf. Fig. 1(c)], the gas reaches higher densities, causing the mean inter-particle distance to become comparable to the size of the repulsive shielding core. This leads to a more pronounced antibunching effect, as illustrated by the density-density correlation function in the x - y plane:

$$G_2(\rho) \equiv \frac{\langle \sum_{i \neq j} \int dz \int dz' \delta(\mathbf{r}_i - \mathbf{r}) \delta(\mathbf{r}_j) \rangle}{\bar{n}(\rho) \bar{n}(0)}, \quad (3)$$

where $\mathbf{r}_i$ is the position of the particle i , and $\bar{n}(\rho) = \int dz n(\mathbf{r})$ is the integrated density distribution. As shown in

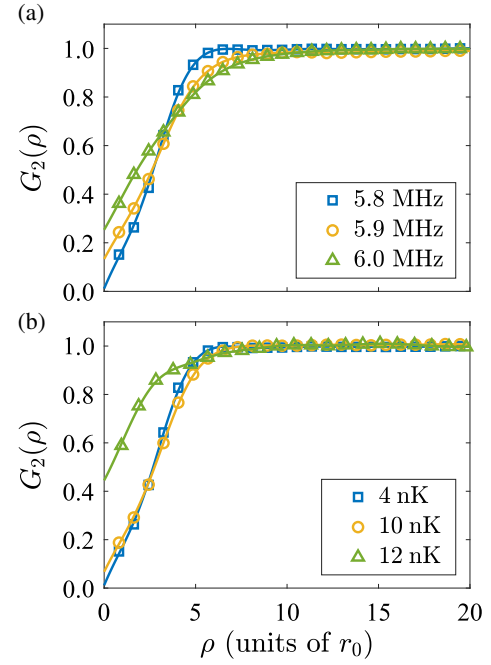


FIG. 4. Density-density correlation functions for $N = 200$ molecules: (a) $T = 4$ nK; (b) $\Omega_\pi = 2\pi \times 5.8$ MHz.

Fig. 4, when the dipolar interaction strength increases (or the temperature decreases), $G_2(\rho)$ exhibits stronger antibunching behavior. This indicates that the possibility of detecting two particles within the distance $\rho \sim r_0$ in the x - y plane decreases, and more depletion is generated. As Ω_π is further decreased, the increased depletion suppresses the establishment of phase coherence within the length scale r_0 , thereby reducing the condensate fraction.

Thus, we conclude that in the weak DDI regime, the density remains low and the condensate fraction f_c is primarily governed by the long-range dipolar interaction. In contrast, in the strong DDI regime, the density increases and f_c is dominated by the short-range shielding potential. The non-monotonic behavior of f_c can be inferred from the momentum distribution shown in Appendix A, as measured in time-of-flight (TOF) experiments [11]. We note that in the EG phase, the gas expands freely upon release of the trap in a TOF experiment. In contrast, in the SBG phase, expansion can be enabled by simultaneously releasing the trap and turning off the dipolar interactions ($C_3 = 0$) achieved by rapidly increasing the Rabi frequency Ω_π to $2\pi \times 6.66$ MHz. Turning to the superfluid fraction, f_{sf} is closely related to the properties of low-lying excitations, e.g., the phonon velocity, which depend on the long-range behavior governed by the dipolar interaction. Since the dipolar interaction strength C_3 increases monotonically as Ω_π decreases [cf. Fig. 1(c)], the superfluid fraction f_{sf} also varies monotonically with Ω_π , as shown in Fig. 3(b).

V. EG-TO-SBG TRANSITION

As analyzed in Secs. III and IV, the EG-to-SBG transition is obscured by the external trap and is not directly reflected in f_c or f_{sf} . In this section, we propose a robust signature for identifying the EG-to-SBG transition point in the presence of the trap.

We examine the geometric properties of the gases in Fig. 5, such as its “volume” $v \equiv \sqrt{\langle x^2 \rangle \langle y^2 \rangle \langle z^2 \rangle}$ and aspect ratio $\gamma \equiv \sqrt{\langle x^2 \rangle / \langle z^2 \rangle}$. Although the EG-to-SBG transition boundary T_{sb} is determined in free space, independent of the radial trap, the presence of radial confinement obscures the phase boundary, as the geometric properties of the gas strongly depend on $\omega_\perp$. Here, to make a clear comparison, in addition to the trap with frequency $\omega_\perp = 2\pi \times 33.6$ Hz, we also consider a weaker radial trap with $\omega_\perp = 2\pi \times 10.6$ Hz [72]. In Fig. 5(a), the blue lines show the volume v as a function of Ω_π for two different values of $\omega_\perp$. Since a smaller Rabi frequency enhances the attractive interaction, the volume v monotonically decreases as Ω_π is reduced. Particularly, when Ω_π becomes small, e.g., $2\pi \times 5.75$ MHz, the two volumes converge to approximately the same value, signaling the formation of SBG. In fact, as long as the radial size of SBG is smaller than that of the external trap, the volumes of the gases corresponding to different radial trap frequencies should converge for control parameters deep within the SBG phase. However, it is important to note that, due to the radial trap, the point where two volumes converge does not coincide with the EG-to-SBG transition boundary denoted by the solid black curve in Fig. 2(a).

Next, we consider the aspect ratio of the gases. To this end, we note that the aspect ratio γ is, $\omega_z/\omega_\perp$ for thermal gases or depleted particles with high energies, and $\sqrt{\omega_z/\omega_\perp}$ for a pure condensate occupying the low-energy state. Therefore, for $\omega_z/\omega_\perp > 1$, γ decreases as the condensate fraction increases. In the presence of DDI, however, the gas tends to stretch (measured in terms of aspect ratio) along the attractive (radial) directions to minimize the interaction energy. As a result, γ increases with the interaction strength for a negated DDI. In summary, γ is determined by the competition between condensation and interaction: condensation (interaction) favors a smaller (larger) γ .

The red lines in Fig. 5(a) show the Ω_π dependence of γ for two values of $\omega_\perp$ with $T = 4$ nK. As Ω_π decreases, γ for the trap with $\omega_\perp = 2\pi \times 33.6$ Hz increases monotonically, indicating that DDI dominates the behavior of γ . This is expected, as the higher gas density in a stronger radial trap enhances the role of interactions. In contrast, for $\omega_\perp = 2\pi \times 10.6$ Hz, the Ω_π dependence of γ becomes more complex and non-monotonic. Starting from the weak DDI regime $\Omega_\pi = 2\pi \times 6.5$ MHz, γ initially decreases upon lowering Ω_π , because the condensate fraction increases and governs the behavior of γ in

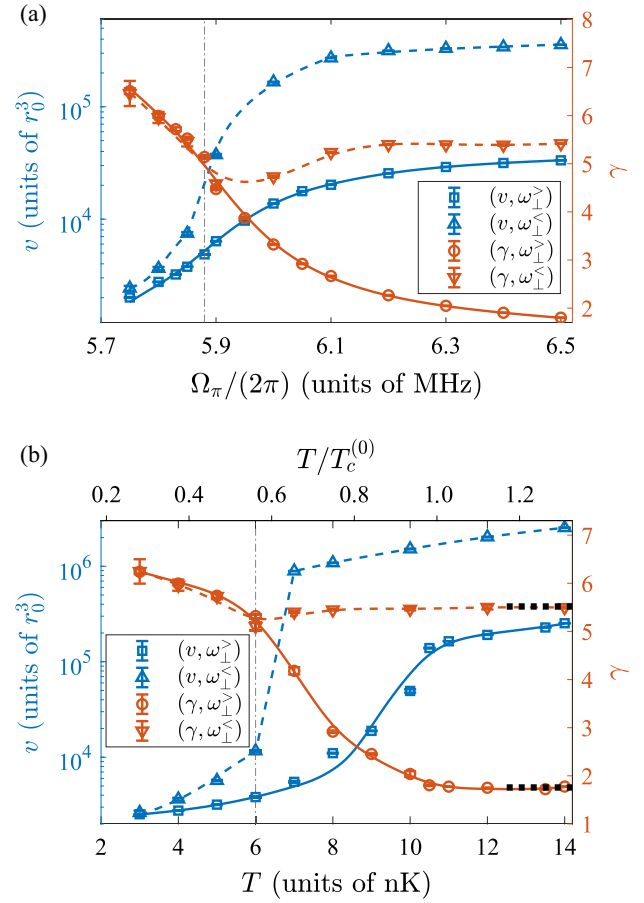


FIG. 5. (a) Gas volume v (blue, left axis) and aspect ratio γ (red, right axis) as functions of Ω_π at $T = 4$ nK. (b) Gas volume (left axis) and aspect ratio (right axis) as functions of T at $\Omega_\pi = 2\pi \times 5.8$ MHz. Two short dotted lines denote the aspect ratios of trapped non-interacting thermal gases. For both (a),(b), the number of molecules is $N = 200$. The solid (dashed) lines correspond to a higher (lower) radial trap frequency $\omega_\perp = \omega_\perp^+ \equiv 2\pi \times 33.6$ Hz ($\omega_\perp = \omega_\perp^- \equiv 2\pi \times 10.6$ Hz). Black dot-dashed lines indicate the EG-to-SBG transition boundaries.

the low-density regime. However, when Ω_π drops below approximately $2\pi \times 5.9$ MHz, the gas density becomes sufficiently large in the SBG phase, and the repulsive shielding potential enhances antibunching effects, thereby suppressing the condensate fraction. As a result, the strong DDI becomes the dominant factor determining the aspect ratio, and γ begins to increase as Ω_π is further reduced. The behavior of γ thus reflects the non-monotonic variation of condensate fraction controlled by Ω_π . Furthermore, γ for different values of $\omega_\perp$ converge at sufficiently small Ω_π , similar to the behavior of v . However, unlike v , the value of Ω_π at which γ converges for different $\omega_\perp$ aligns with the EG-to-SBG transition boundary in Fig. 2(a), suggesting that γ can serve as a reliable indicator of

the EG-to-SBG transition even in the presence of radial confinement.

To gain deeper insight into the geometric properties of the gas, we plot in Fig. 5(b) the dependence of v and γ on T for two radial trap frequencies, $\omega_{\perp} = 2\pi \times 33.6$ Hz and $2\pi \times 10.6$ Hz. As expected, the volume v decreases monotonically as T is lowered. Similarly, the two blue curves for the volume converge to approximately the same value (distinct from the transition boundary T_{sb}) in the SBG phase. Regarding the aspect ratio, distinct behaviors are observed for the two trap frequencies, as shown by the red lines in Fig. 5(b). In the high-temperature limit, the aspect ratios agree with those of non-interacting thermal gases, indicated by the black dashed lines. For $\omega_{\perp} = 2\pi \times 33.6$ Hz, γ increases monotonically as T is lowered, as the gas density is sufficiently high, with the aspect ratio primarily governed by interactions. In contrast, for $\omega_{\perp} = 2\pi \times 10.6$ Hz, the temperature dependence of γ exhibits non-monotonic behavior. In the high-temperature regime ($T > 8$ nK), the gas density is low, and γ remains nearly constant as the temperature decreases. Between 8 and 6 nK, γ experiences a slight decrease, which can be attributed to an increase in the condensate fraction. As T is further lowered, the gas density increases in the SBG phase, enhancing the role of DDI, and γ begins to increase. Again, the EG to SBG transition boundary identified by the convergence of γ is more accurately determined than that obtained via the convergence of v . This highlights the sensitivity of the aspect ratio to the underlying strong-correlation mechanism governing the EG-to-SBG transition.

VI. DISCUSSIONS AND CONCLUSIONS

In conclusion, we have employed the path integral Monte Carlo method with the worm algorithm to investigate the finite-temperature phase diagram of bosonic MSPMs, uncovering both the EG and SBG phases. These phases exhibit many-body correlations that go beyond what is observed in atomic BECs. The interplay between long-range DDI and short-range shielding potentials gives rise to striking molecular antibunching effects and intriguing behaviors in both the condensate and superfluid fractions. Notably, in the SBG phase, MSPMs display a low condensate fraction and a high superfluid fraction, similar to the strong correlations seen in superfluid ^4He . This positions MSPMs as a promising new platform for simulating strongly correlated systems. In addition, we demonstrate that while radial confinement may obscure the phase boundary as measured by the gas volume, the aspect ratio of the density profile offers a robust and precise tool to identify the EG-to-SBG transition, even in the presence of a trapping potential. This highlights the significant role of DDI and condensate fractions in this transition.

Our numerically exact simulations provide a solid and quantitative framework for understanding MSPMs under

realistic experimental conditions. These results offer practical insights for experimental efforts aimed at realizing BECs of polar molecules, such as NaRb, CsAg, BaF, and CaF. Additionally, these upcoming experiments will help benchmark effective potentials, validate theoretical models, and refine our understanding of molecular interactions. Furthermore, our study opens avenues for future experimental and theoretical investigations into exotic many-body phases. For instance, exploring alternative molecular species with stronger DDI could facilitate the realization of SBG at higher temperatures $T_{\text{sb}} > T_c$. More importantly, the creation of self-bound droplet arrays and intriguing supersolid phases [73] may be achievable by enhancing DDI through tuning the Rabi frequency and the ellipticity angle of the microwave field [8,9], a promising direction that warrants further theoretical and experimental exploration.

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DATA AVAILABILITY

No data were created or analyzed in this study.

APPENDIX A: FIRST-ORDER CORRELATION FUNCTIONS IN NON-UNIFORM SYSTEMS

In this Appendix, we present a detailed computation of the first-order correlation function, $G_1(\mathbf{r}, \mathbf{r}') = \langle \hat{\psi}^\dagger(\mathbf{r}) \hat{\psi}(\mathbf{r}') \rangle$, in a non-uniform system using the path integral Monte Carlo combined with the worm algorithm. The condensate fraction is determined by the largest eigenvalue of $G_1(\mathbf{r}, \mathbf{r}')$, and the momentum distribution is obtained from its Fourier transform.

The first-order correlation function, $G_1(\mathbf{r}, \mathbf{r}') = \langle \hat{\psi}^\dagger(\mathbf{r}) \hat{\psi}(\mathbf{r}') \rangle$, satisfies the following properties: (1) Normalization condition: $\text{Tr} G_1 \equiv \int d\mathbf{r} G_1(\mathbf{r}, \mathbf{r}) = N$, where N denotes the total particle number. (2) Positivity: The eigenvalues of G_1 are non-negative. (3) Hermiticity: $G_1(\mathbf{r}, \mathbf{r}') = G_1^*(\mathbf{r}', \mathbf{r})$. The eigenvalues λ_n of G_1 are determined by the eigenequation

$$\int d\mathbf{r}' G_1(\mathbf{r}, \mathbf{r}') \phi_n(\mathbf{r}') = \lambda_n \phi_n(\mathbf{r}), \quad (\text{A1})$$

where the eigenvalues are sorted in descending order, $\lambda_0 \geq \lambda_1 \geq \lambda_2 \geq \dots \geq 0$, and satisfy the normalization condition

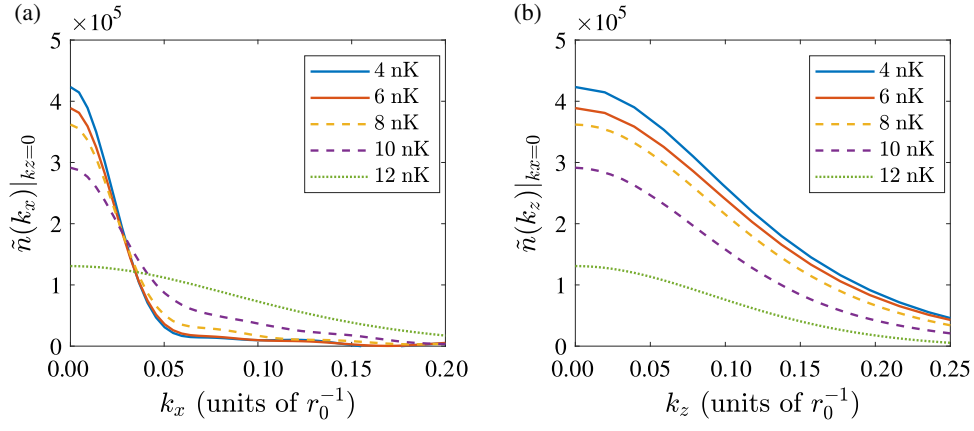


FIG. 6. Integrated momentum distributions: (a) $\tilde{n}(k_x)|_{k_z=0} \equiv \tilde{n}(k_x, k_z = 0)$; (b) $\tilde{n}(k_z)|_{k_x=0} \equiv \tilde{n}(k_x = 0, k_z)$. Here, $\Omega_\pi = 2\pi \times 5.8$ MHz, $N = 200$, and trap frequencies $\omega_\perp = 2\pi \times 33.6$ Hz, $\omega_z = 2\pi \times 59$ Hz. For this parameter set, $T = 4$ nK and $T = 6$ nK correspond to the SBG phase, while higher temperatures correspond to the EG phase. The BEC transition occurs at approximately $T = 10.5$ nK.

$\sum_i \lambda_i = N$. The condensate fraction is then defined as $f_c = N_0/N$, where $N_0 \equiv \lambda_0$.

To efficiently compute G_1 through sampling, we first select an appropriate set of basis functions:

$$\begin{aligned}
 g_{n_\rho, m, n_z}^{(e)}(\rho, \varphi, z) &= \frac{1}{\sqrt{\mathcal{N}_{n_\rho m n_z}^{(1)}}} \\
 &\times \cos(m\varphi) \psi_{n_\rho, m}(\rho/\xi_\rho) \phi_{n_z}(z/\xi_z), \quad m = 0, 1, 2, \dots, \\
 g_{n_\rho, m, n_z}^{(o)}(\rho, \varphi, z) &= \frac{1}{\sqrt{\mathcal{N}_{n_\rho m n_z}^{(2)}}} \\
 &\times \sin(m\varphi) \psi_{n_\rho, m}(\rho/\xi_\rho) \phi_{n_z}(z/\xi_z), \quad m = 1, 2, 3, \dots
 \end{aligned} \tag{A2}$$

Here, m is the quantum number of the projected angular momentum along the z -direction, and $\mathbf{r} = (\rho, z, \varphi)$ with ρ , φ , and z representing the radial coordinate, azimuthal

angle, and axial coordinate, respectively. The functions $\psi_{n_\rho, m}(x) = x^{|m|} e^{-x^2/2} L_{n_\rho}^{(|m|)}(x^2)$ and $\phi_{n_z}(x) = e^{-x^2/2} H_{n_z}(x)$ are eigenfunctions of the two- and one-dimensional harmonic oscillators, respectively, expressed in terms of the generalized Laguerre polynomials $L_n^{(\alpha)}(x)$ and Hermite polynomials $H_n(x)$. The characteristic lengths ξ_ρ and ξ_z should be chosen to match the spatial widths of the density profile in the x - y plane and along the z -axis, ensuring rapid convergence as the cutoffs for n_ρ and n_z are increased. The corresponding normalization factors are

$$\begin{aligned}
 \mathcal{N}_{n_\rho m n_z}^{(1)} &= s_m \pi^{3/2} \frac{\xi_\rho^2 (n_\rho + |m|)!}{2n_\rho!} 2^{n_z} n_z! \xi_z, \\
 \mathcal{N}_{n_\rho m n_z}^{(2)} &= \pi^{3/2} \frac{\xi_\rho^2 (n_\rho + |m|)!}{2n_\rho!} 2^{n_z} n_z! \xi_z,
 \end{aligned} \tag{A3}$$

where $s_m = 2$ for $m = 0$ and $s_m = 1$ for $m > 0$.

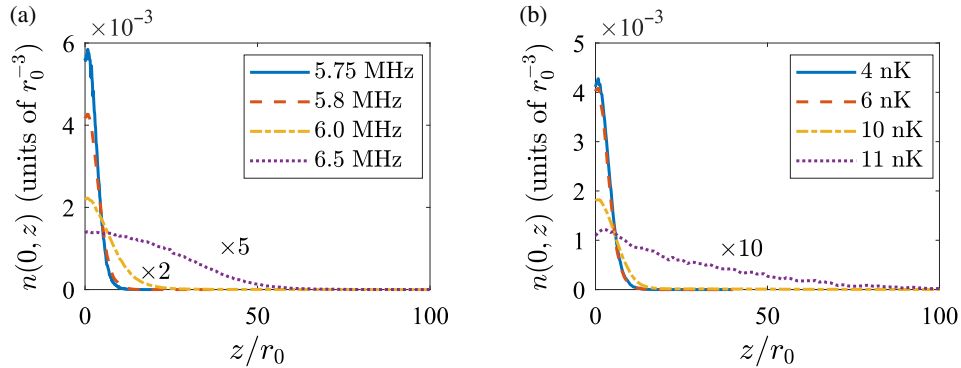


FIG. 7. (a) Density profiles along the z -axis at $T = 4$ nK for different Ω_π 's. (b) Density profiles along the z -axis at $\Omega_\pi = 2\pi \times 5.8$ MHz for different T 's. To improve visibility, some density profiles in (a),(b) are scaled by appropriate factors. All results presented here are for a system of $N = 200$ molecules and the radial trap frequency $\omega_\perp = 2\pi \times 33.6$ Hz.

By projecting $G_1(\mathbf{r}, \mathbf{r}')$ onto the space spanned by the orthonormal basis $g_{n_\rho, m, n_z}^{(s=e, o)}(\rho, \varphi, z)$, we obtain

$$\begin{aligned}
 (G_1)_{(n_\rho m n_z), (n'_\rho m' n'_z)}^{(s, s')} &= \int d\mathbf{r} \int d\mathbf{r}' g_{n_\rho, m, n_z}^{(s)}(\rho, \varphi, z) G_1(\mathbf{r}, \mathbf{r}') g_{n'_\rho, m', n'_z}^{(s')}(\rho', \varphi', z') \\
 &= \frac{\int d\mathbf{r} \int d\mathbf{r}' g_{n_\rho, m, n_z}^{(s)}(\rho, \varphi, z) \text{Tr}[e^{-\beta \hat{H}} \hat{\psi}^\dagger(\mathbf{r}) \hat{\psi}(\mathbf{r}')] g_{n'_\rho, m', n'_z}^{(s')}(\rho', \varphi', z')}{\text{Tr}[e^{-\beta \hat{H}}]} \\
 &= \frac{\int d\mathbf{r} \int d\mathbf{r}' \text{Tr}[e^{-\beta \hat{H}} \hat{\psi}^\dagger(\mathbf{r}) \hat{\psi}(\mathbf{r}')] g_{n_\rho, m, n_z}^{(s)}(\rho, \varphi, z) g_{n'_\rho, m', n'_z}^{(s')}(\rho', \varphi', z')}{\int d\mathbf{r} \int d\mathbf{r}' \text{Tr}[e^{-\beta \hat{H}} \hat{\psi}^\dagger(\mathbf{r}) \hat{\psi}(\mathbf{r}')] } \\
 &\quad \times \frac{\int d\mathbf{r} \int d\mathbf{r}' \text{Tr}[e^{-\beta \hat{H}} \hat{\psi}^\dagger(\mathbf{r}) \hat{\psi}(\mathbf{r}')] }{\text{Tr}[e^{-\beta \hat{H}}]}. \tag{A4}
 \end{aligned}$$

In this new basis, $G_1(\mathbf{r}, \mathbf{r}')$ can be efficiently computed using the worm algorithm. The first factor in the last equality of Eq. (A4) can be sampled in the off-diagonal sector (when the worm is present), while the second factor represents the relative probability between the diagonal and off-diagonal sectors.

For a system with cylindrical symmetry along the z -axis, G_1 is block diagonal, i.e., $m = m'$. By diagonalizing G_1 in different sectors m , we obtain the largest eigenvalue λ_0 and the condensate fraction f_c . The convergence of all results is verified by systematically increasing the cutoffs for n_ρ and n_z .

The momentum distribution is obtained from the Fourier transform of $G_1(\mathbf{r}, \mathbf{r}')$ as

$$\tilde{n}(\mathbf{k}) = \int d\mathbf{r} \int d\mathbf{r}' G_1(\mathbf{r}, \mathbf{r}') e^{-i\mathbf{k} \cdot (\mathbf{r} - \mathbf{r}')}.$$
 (A5)

The integrated momentum distribution $\int dk_y \tilde{n}(\mathbf{k}) / (2\pi)$ is shown in Fig. 6.

APPENDIX B: DENSITY PROFILES ALONG THE z -AXIS

In Fig. 7, we present the density distributions along the z -axis, complementing the radial density profiles shown in the main text. Similar to the radial density profiles, the width of the gas along the z -axis decreases as the Rabi frequency Ω_π and the temperature T are reduced. However, the sharp change in size does not coincide with the EG-to-SBG transition boundary T_{sb} , suggesting that the presence of the external trap smooths out the transition, as discussed in Secs. III and V.

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