Polaritons under Extensive Disordered Molecular Rotation in Optical Cavities

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This study investigates the dynamic behavior of polaritons in an optical cavity containing one million molecules, emphasizing the influence of molecular rotation and level disorder on the coupling between molecules and photons. Through rigorous theoretical simulations and numerical analyses, we systematically explore the formation and spectral characteristics of polaritons in this complex environment. Our findings reveal that the rotational motion of molecules significantly affects the electromagnetic field distribution within the cavity, leading to distinct alterations in polariton properties. Simultaneously, the presence of level disorder induces diverse energy level structures, influencing the energy distribution of polaritons. The comprehensive examination of these factors provides valuable insights into the intricate interplay between molecules and photons in large-scale cavity systems. This research not only advances the fundamental understanding of molecular-photon coupling but also offers theoretical guidance for practical applications in the design and exploration of optical cavities.

Introduction.—The interaction between matter (atoms, molecules, or solid materials) and the quantized electromagnetic field inside an optical cavity gives rise to a distinct set of states known as polariton states [1–3]. These states involve shared excitations between molecules and the cavity, influencing chemical reactivity [1, 3–14]. Describing them comprehensively is challenging due to the numerous light modes in the cavity and energetic, spatial, and orientational disorder.

Many theories simplify light by using a single cavity mode coupled to numerous quantum emitters [8, 15–32]. Recent research predicts changes in transport and relaxation based on disorder but often neglects practical factors like molecular rotation [15, 16]. The impact of molecular rotation on polaritons, especially in cavities with numerous molecules, remains unclear. As molecules rotate, the dynamic nature of light-matter coupling, exhibiting time-dependent oscillations, comes to the forefront. The periodically driven Hamiltonian has been studied using Floquet theory [33–36], and disorder's impact on polaritons has been explored [16, 37–41]. However, a comprehensive understanding of the dynamics of light and matter for quantum emitters driven by both Floquet and disorder is still lacking.

In this letter, we delve into a large cavity system comprising 10^6 molecules and one photon, considering molecular rotation and disorder. Through theoretical simulations and calculations, we investigate the dynamic behavior and spectral features of polaritons in this intricate setting. The introduction of molecular rotation and disorder induces shifts in the energy levels of polaritons,

impacting the oscillation period of photon-matter hybrid states. Increasing disorder leads to a rapid decay of these hybrid states, almost causing their disappearance. In the case of weak coupling, even though polaritons are not generated, molecular rotation is observed to extend the lifetime of the photon state.

Floquet Disordered Tavis-Cummings Model.—As shown in Fig. 1 (a) and (b), we consider that many molecules (10^6) are contained in a microcavity formed by two nearly 100% reflecting mirrors, involving the rotation of molecules. We adopt a singlemode Floquet-driven disordered Tavis-Cummings model, whose Hamiltonian is given as $\hat{H} = \hat{H}_M + \hat{H}_C + \hat{H}_{MC}$, where

$$\hat{H}_M = \sum_{j=1}^{N_M} \hbar\omega_j \hat{B}_j^\dagger \hat{B}_j, \ \ \hat{H}_C = \hbar\omega_c \hat{a}^\dagger \hat{a},$$

$$\hat{H}_{MC} = \sum_{j=1}^{N_M} g_c \cos(\Omega t)\hat{B}_j^\dagger \hat{a} + H.c. \tag{1}$$

The molecules j are described by bosonic operators $\hat{B}_j$. Here, the excitation energies $\hbar\omega_j$ are distributed according to a Gaussian function $P(\omega) = (1/\sqrt{\pi}\sigma)e^{-\hbar(\omega-\omega_m)^2/(2\sigma^2)}$, with center $E_M = \hbar\omega_m$ and disorder width σ. We define the range of the Gaussian distribution of excitation energies as ΔE. The light field is quantized by the photonic operators $\hat{a}$ (under the single mode approximation). The light-matter interaction in $\hat{H}_{MC}$ is given by $g_c \cos(\Omega t)$, where Ω means the molecular vibration frequency. When $2g_c\sqrt{N_M} \gg \Delta E$, there is a strong interaction between light and matter, and when $2g_c\sqrt{N_M} \ll \Delta E$, there is a weak interaction between light and matter. Due to the temporal periodicity of molecular rotations, we will use Floquet theory to treat
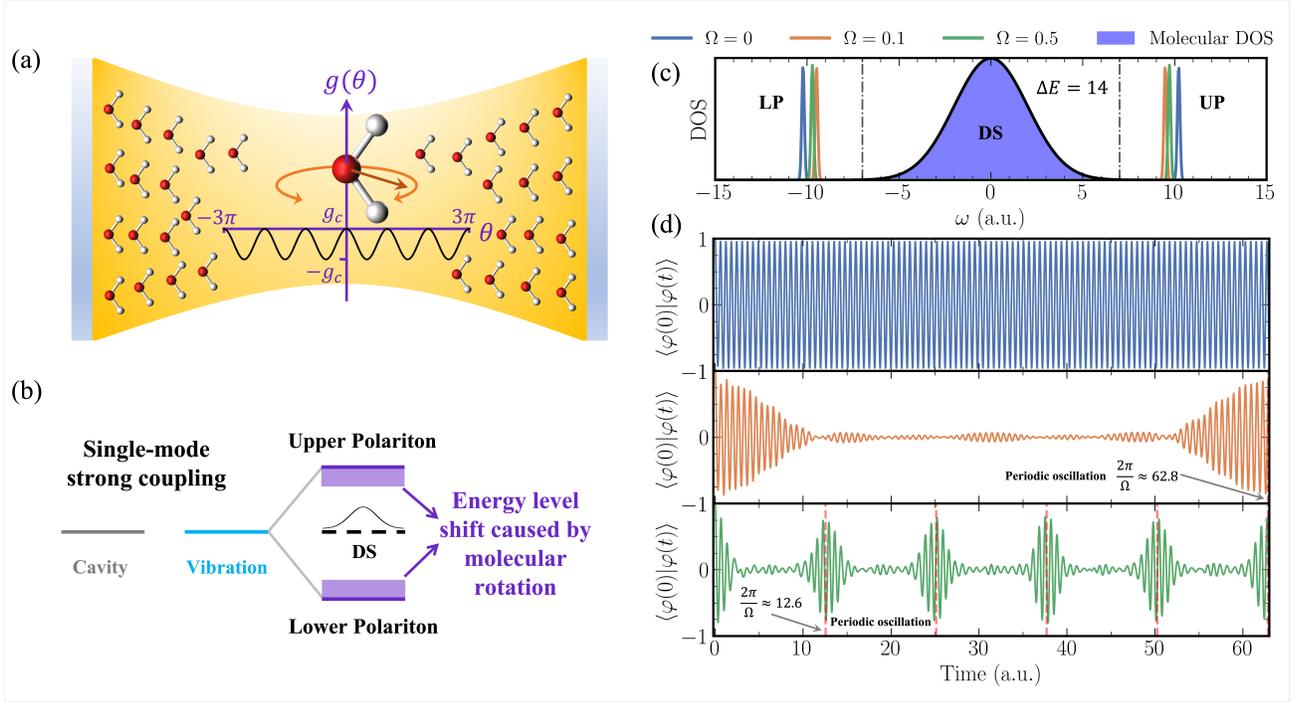
FIG. 1. (a) Schematic displays that many molecules (10^6) are contained in a microcavity formed by two nearly 100% reflecting mirrors, considering the rotation of molecules. The intensity of light-matter interactions, g, fluctuates within the range of $[-g_c, g_c]$ with the rotation angle θ of the molecule. (b) When a cavity resonance is tuned to match a molecular transition, it is possible to establish strong coupling, leading to two new "polariton" states split in energy proportionally to the coupling. The energy levels of these polaritons can be shifted by different molecular rotational angular velocities. The Gaussian distribution of dark states (DS) represents energy level disorder. (c) Normalized density of states (DOS) with respect to the energy frequency ω under different molecular rotational angular velocity Ω. The black dotted line represents the truncation position of the Gaussian distribution. ΔE represents the range of disordered molecular energy levels. Note that the amplitude of polariton and molecular DOS has been adjusted to display on the same graph. (d) Time evolution of correlation functions of photon-matter hybrid wavefunctions φ under different Ω, as defined in Eq. 9. After introducing molecular rotation, the oscillation period of the photon-matter hybrid states will depend on the molecular rotation period ($\frac{2\pi}{\Omega}$, red dash lines). Parameters: $\Omega = 0, 0.1, 0.5$, $k = 30$, $\hbar = 1$, $g_c = 0.01$, $\omega_c = \omega_m = 0$, $\sigma = 2$, $\gamma = 0.1$, $N_F = 3$, $\Delta t = 0.001$, $N_M = 10^6$, $N_C = 1$.

time-dependent Hamiltonian $\hat{H}(t)$ as time-independent $\hat{H}^F$:

$$\hat{H}^F = \sum_{n,m=-N_F}^{N_F} (\hat{H}_0 + n\hbar\Omega \otimes \hat{I})\delta_{nm} + \hat{H}_1(\frac{1}{2}\delta_{n,m+1} + \frac{1}{2}\delta_{n,m-1}) \tag{2}$$

where $\hat{H}_0$ means the diagonal part of $\hat{H}$ and $\hat{H}_1$ means the off-diagonal part of $\hat{H}$ removing $\cos(\Omega t)$. $\hat{I}$ is identity matrix. Dirac delta function δ is used. N_F is the number of Floquet levels.

We restrict the current investigation to molecular ro-

tational angular velocity and energetic disorder under a system of $N_M = 10^6$ molecules with $N_C = 1$ cavity mode.

Sparse Floquet Hamiltonian.—In dealing with such an extensive system, the primary obstacle shifts from computing speed to memory usage. To address this, and mindful of the sparsity within $\hat{H}^F$ whose dimension is $[(2N_F + 1)(N_M + N_C), (2N_F + 1)(N_M + N_C)]$, we opt for a memory-efficient approach. The entire matrix multiplication is founded on the non-zero elements using the Co-ordinate Format (COO) sparse matrix framework, identified as $\hat{H}_s^F$ whose total number of the non-zero elements is $[3 \times (2N_F + 1)(N_M + N_C)]$. This not only helps mitigate memory constraints but also streamlines the computational process.

Density of States (DOS).—We use the implicitly restarted Lanczos method to compute half $(k/2)$ eigenvalues (denoted as Λ) and their corresponding eigenvectors (denoted as U) from each end of the spectrum of

the sparse matrix H_s^F. This method is a wrapper for the ARPACK [42] SSEUPD and DSEUPD functions [43]. Then the DOS ρ of polaritons can be written as :

$$\rho(\omega) = Tr[\delta(\tilde{H} - \omega)U^\dagger P_C U] \tag{3}$$
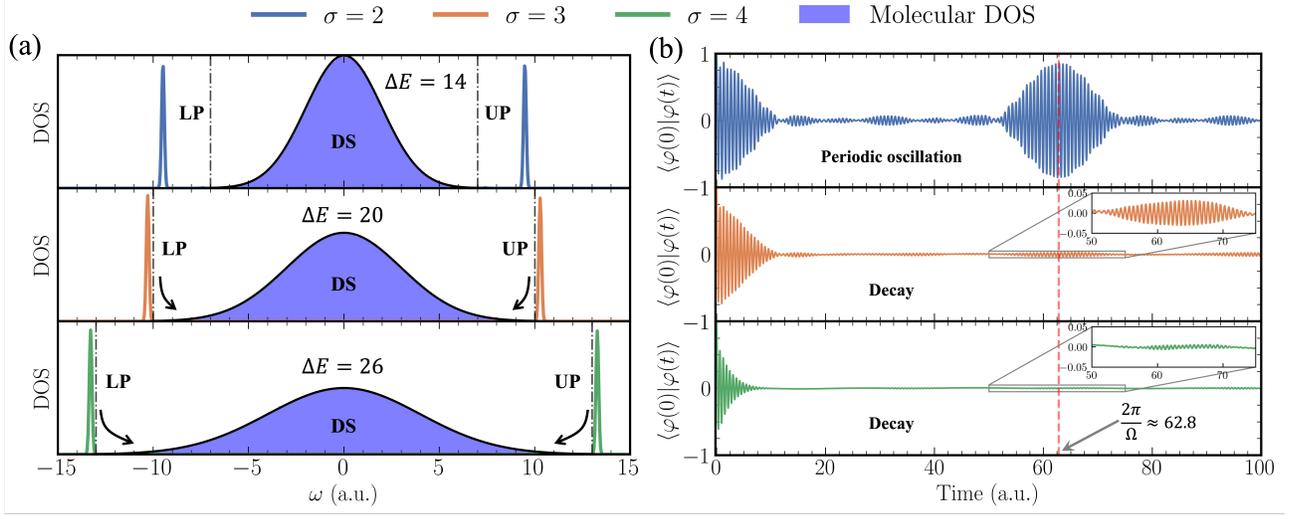
FIG. 2. (a) Normalized density of states (DOS) with respect to the energy frequency ω under different disorder σ. The black dotted line represents the truncation position of the Gaussian distribution. As the disorder increases, the polaritons will become closer to the dark states (DS). ΔE represents the range of disordered molecular energy levels. Note that the amplitude of polariton and molecular DOS has been adjusted to display on the same graph. (b) Time evolution of correlation functions of photon-matter hybrid wavefunctions φ under different disorder σ, as defined in Eq. 9. After introducing molecular rotation, the oscillation period of the photon-matter hybrid states will depend on the molecular rotation period ($\frac{2\pi}{\Omega}$, red dash lines). When the disorder increases, the lifetime of polaritons rapidly decreases and approaches disappearance. Parameters: $\sigma = 2, 3, 4$, $k = 30$, $\hbar = 1$, $g_c = 0.01$, $\omega_c = \omega_m = 0$, $\Omega = 0.1$, $\gamma = 0.1$, $N_F = 3$, $\Delta t = 0.001$, $N_M = 10^6$, $N_C = 1$.

where $\tilde{H} = \mathbf{diag}(\Lambda)$ and $\mathbf{diag}$ means diagonal matrix mapping. $P_C = |\psi_C^F(0)\rangle \langle \psi_C^F(0)|$ is the projection operator to the cavity(photon) space and

$$\psi_C^F(0) = \frac{1}{\sqrt{N_C}} \sum_{q=-N_F}^{N_F} \mathbf{1}_{[\text{start}(q):\text{end}(q)]} \quad (4)$$

where ψ_C^F is an array with $N = (2N_F + 1)(N_M + N_C)$ elements, and $\mathbf{1}_{[\text{start}(q):\text{end}(q)]}$ is an array of length $(N_M + N_C)$, where all elements are zero except for the range $[\text{start}(q), \text{end}(q)]$, where the elements are set to 1. The specific forms of $\text{start}(q)$ and $\text{end}(q)$ are given by:

$$\begin{aligned} \text{start}(q) =& (N_M + N_C)(q + N_F), \\ \text{end}(q) =& (N_M + N_C)(q + N_F + N_C). \end{aligned} \quad (5)$$

Note that $\sum$ in Eq. 4 means array concatenation instead of numerical summation. Before taking the trace, the delta function is initially implemented numerically with Gaussian regularization.[44–46] The Gaussian regularization can be interpreted as applying a Gaussian linebroadening to the traditional time correlation function. The density of states is then directly related to this transformed function by a specific transform:

$$\begin{aligned} \rho(\omega) &= Tr(\int dt \ e^{-i\tilde{H}t} e^{i\omega t} e^{\frac{\gamma^2 t^2}{2}} \ U^\dagger P_C U) \\ &= Tr(\frac{1}{\gamma\sqrt{\pi}} e^{-\frac{(\tilde{H}-\omega)^2}{\gamma^2}} \ U^\dagger P_C U) \end{aligned} \quad (6)$$

in the small γ limit. Through the transformation, we have reduced the originally calculated $[(2N_F + 1)(N_M + N_C), (2N_F + 1)(N_M + N_C)]$ dimension to a controllable $[k, k]$ dimension, greatly reducing computational memory while not losing the key information we need about polaritons.

Correlation Function.—We calculate the correlation function in non-Floquet space, but at the same time, we need to use Floquet Hamiltonian to evolve the wavefunction, such as

$$i\hbar \frac{\partial \psi_C^F(t)}{\partial t} = \hat{H}_s^F \psi_C^F(t). \quad (7)$$

So we need a transformation from the wavefunction in Floquet space to the wavefunction in non-Floquet space.

$$\begin{aligned} \varphi(t) = \\ \frac{1}{2N_F + 1} \sum_{j=0}^{2N_F} \psi_C^F(t)[j(N_M + N_C)] \exp\left(i\Omega(j - N_F)t\right). \end{aligned} \quad (8)$$

Then the correlation function of photonic wavefunction

will be

$$\langle \varphi(0)|\varphi(t)\rangle = \varphi(0)^{\dagger}\varphi(t). \tag{9}$$

Analysis.—In Fig. 1 (c), we explore the Density of States (DOS) of polaritons for varying parameters $\Omega = 0, 0.1, 0.5$ with a strong light-matter interaction regime ($g_c = 0.01$), considering $N_M = 10^6$ molecules coupled to a single cavity mode. For $\Omega = 0$ (no molecular rotation), the system exhibits a Rabi splitting of $\Omega_{Rabi} \approx 2g_c\sqrt{N_M} = 10$, with $\omega_c = \omega_m = 0$ leading to lower and upper polaritons. As Ω increases, the DOS shifts, indicating a dependency on molecular rotation. Particularly, at $\Omega = 0.5$, the polariton DOS approaches that of the non-rotational case, revealing enhanced coupling strength compared to $\Omega = 0.1$. The rotational motion induces inertial effects within molecules, altering the resonance frequency of polaritons. Molecular rotation influences the resonance frequency of molecular vibration, subsequently impacting polariton frequencies.

In Fig. 1 (d), the correlation function of the photonic wavefunction undergoes substantial changes for $\Omega = 0, 0.1, 0.5$. For $\Omega = 0$, the correlation function evolves uniformly and periodically. As Ω increases, the fluctuation period becomes dominated by molecular rotational angular velocity, with $T = 2\pi/\Omega$. The change in fluctuation period from $\Omega = 0.1$ to $\Omega = 0.5$ highlights the profound impact of molecular rotation on photon state evolution. Molecular rotation induces changes in the spatial charge distribution, leading to a time-varying dipole moment that influences the amplitude and phase of polaritons.

In Fig. 2 (a), we investigate the influence of molecular energy disorder (varying σ) on the polariton DOS. As σ increases from 2 to 4, polariton energies deviate significantly from the center of molecular DOS. At the same time, these polaritons are very close to the dark states. Elevated disorder results in stronger spatial localization of polaritons, affecting their propagation and interaction within the optical cavity.

In Fig. 2 (b), we analyze the impact of disorder σ on the correlation function of photons. For $\sigma = 3$ and 4, the correlation function decays close to zero and loses its periodic fluctuations. Molecular energy level disorder broadens the energy levels, diminishing exciton lifetimes by increasing the transition rate between energy levels. This, in turn, affects the stability and duration of polaritons in the optical cavity. The interplay between energy level disorder and rotational motion introduces complexity to the excitation and relaxation processes of excitons, resulting in a dynamic behavior in the lifetime of polaritons.

Finally, in Fig. 3, under weak light-matter interactions ($g_c = 0.0005$), we observe that the introduction of molecular rotation enhances the lifetime of photon states.

Conclusion.—Our investigation delves into the intricate interplay of molecular rotation and energy disorder in the context of strong and weak light-matter interactions. In the regime of strong interactions ($g_c = 0.01$) il-
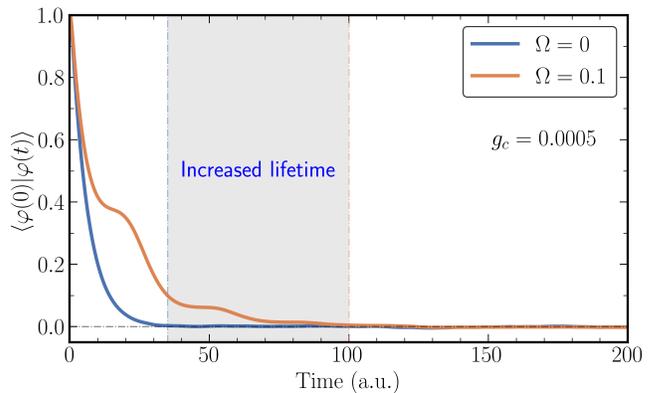
FIG. 3. Time evolution of correlation functions of photonic wavefunctions φ under weak coupling $g_c = 0.0005$. We observe that molecular rotation increases the lifetime of the photon state. Parameters: $\Omega = 0, 0.1$, $k = 30$, $\hbar = 1$, $g_c = 0.0005$, $\omega_c = \omega_m = 0$, $\sigma = 2$, $\gamma = 0.1$, $N_F = 3$, $\Delta t = 0.001$, $N_M = 10^6$, $N_C = 1$.

lustrated in Fig. 1 (c), we find that the rotational motion of molecules significantly influences the Density of States (DOS) of polaritons. Notably, as the angular velocity Ω increases, a pronounced shift in the polariton DOS is observed, indicative of enhanced coupling strength. This effect is attributed to the inertial burden introduced by molecular rotation, impacting the resonance frequency of polaritons. Furthermore, the correlation function of the photonic wavefunction, as depicted in Fig. 1 (d), reveals a direct correlation between molecular rotation and temporal fluctuations in photon states. Molecular rotation induces changes in the charge distribution, impacting both the amplitude and phase of polaritons.

Turning our attention to disorder effects in Fig. 2, we analyze the impact of molecular energy disorder on polaritons. Increasing disorder ($\sigma = 2$ to $\sigma = 4$) leads to a substantial shift in polariton energies away from the molecular DOS center, indicating stronger spatial localization. This spatial confinement has profound implications for polariton propagation and interaction within the optical cavity. Correspondingly, disorder in molecular energy levels, as portrayed in Fig. 2 (b), introduces energy level broadening, diminishing the lifetime of excitons. This reduction in lifetime is attributed to an increased transition rate between energy levels, affecting the stability and existence time of polaritons in the optical cavity. The combined influence of energy disorder and rotational motion results in a dynamic and complex interplay during the excitation and relaxation processes of excitons.

In the context of weak light-matter interactions ($g_c = 0.0005$), Fig. 3 demonstrates that the introduction of molecular rotation has a notable impact on the lifetime of photon states. The observed increase in photon state lifetime underscores the significance of molecular rotation even in the weak coupling regime.

In summary, our findings shed light on the intricate dynamics of polaritons influenced by molecular rotation and energy disorder. The presented results pave the way for a deeper understanding of these phenomena in the realm of cavity quantum electrodynamics and hold implications for the design and optimization of polaritonic devices. Future research in this direction could explore the potential applications of these insights in the design and optimization of optical cavities for specific functionalities, paving the way for advancements in the field of molecular-photon coupling.

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