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Magnetopolariton in bilayer graphene: A tunable ultrastrong light-matter couplingTao Liu¹ and Qi Jie Wang^{1,2,*}¹*NOVITAS, Nanoelectronics Centre of Excellence, and OPTIMUS, Photonics Centre of Excellence, School of Electrical and Electronic Engineering, Nanyang Technological University, 50 Nanyang Avenue, Singapore 639798*²*CDPT, Centre for Disruptive Photonic Technologies, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 637371*

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Magnetopolariton in bilayer graphene (BLG) is theoretically investigated with the consideration of the influence of asymmetry between on-site energies in the two layers of BLG. The results show that an ultrastrong light-matter coupling regime can be achieved in a high filling factor and asymmetry has a strong effect on it. Although BLG in the low-energy regime and semiconductor have a similar quadratic dispersion of quasiparticles, a remarkably different cavity quantum electrodynamics (QED) effect occurs in BLG. In particular, a quantum phase transition, as predicted by the Dicke model, occurs in BLG in spite of the Schrödinger-like term $p^2/2m$ in the system Hamiltonian, while such quantum phase transition does not exist in semiconductors. Most noticeably, the ultrastrong light-matter coupling can be easily controlled by modulating the asymmetry in BLG, which provides an excellent platform to observe interesting QED effects and can lead to tunable polariton-based devices and cavity-controlled magnetotransport in BLG.

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I. INTRODUCTION

Light-matter coupling in a microcavity has been a fascinating topic for fundamental studies of cavity quantum electrodynamics (QED) [1–3]. For applications in quantum devices, enhancing and tuning the coupling strength are crucial. Recently, a newly developed so-called ultrastrong light-matter coupling regime of cavity QED has stimulated great interest and has been experimentally observed [3–7]. This regime can be achieved when the interaction strength between an excitation and the cavity photon, quantified by vacuum Rabi frequency, becomes comparable to or larger than the corresponding electronic transition frequency in a high-finesse cavity. In this regime, the standard rotating-wave approximation is no longer applicable and the antiresonant term of the interaction Hamiltonian starts to play a significant role. As a result, a dramatic modification to the properties of quantum ground state is achieved [4], which leads to very interesting nonadiabatic cavity QED phenomena [5,8].

The ultrastrong coupling regime has been investigated in several systems, such as superconducting quantum circuits in transmission line resonators [9] and exciton and cyclotron transitions in semiconductor microcavities [3,10]. Recently, theoretical prediction has shown that the ultrastrong coupling between a cavity resonator and the cyclotron transition is achievable in monolayer graphene [11]. Moreover, due to the linear dispersion in the low-energy regime, the squared electromagnetic vector potential $\mathbf{A}_{em}^2$ (i.e., diamagnetic term) is neglected when the lattice constant is much smaller than the magnetic length. In this case, the system can be described by the Dicke model [12]. Therefore, a vacuum instability can occur in monolayer graphene with massless fermions, which is absent for massive electrons in semiconductors [11]. This investigation shows that graphene, a two-dimensional (2D) honeycomb crystal of carbon atoms [13], has more diverse

cavity QED phenomena for both fundamental and applied studies. Moreover, recent hybrid graphene/metamaterial devices further pave the way for cavity QED experiments in graphene resonators [14].

In contrast to monolayer graphene with linear dispersion, bilayer graphene (BLG), stacked in the Bernal sequence (A-B stacking), is characterized by a massive Dirac spectrum with the quadratic dispersion in the low-energy regime [15–17]. The Hamiltonian has the off-diagonal structure similar to massless Dirac fermions, but it is combined with Schrödinger-like terms $p^2/2m$ ($p = p_x + ip_y$ is the momentum and $m \approx 0.054m_e$ is the effective mass of quasiparticles in BLG) [16]. Due to this peculiar dispersion relation, BLGs have shown a unique Landau level (LL) spectrum and associated unique quantum phenomena, e.g., unconventional quantum Hall effect, in the presence of a magnetic field [15–20], which is entirely different from monolayer graphene. Most notably, the inversion symmetric AB-stacked BLG is a zero-band-gap semiconductor with a parabolic dispersion in the low-energy regime, but a nonzero band gap can be induced and tuned by breaking the inversion symmetry of the two layers. Indeed, the interlayer asymmetry, which does not exist in monolayer graphene, between the on-site energies in two layers can lead to a significant modification of the band structure and the LL spectrum in BLG [17,21]. Overall, these remarkable differences in the band structure and the LL spectrum make the BLG an interesting platform for the study of cavity QED phenomena. One important opening question is whether a vacuum instability can occur in BLG resonators in spite of the Schrödinger-like term $p^2/2m$ in the system Hamiltonian, and how such asymmetry influences the light-matter coupling.

In this paper, we theoretically investigate the magnetopolariton in BLG under a perpendicular magnetic field by using the quantum field theory. The asymmetry between on-site energies in two layers is taken into account. The results show that an ultrastrong light-matter coupling regime can be achieved in a high filling factor and asymmetry has a strong effect on it. Specifically, a quantum phase transition exists in

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the ultrastrong light-matter coupling regime in spite of the Schrödinger-like terms $p^2/2m$ in the system Hamiltonian in BLG. However, this quantum phase transition is not observed in semiconductors although semiconductors have the same quadratic dispersions as the BLG in the low-energy regime. Most noticeably, the ultrastrong light-matter coupling can be easily controlled by modulating the asymmetry in BLG.

The paper is organized as follows: In Sec. II, we develop the general theory of the coupling between a cavity resonator and cyclotron transition and establish the second quantized quantum light-matter Hamiltonian for BLG system. The asymmetry between the on-site energies in the two layers in BLG is considered. In Sec. III, we firstly demonstrate the tunability of vacuum Rabi frequency by tuning the asymmetry. Then the vacuum instability, magnetopolariton dispersion, and its dependence on asymmetry in BLG are discussed. Finally, conclusions are drawn in Sec. IV. Additional theoretical derivations are presented in Appendixes A and B.

II. PHYSICAL SYSTEM AND INTERACTION HAMILTONIAN

The BLG is modeled as two coupled hexagonal lattices with inequivalent sites A_1 , B_1 and A_2 , B_2 on the top and bottom graphene sheets, respectively. They are arranged according to the Bernal (A_2 - B_1) stacking [13], as shown in Fig. 1. This lattice has a degeneracy point at each of the two inequivalent corners, K and K' . When trigonal warping is neglected, the low-energy states of electrons in, e.g., the valley K can be described by an effective two-component Hamiltonian (see Appendix A):

$$H = -\frac{1}{2m} \begin{pmatrix} 0 & (p^\dagger)^2 \\ p^2 & 0 \end{pmatrix} - \frac{1}{2}U \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} + \frac{1}{2m} \frac{U}{\gamma_1} \begin{pmatrix} p^\dagger p & 0 \\ 0 & -pp^\dagger \end{pmatrix}, \quad (1)$$

where $p = p_x + ip_y$. $\gamma_1 \approx 0.4$ eV is the interlayer hopping amplitude and the effective mass $m = \gamma_1/2v^2 \approx 0.054m_e$ (v is the single-layer Dirac velocity and m_e is the free electron mass). U describes the asymmetry between the top

FIG. 1. (Color online) Schematic illustration of the structure of bilayer graphene with the Bernal (A_2 - B_1) stacking. γ_0 and γ_1 are the intralayer and interlayer hopping amplitudes, respectively.

and the bottom layers, which open a minigap. The two-component Hamiltonian is applicable within the energy range smaller than γ_1 .

When a perpendicular magnetic field $\mathbf{B}$ is applied to the graphene plane, the electrons occupy highly degenerate LLs. The LL spectrum can be exactly solved by replacing the $\mathbf{p}$ with $\Pi_0 = \mathbf{p} + (e/c)\mathbf{A}_0$ within the limit $a/\ell_0 \ll 1$ (see Appendix A), where $\mathbf{A}_0 = -By\mathbf{u}_x$ is the vector potential in Landau gauge. The Dirac fermion field operator in the valley K can be written on a Landau level basis as

$$\psi(r) = \sum_{\zeta,n,k} \begin{pmatrix} \varphi_{\zeta,n,k}^{A_1}(r) \\ \varphi_{\zeta,n,k}^{B_2}(r) \end{pmatrix} c_{\zeta,n,k} = \sum_{\zeta,n,k} \frac{e^{ikx}}{\sqrt{L}} \begin{pmatrix} a_n \phi_{n-2,k}(y) \\ b_n \phi_{n,k}(y) \end{pmatrix} c_{\zeta,n,k}, \quad (2)$$

where $c_{\zeta,n,k}$ is the annihilation operator of electrons in the state with quantum numbers (ζ, n, k) . k is the wave vector which is the eigenvalue of the magnetic translation operator in the x direction. $\zeta = +(-)$ indicates the conduction (valence) band levels and LL index $n \in \mathbb{N}$. a_n and b_n are normalized coefficients (see Appendix A). $\phi_{n,k}(y)$ refers to the eigenfunction of the one-dimensional harmonic oscillator problem shifted by the guiding center position $y_0 = k\ell_0^2$ [22]. Each LL has degeneracy of $\mathcal{N} = g_s S/2\pi\ell_0^2$, where $g_s = 4$ is the spin and valley isospin degeneracy, and S is the surface area of the graphene sheet. The cyclotron frequency and magnetic length are $\omega_0 = eB/mc$ and $\ell_0 = \sqrt{\hbar c/eB}$, respectively. The first unoccupied LL index n is determined by the filling factor $\nu = \rho S/\mathcal{N}$ (ρ is electron density). In the paper, for the sake of simplicity, we will consider the case of an integer filling factor ν and assume the Fermi level is within the conduction band. As we deal with the coupling between the light and high filling factor LLs, we have to take into account the system at cryogenic temperature. Therefore, we assume zero temperature in the rest of the calculations of this paper to ensure the cyclotron transition energy is larger than thermal energy.

The cavity geometry (see Fig. 2) that we consider has a dimension with volume $V = L_z L^2$. The graphene sheet is perpendicular to the z direction and is placed at the center of the cavity. The cavity length L_z along the z direction is

FIG. 2. (Color online) Sketch of a cavity resonator embedding a bilayer graphene. A uniform and static magnetic field B is perpendicular to the graphene layer.

assumed to be much smaller than the cavity transverse size L . The electromagnetic modes are confined along the three spatial directions. The BLG is placed at the center of the cavity. We consider a particular cavity mode with wave vector $\mathbf{q} = (q_x, q_y, q_z) = (2\pi/L, 2\pi/L, 2\pi/L_z)$ resonant with the active cyclotron transition. For this considered cavity mode, the electromagnetic vector potential can be written as

$$\mathbf{A}_{em}(r) = \sum_{\eta=1,2} \sqrt{\frac{2\pi\hbar c^2}{\varepsilon\omega_c V}} \mathbf{u}_\eta (a_\eta + a_\eta^\dagger), \quad (3)$$

where $a_\eta^\dagger$ is the bosonic creation operator for a given photon mode $\mathbf{q}$ with the polarization $\eta = 1, 2$. $\omega_c = (\pi c/L_z \sqrt{\varepsilon}) \sqrt{1 + 8(L_z/L)^2}$ is cavity frequency, where ε is the cavity dielectric constant. The mode spatial profile $\mathbf{u}_\eta$ is written as [11,23]

$$u_1 = \begin{pmatrix} 2 \cos(2\pi x/L) \sin(2\pi y/L) \cos(\theta) \\ 2 \sin(2\pi x/L) \cos(2\pi y/L) \cos(\theta) \\ 0 \end{pmatrix}, \quad (4)$$

$$u_2 = \begin{pmatrix} -2 \cos(2\pi x/L) \sin(2\pi y/L) \\ 2 \sin(2\pi x/L) \cos(2\pi y/L) \\ 0 \end{pmatrix}, \quad (5)$$

where $\cos(\theta) = 1/\sqrt{1 + 8(L_z/L)^2}$.

The light-matter interactions are described by the minimal coupling $\mathbf{p} \rightarrow \Pi_0 + (e/c)\mathbf{A}_{em}$ in the perpendicular magnetic field. Then the second quantized form of coupling Hamiltonian can be extracted from $\int dr \psi^\dagger(r) H_m \psi(r)$, where H_m is the minimal Hamiltonian, by replacing $\mathbf{p}$ in Eq. (1) with $\Pi_0 + (e/c)\mathbf{A}_{em}$ within the limits $a/\ell_0 \ll 1$ and $a/L \ll 1$ (see Appendix B). It is noted that, due to Schrödinger-like terms $p^2/2m$ in Eq. (1), we can see the diamagnetic term in this minimal Hamiltonian H_m when replacing $\mathbf{p}$ in Eq. (1) with $\Pi_0 + (e/c)\mathbf{A}_{em}$. In order to obtain the bosonized version of the system second quantized Hamiltonian $\int dr \psi^\dagger(r) H_m \psi(r)$, we can perform a procedure of LL bosonization [24].

In analogy to the case of the monolayer graphene and by following the procedure of LL bosonization [11,24,25], we obtain the bosonic bright mode annihilation operator between the transitions $v \rightarrow v-1$ coupled to cavity modes 1 and 2 considering the condition $|\mathbf{q}|\ell_0 \ll 1$ (for the photonic wave vector, this condition is always satisfied which allows us to neglect the LL mixing [11]):

$$d_1 = \frac{1}{\sqrt{N}} \sum_k \sum_{\pm} \sin \left[\frac{2\pi}{L} \left(k \pm \frac{\pi}{L} \right) \ell_0^2 \mp \frac{\pi}{4} \right] c_{v-1,k}^\dagger c_{v,k \pm 2\pi/L}, \quad (6)$$

$$d_2 = \frac{1}{\sqrt{N}} \sum_k \sum_{\pm} \pm \cos \left[\frac{2\pi}{L} \left(k \pm \frac{\pi}{L} \right) \ell_0^2 \mp \frac{\pi}{4} \right] c_{v-1,k}^\dagger c_{v,k \pm 2\pi/L}, \quad (7)$$

where d_η and $d_\eta^\dagger$ are approximately bosonic operators in the dilute regime under consideration and they satisfy the

commutation relation by its average value in the ground state $|F\rangle = \prod_{n=0}^{v-1} \prod_{k=0}^N c_{n,k}^\dagger |0\rangle$ as

$$[d_\eta(k), d_\eta^\dagger(k')] \approx \delta_{k,k'}. \quad (8)$$

Starting from the bosonic bright mode operator, after some calculations that are detailed in Appendix B, we can get the following bosonized version of the single-particle and light-matter interaction Hamiltonian and Hamiltonian with diamagnetic term:

$$\mathcal{H}_L = \sum_\eta \hbar \omega_L d_\eta^\dagger d_\eta, \quad (9)$$

$$\mathcal{H}_{\text{int}} = \sum_\eta \hbar \Omega_\eta \chi_\eta (d_\eta + d_\eta^\dagger) (a_\eta + a_\eta^\dagger), \quad (10)$$

$$\mathcal{H}_{\text{dia}} = \sum_\eta \hbar D_\eta (a_\eta + a_\eta^\dagger)^2, \quad (11)$$

with $\chi_1 = -1$, $\chi_2 = 1$, $\Omega_1 = \Omega_2 \cos(\theta)$, $D_1 = D_2 \cos^2(\theta)$, and the vacuum Rabi frequency is

$$\Omega_2 = -\omega_0 \sqrt{\frac{\alpha c g_s}{\varepsilon \omega_c L_z}} [2a_v b_{v-1} \sqrt{v-1} + (U/\gamma_1)(a_{v-1} a_v \sqrt{v-1} - b_{v-1} b_v \sqrt{v})], \quad (12)$$

where $\alpha = e^2/\hbar c$. ω_L and D_2 are

$$\begin{aligned} \omega_L = & -2\omega_0 [a_v b_v \sqrt{v(v-1)} - a_{v-1} b_{v-1} \sqrt{(v-1)(v-2)}] \\ & - \frac{U}{2\hbar} [(a_v^2 - b_v^2) - (a_{v-1}^2 - b_{v-1}^2)] \\ & + \frac{U}{2\gamma_1} \omega_0 [a_v^2 (2v-2) - b_v^2 (2v+2) \\ & - a_{v-1}^2 (2v-4) + b_{v-1}^2 (2v)], \end{aligned} \quad (13)$$

$$D_2 = \frac{U}{\gamma_1} \frac{2\pi \alpha \omega_0 c \ell_0^2}{\varepsilon \omega_c V} \sum_{n=0}^{v-1} (a_n^2 - b_n^2). \quad (14)$$

Since we only consider the weak asymmetry in this paper, i.e., $|U/\gamma_1| \ll 1$ together with $|\mathbf{q}|\ell_0 \ll 1$, by evaluating Eqs. (12) and (14), we have

$$\frac{\Omega_2^2/\omega_{eg}}{|D_2|} > \frac{2g_s \gamma_1 L^2 b_{v-1}^2}{\pi |U| \ell_0^2} \gg 1. \quad (15)$$

Therefore, the diamagnetic term $\mathcal{H}_{\text{dia}}$ of the system Hamiltonian can be neglected. A quantum phase transition occurs in our considered BLG system [12]. It is noted that an additional diamagnetic term can be derived when the higher-order expansion of the vector potential term in Eq. (B1) is taken into account, which can also be neglected within the limits $a/\ell_0 \ll 1$ and $a/L \ll 1$.

In addition to the light-matter interaction, Coulomb interaction can play a crucial role in BLG under the magnetic field [26]. Based on our cavity structure, we expand the Coulomb potential $V(r-r')$ in terms of the 2D Fourier series [11]. Because for the photonic wave vector $\mathbf{q}$, $|\mathbf{q}|\ell_0 \ll 1$ is always satisfied, we only take into account the Coulomb interaction between the transitions $v \rightarrow v-1$. By the bosonization of the Coulomb Hamiltonian (here we only consider the Coulomb

depolarization effects), in addition to the bosonic bright mode, two additional dark modes can be obtained [11]. Since the bright and dark modes are decoupled by the Coulomb interaction, we can write the Coulomb Hamiltonian in terms of only the bright mode as

$$\mathcal{H}_{\text{Coul}} = \sum_{\eta} \hbar V_c \chi_{\eta} (\chi_{\eta} d_{\eta} + d_{\eta}^{\dagger})^2, \quad (16)$$

where

$$V_c = \frac{\pi g_s \alpha c}{4\sqrt{2}\varepsilon L} (a_{v-1} a_v \sqrt{v-2} + b_{v-1} b_v \sqrt{v})^2. \quad (17)$$

By diagonalizing the single-particle and Coulomb Hamiltonian, we can obtain the magnetoplasmon modes,

$$\begin{aligned} \mathcal{H}_{mp} &= \sum_{\eta} \hbar [\omega_L d_{\eta}^{\dagger} d_{\eta} + V_c \chi_{\eta} (\chi_{\eta} d_{\eta} + d_{\eta}^{\dagger})^2] \\ &= \sum_{\eta} \hbar \omega_p m_{\eta}^{\dagger} m_{\eta} + \text{const}, \end{aligned} \quad (18)$$

where m_{η} is the magnetoplasmon mode with frequency $\omega_p = \sqrt{\omega_L(\omega_L + 4V_c)}$. Then the considered system's Hamiltonian is written in terms of the magnetoplasmon and photonic modes as

$$\begin{aligned} \mathcal{H}_{\text{tot}} &= \sum_{\eta} [\hbar \omega_p m_{\eta}^{\dagger} m_{\eta} \\ &\quad + \hbar \omega_c a_{\eta}^{\dagger} a_{\eta} + \hbar \Lambda_{\eta} (m_{\eta} + m_{\eta}^{\dagger})(a_{\eta} + a_{\eta}^{\dagger})], \end{aligned} \quad (19)$$

where $\Lambda_1 = -\Omega_1 \sqrt{\omega_p/\omega_L}$ and $\Lambda_2 = -\Omega_2 \sqrt{\omega_L/\omega_p}$.

We introduce the lower polariton (LP) and upper polariton (UP) annihilation operators,

$$\beta_{j,\eta} = X_{j,\eta} a_{\eta} + Y_{j,\eta} m_{\eta} + Z_{j,\eta} a_{\eta}^{\dagger} + R_{j,\eta} m_{\eta}^{\dagger}, \quad (20)$$

where $j \in \{\text{LP}, \text{UP}\}$ and $[\beta_{\eta}(k), \beta_{\eta}^{\dagger}(k')] = \delta_{k,k'}$. By using a Hopfield-Bogoliubov transformation [27], the Hamiltonian of the system can be rewritten in the diagonal form,

$$\mathcal{H}'_{\text{tot}} = \sum_{\eta} \sum_{j \in \{\text{LP}, \text{UP}\}} \hbar \omega_{j,\eta} \beta_{j,\eta}^{\dagger} \beta_{j,\eta}, \quad (21)$$

where, by using the commutation relation $[\beta_{j,\eta}, \mathcal{H}_{\text{tot}}] = \hbar \omega_{j,\eta} \beta_{j,\eta}$, we can obtain the frequency $\omega_{j,\eta}$ of magnetopolariton where the Hopfield-Bogoliubov matrix $\mathcal{M}_{\eta}$ satisfies $\mathcal{M}_{\eta} \mathbf{v}_{\eta} = \hbar \omega_{j,\eta} \mathbf{v}_{\eta}$, and $\mathbf{v}_{\eta}$ is the vector $\mathbf{v}_{\eta} = (X_{j,\eta}, Y_{j,\eta}, Z_{j,\eta}, R_{j,\eta})^T$.

III. RESULTS AND DISCUSSIONS

A. Ultrastrong light-matter coupling and its dependence on asymmetry

We have obtained the vacuum Rabi frequency of the BLG system in Eq. (10). The first conclusion is that the BLG system can be operated in the ultrastrong coupling regime. In particular, when $|U| \leq \hbar \omega_0$, we have $a_n \approx -b_n = 1/\sqrt{2}$, then $\Omega_2 \approx \omega_0 \sqrt{\alpha c g_s (v-1)/(\varepsilon \omega_c L_z)}$, $\omega_L \approx \omega_0$ ($v \gg 1$), and the LL transition frequency between $v \rightarrow v-1$ is $\omega_{eg} \approx \omega_0$ ($v \gg 1$). Compared with semiconductors [28], we find that BLG has the same equidistant LL spectrum with semiconductor in high filling factor when $|U| \leq \hbar \omega_0$, and their vacuum Rabi frequencies have the same forms, which are all proportional to

FIG. 3. (Color online) Vacuum Rabi frequency and Ω_2/ω_{eg} as a function of asymmetry U for a given magnetic field $B = 120$ mT and doping density $\rho = 4 \times 10^{11} \text{ cm}^{-2}$. Other parameters are $\varepsilon = 4.0$, $L_z = 1 \text{ mm}$, and $L = 10L_z$.

the square root of the filling factor due to their similar parabolic dispersions. The vacuum Rabi frequency in BLG normalized to the transition frequency in high filling factor is

$$\frac{\Omega_2}{\omega_{eg}} \approx \sqrt{\alpha g_s (v-1)/\pi \sqrt{\varepsilon [1 + 8(L_z/L)^2]}}, \quad (22)$$

which can be comparable to or larger than 1 for large enough filling factors (e.g., $\Omega_2/\omega_{eg} \approx 0.48$ for $v = 50$). Therefore, the BLG can achieve the ultrastrong coupling in the limit $|U| \leq \hbar \omega_0$. Most noticeably, when we do not consider the dependence of asymmetry U on the carrier density ρ , the vacuum Rabi frequency shows a strong dependence on the asymmetry U in the limit $\hbar \omega_0 < |U| \ll \gamma_1$ at the fixed carrier density $\rho = 4 \times 10^{11} \text{ cm}^{-2}$, as shown in Fig. 3. Since the vacuum Rabi frequency is comparable to the transition frequency, an ultrastrong coupling regime can always be achieved in BLG when $|U| \ll \gamma_1$. However, the asymmetry U between on-site energies usually depends strongly on the doping density, and the asymmetry can achieve $U \sim \gamma_1/100$ at doping density $\rho = 4 \times 10^{11} \text{ cm}^{-2}$ when a single gate is applied to control the carrier density [21]. The corresponding vacuum Rabi frequency is actually $\Omega_2 \approx 0.62 \text{ GHz rad}^{-1}$, which prevents us from experimentally observing the strong light-matter coupling phenomena in BLG. Therefore, in order to achieve the ultrastrong light-matter coupling in experiments, we must greatly reduce the influence of doping density on asymmetry. For example, we can simultaneously use the top and the bottom gates to control independently the asymmetry and doping density [29]. In this scheme, doping density is determined by $\delta D = D_b - D_t$, and asymmetry by $\Delta D = (D_b + D_t)/2$, where D_b and D_t are the bottom and top electrical displacement fields, respectively. Then we can achieve a suitable doping density but a small asymmetry. Moreover, we can tune the asymmetry to modulate the ultrastrong light-matter coupling by using this scheme. This shows that the BLG could potentially be used to experimentally study the interesting QED, e.g., generating correlated photon pairs out of the vacuum [4]. We would like to point out that, when the asymmetry satisfies $|U| > \gamma_1$, the effective two-component Hamiltonian [Eq. (1)] is not

FIG. 4. (Color online) (a) Normalized frequencies of LP and UP branches of magnetopolariton with the limit $|U| \leq \hbar\omega_0$ as a function of carrier density for $\eta = 1$ and $\eta = 2$. Parameters are $B = 120$ mT, $\varepsilon = 4.0$, $L_z = 1$ mm, $L = 10L_z$, and $\omega_c = 0.49$ THz rad $^{-1}$. The critical density is $\rho_c = 8.8 \times 10^{11} \text{ cm}^{-2}$. (b) Same quantities as a function of external magnetic field for $\eta = 1$ and $\eta = 2$. The density is $\rho = 8.78 \times 10^{11} \text{ cm}^{-2}$ which is just below the critical density.

applicable. The system cavity QED phenomena should be described by the four-component Hamiltonian, and this will be the scope of future investigations.

B. Magnetopolariton dispersion and vacuum instability

As written in Eq. (19), the quantum Hamiltonian $\mathcal{H}_{\text{tot}}$ of massive fermions in BLG recalls the celebrated Dicke model [30] of cavity QED in spite of Schrödinger-like terms $p^2/2m$, which is in contrast to the case for the massive quasiparticles in semiconductor resonators [6,28]. In semiconductors, the diamagnetic term is even dominant in ultrastrong coupling regime between a cavity resonator and cyclotron transitions [28]. In BLG resonators, a quantum critical point exists beyond which the normal ground state becomes unstable. The Dicke-like Hamiltonian $\mathcal{H}_{\text{tot}}$ in BLG is attributed to the peculiar crystal structure, while in semiconductors, the crystal structure is usually not important for many physical effects, and the effective mass approximation can work very well in a broad range of conditions [31].

To analyze the magnetopolariton characteristics in BLG, we firstly consider the limit $|U| \leq \hbar\omega_0$ (in the below discussions, we consider asymmetry independent of doping density, which is achievable in experiments). In this case, the system's

Hamiltonian $\mathcal{H}_{\text{tot}}$ has a very simple expression. Figure 4 shows the density and magnetic field dependences of frequencies of magnetopolariton normalized to the cavity mode, where the LP and UP branches are the two spectrally separated light-matter eigenstates in strong coupling regime. By increasing the density, the LP branches for $\eta = 1$ disappear (i.e., $\omega_{LP,1} = 0$) in the critical density $\rho_c = 8.8 \times 10^{11} \text{ cm}^{-2}$. The critical density can be evaluated by setting $\text{Det}(\mathcal{M}_\eta) = 0$, and it is, in particular, $\rho_c = m\varepsilon\omega_c L_z / [8\pi \cos^2(\theta)\hbar\alpha]$ for $\eta = 1$ within the limit $|U| \leq \hbar\omega_0$. Above the critical point, the vacuum state of the cavity resonator is twice degenerate and coherent spontaneous emission occurs. In Fig. 4(b), when we increase the magnetic field at just below the critical density, a strong asymmetric dispersion is exhibited, and the LP branches for $\eta = 1$ disappear at $B = 175$ mT, which shows the signature of such a vacuum instability. We would like to demonstrate that BLG has a quite different polariton dispersion from monolayer graphene due to their different band structures [11].

C. Magnetopolariton dispersion as a function of asymmetry

As discussed in Sec. III A (see Fig. 3), the vacuum Rabi frequency has a strong dependence on the asymmetry when $|U| > \hbar\omega_0$. It is shown that the asymmetry can be easily

FIG. 5. (Color online) (a) Normalized frequencies of LP and UP branches as function of the normalized asymmetry U/γ_1 for $\eta = 1$. Parameter are $\rho = 6 \times 10^{11} \text{ cm}^{-2}$ and $B = 120$ mT. (b) The critical density ρ_c as a function of the normalized asymmetry U/γ_1 for $\eta = 1$. $B = 120$ mT is used.

FIG. 6. (Color online) Normalized frequencies of LP and UP branches as a function of carrier density ρ with (a) $U/\gamma_1 = 10^{-4}$ and (b) $U/\gamma_1 = 5 \times 10^{-4}$ for $\eta = 1$. Parameter is $B = 120$ mT.

controlled by, e.g., doping [32] and external gate voltage [29,33], which can enable us to control the ultrastrong light-matter coupling and even manipulate the quantum vacuum state by modulating the asymmetry U in BLG. In the ultrastrong light-matter coupling regime, the ground state of the system is a squeezed vacuum and has correlated photon pairs [4]. However, these photon pairs in the ground state are virtual and cannot be coupled out of the cavity if there is no modulation of system parameters [8]. The possibility of tuning the properties of the ground state of the system suggests that the BLG could potentially be a good platform to study and observe interesting QED phenomena. In Fig. 5(a), we show the normalized frequencies of magnetopolariton versus the normalized asymmetry U . A clear dependence of polariton frequencies on the asymmetry U is observed in the considered cavity modes. As an example, we show the polariton dispersions with $U/\gamma_1 = 10^{-4}$ and $U/\gamma_1 = 5 \times 10^{-4}$ in Figs. 6(a) and 6(b), respectively. In addition to quite different polariton dispersions, as the density increases, both LP branches will disappear, but at different critical values. The dependence of critical value of the quantum phase transition on the asymmetry U is shown in Fig. 5(b). We see a strong increase of the critical density as U increases.

IV. CONCLUSIONS

In conclusion, we have theoretically investigated the cavity QED in BLG, and the asymmetry between on-site energies in two layers is taken into account. The results show that, due to the peculiar crystal structure, BLG shows a quantum phase transition, as predicted by the Dicke model, in spite of the Schrödinger-like terms $p^2/2m$ in the system Hamiltonian within the low-energy regime. This is in contrast to the case for semiconductor resonators with massive quasiparticles. In addition, the characteristics of the light-matter coupling in BLG are different from those of monolayer graphene due to their different band structure. Most noticeably, the vacuum Rabi frequency drastically reduces as the asymmetry increases, which prevents us from observing the ultrastrong light-matter coupling phenomena experimentally due to the strong dependence of asymmetry on the doping density. However, the dilemma can be overcome by independent control of the asymmetry and doping density, e.g., using a double-gate tuning structure. Therefore, we can control the ultrastrong

light-matter coupling and manipulate the quantum vacuum state by modulating the asymmetry in BLG. Our study provides a theoretical foundation on observation and investigation of interesting QED, e.g., generating the correlated photon pair out of the vacuum, for fundamental studies and quantum applications in BLG.

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APPENDIX A: EFFECTIVE TWO-COMPONENT HAMILTONIAN AND LL SPECTRUM

As shown in Fig. 1, when trigonal warping is neglected, the tight-binding Hamiltonian of BLG with Bernal stacking in the space of wave functions $\psi = (\psi_{A_1}, \psi_{B_1}, \psi_{A_2}, \psi_{B_2})^T$ can be expressed as

$$H = \begin{pmatrix} -U/2 & -\gamma_0 f(k) & 0 & 0 \\ -\gamma_0 f^*(k) & -U/2 & \gamma_1 & 0 \\ 0 & \gamma_1 & U/2 & -\gamma_0 f(k) \\ 0 & 0 & -\gamma_0 f^*(k) & U/2 \end{pmatrix}, \quad (A1)$$

where $f(k) = \sum_{j=1}^3 e^{-ik \cdot \delta_j}$ describes nearest-neighbor hopping in bottom graphene sheets, $\mathbf{k}$ is the in-plane wave vector, and $\delta_1 = (a/2, a/2\sqrt{3})$, $\delta_2 = (-a/2, a/2\sqrt{3})$, $\delta_3 = (0, -a/\sqrt{3})$ is the position of the three nearest B_1 atoms relative to a given A_1 atom ($a = 0.246$ nm is the lattice constant). γ_0 and γ_1 are the intralayer and interlayer hopping parameters, respectively.

The graphene has a degeneracy point at each of the two inequivalent corners, K and K' . We consider the properties of electrons in the vicinity of, in particular, the K point with $\mathbf{K} = [4\pi/(3a), 0]$. For the low-energy electronic band structure (i.e., $|p|a/\hbar \ll 1$, where momentum $p = p_x + ip_y$), the function $f(k)$ can be approximately given by

$f(k) \approx -\sqrt{3}a(p_x - ip_y)/2\hbar$. Then we can obtain an effective two-component Hamiltonian in the space of wave function $\psi = (\psi_{A_1}, \psi_{B_2})^T$ in the valley K by using the following equations [18]:

$$H \approx (1 + uh_{\beta}^{-2}u^{\dagger})^{-1/2}(h_{\theta} - uh_{\beta}^{-1}u^{\dagger})(1 + uh_{\beta}^{-2}u^{\dagger})^{-1/2}, \quad (\text{A2})$$

with

$$h_{\theta} = \begin{pmatrix} -U/2 & 0 \\ 0 & U/2 \end{pmatrix}, \quad h_{\beta} = \begin{pmatrix} U/2 & \gamma_1 \\ \gamma_1 & -U/2 \end{pmatrix}, \quad (\text{A3})$$

$$u = \begin{pmatrix} 0 & vp^{\dagger} \\ vp & 0 \end{pmatrix}, \quad u^{\dagger} = \begin{pmatrix} 0 & vp^{\dagger} \\ vp & 0 \end{pmatrix}, \quad (\text{A4})$$

where $v = \sqrt{3}a\gamma_0/2\hbar$ is the velocity.

By assuming $|E/\gamma_1| \ll 1$ and $|U/\gamma_1| \ll 1$, the effective two-component Hamiltonian of Eq. (A2) becomes

$$H = -\frac{1}{2m} \begin{pmatrix} 0 & h_1 \\ h_1^{\dagger} & 0 \end{pmatrix} - \frac{1}{2}U \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} + \frac{1}{2m} \frac{U}{\gamma_1} \begin{pmatrix} h_2 & 0 \\ 0 & -h_2^{\dagger} \end{pmatrix}, \quad (\text{A5})$$

where $m = \gamma_1/2v^2$ is the effective mass of Dirac fermions and the Hamiltonian h_1 and h_2 are expressed as $h_1 = (p^*)^2$ and $h_2 = p^*p$.

When a static magnetic field $\mathbf{B}$ is perpendicularly applied to the BLG plane, the operator $\mathbf{p}$ in Eq. (A5) can be replaced by $\Pi_0 = \mathbf{p} + (e/c)\mathbf{A}_0$ within the limit $a/\ell_0 \ll 1$, where $\mathbf{A}_0 = -By\mathbf{u}_x$ is the vector potential in the Landau gauge. The moment operator Π_0 can be expressed in terms of the harmonic oscillator ladder operator dr and $dr^{\dagger}$ such that $d_l|n, k\rangle = \sqrt{n}|n-1, k\rangle$, $d_l^{\dagger}|n, k\rangle = \sqrt{n+1}|n+1, k\rangle$, and $[d_l, d_l^{\dagger}] = 1$:

$$\Pi_{0,x} = -\frac{\sqrt{2}\hbar}{2\ell_0}(d_l + d_l^{\dagger}), \quad (\text{A6})$$

$$\Pi_{0,x} = i\frac{\sqrt{2}\hbar}{2\ell_0}(d_l^{\dagger} - d_l), \quad (\text{A7})$$

where $\ell_0 = \sqrt{\hbar c/eB}$ is the magnetic length. By substituting Eqs. (A6) and (A7) into Eq. (A5), we can obtain the LL spectrum as

For $n = 0, 1$,

$$\begin{pmatrix} \varphi_{0,k}^{A_1}(r) \\ \varphi_{0,k}^{B_2}(r) \end{pmatrix} \sim \begin{pmatrix} 0 \\ \langle r|0, k \rangle \end{pmatrix} \quad \text{and} \quad \begin{pmatrix} \varphi_{1,k}^{A_1}(r) \\ \varphi_{1,k}^{B_2}(r) \end{pmatrix} \sim \begin{pmatrix} 0 \\ \langle r|1, k \rangle \end{pmatrix}, \quad (\text{A8})$$

$$E_0 = U/2 \quad \text{and} \quad E_1 = U/2 - \Delta, \quad (\text{A9})$$

For $n \geq 2$,

$$\begin{pmatrix} \varphi_{n,k}^{A_1}(r) \\ \varphi_{n,k}^{B_2}(r) \end{pmatrix} \sim \begin{pmatrix} a_n \langle r|n-2, k \rangle \\ b_n \langle r|n, k \rangle \end{pmatrix}, \quad (\text{A10})$$

$$E_n = \pm \sqrt{(\hbar\omega_0)^2 n(n-1) + (U/2 - n\Delta)(U/2 - n\Delta + \Delta) + \Delta^2} - \Delta/2, \quad (\text{A11})$$

where $+$ ($-$) refers to electron (hole) states, and

$$\Delta = U\hbar\omega_0/\gamma_1,$$

$$a_n = 1/\sqrt{1 + D_n^2},$$

$$b_n = -D_n/\sqrt{1 + D_n^2},$$

$$D_n = (E_n - U/2 + n\Delta)/[\hbar\omega_0\sqrt{n(n-1)}].$$

In particular, when $|U| \leq \hbar\omega_0$, $E_n = \pm\hbar\omega_0\sqrt{n(n-1)} - \Delta/2$. In this case, the BLG has almost equidistant energy levels in the high filling factor, which is similar to semiconductors.

APPENDIX B: LIGHT-MATTER COUPLED HAMILTONIAN

The coupling Hamiltonian between cavity mode and cyclotron transition is obtained by replacing $\hbar\mathbf{k}$ with $\Pi = \Pi_0 + (e/c)\mathbf{A}_{em}$ in Eq. (A1). In particular, in the valley K , the nearest-neighbor hopping function $f(k)$ can be expressed as

$$f(k) = \sum_{j=1}^3 e^{-i\mathbf{K}\cdot\delta_j} \exp \left\{ -\frac{i}{\hbar} [\Pi_0 \cdot \delta_j + (e/c)\mathbf{A}_{em} \cdot \delta_j] \right\}. \quad (\text{B1})$$

By using Eqs. (A6), (A7), and (3) and expanding the exponential factor of Eq. (B1), when $a/\ell_0 \ll 1$ and $a/L \ll 1$, the function $f(k)$ becomes

$$f(k) \approx -\sqrt{3}a(\Pi_x - i\Pi_y)/2\hbar, \quad (\text{B2})$$

where Eq. (B2) is accurate up to the terms which are proportional to a .

We assume $|E/\gamma_1| \ll 1$ and $|U/\gamma_1| \ll 1$. Then, by substituting Eq. (B2) into Eq. (A1) and performing the procedures of Eqs. (A2)–(A4) with replacing $\mathbf{p}$ with $\Pi = \Pi_0 + (e/c)\mathbf{A}_{em}$ in Eq. (A4), the effective two-component minimal coupling Hamiltonian, in the valley K , describing the electrons coupled to the electromagnetic field reads

$$H_m = -\frac{1}{2m} \begin{pmatrix} 0 & \hat{h}_1 \\ \hat{h}_1^{\dagger} & 0 \end{pmatrix} - \frac{1}{2}U \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix} + \frac{1}{2m} \frac{U}{\gamma_1} \begin{pmatrix} \hat{h}_2 & 0 \\ 0 & -\hat{h}_2^{\dagger} \end{pmatrix}, \quad (\text{B3})$$

where

$$\hat{h}_1 = \left[\left(\Pi_{0,x} + \frac{e}{c}A_{em,x} \right) - i \left(\Pi_{0,y} + \frac{e}{c}A_{em,y} \right) \right]^2, \quad (\text{B4})$$

$$\hat{h}_2 = \left[\left(\Pi_{0,x} + \frac{e}{c}A_{em,x} \right) - i \left(\Pi_{0,y} + \frac{e}{c}A_{em,y} \right) \right] \times \left[\left(\Pi_{0,x} + \frac{e}{c}A_{em,x} \right) + i \left(\Pi_{0,y} + \frac{e}{c}A_{em,y} \right) \right]. \quad (\text{B5})$$

From Eqs. (B4) and (B5), we can identify three different terms in the Hamiltonian, i.e., the Hamiltonian of single-particle and light-matter interaction and the Hamiltonian with diamagnetic term,

$$h_1 = h_{L1} + h_{\text{int}1} + h_{\text{dia}1}, \quad (\text{B6})$$

$$h_2 = h_{L2} + h_{\text{int}2} + h_{\text{dia}2}, \quad (\text{B7})$$

where

$$h_{L1} = (\Pi_{0,x}^2 - \Pi_{0,y}^2) - 2i\Pi_{0,x}\Pi_{0,y} - (\hbar/\ell_0)^2, \quad (\text{B8})$$

$$h_{L2} = \Pi_{0,x}^2 + \Pi_{0,y}^2 + (\hbar/\ell_0)^2, \quad (\text{B9})$$

$$h_{\text{int}1} = 2\frac{e}{c}(\Pi_{0,x}A_{em,x} - \Pi_{0,y}A_{em,y}) - 2i\frac{e}{c}(\Pi_{0,x}A_{em,y} + \Pi_{0,y}A_{em,x}), \quad (\text{B10})$$

$$h_{\text{int}2} = 2\frac{e}{c}(\Pi_{0,x}A_{em,x} + \Pi_{0,y}A_{em,y}), \quad (\text{B11})$$

$$h_{\text{dia}1} = \left(\frac{e}{c}\right)^2 (A_{em,x}^2 - A_{em,y}^2) - 2i\left(\frac{e}{c}\right)^2 A_{em,x}A_{em,y}, \quad (\text{B12})$$

$$h_{\text{dia}2} = \left(\frac{e}{c}\right)^2 (A_{em,x}^2 + A_{em,y}^2). \quad (\text{B13})$$

The second quantized Hamiltonian can be written as

$$\mathcal{H}' = \mathcal{H}'_L + \mathcal{H}'_{\text{int}} + \mathcal{H}'_{\text{dia}} = \int dr \psi^\dagger(r) H_m \psi(r), \quad (\text{B14})$$

namely,

$$\begin{aligned} \mathcal{H}' = & -\frac{1}{2m} \sum_{n,k} \sum_{n',k'} a_n b_n (\langle n,k | h_1^\dagger | n' - 2, k' \rangle \\ & + \langle n - 2, k | h_1 | n', k' \rangle) \\ & - \frac{1}{2} U \sum_{n,k} \sum_{n',k'} (a_n^2 \langle n - 2, k | n' - 2, k' \rangle - b_n^2 \langle n, k | n', k' \rangle) \\ & + \frac{1}{2m} \frac{U}{\gamma_1^2} \sum_{n,k} \sum_{n',k'} (a_n^2 \langle n - 2, k | h_2 | n' - 2, k' \rangle \\ & - b_n^2 \langle n, k | h_2^\dagger | n', k' \rangle). \end{aligned} \quad (\text{B15})$$

The above matrix elements can be calculated by using the relation [11,33]

$$\langle n, k | e^{-i\mathbf{q}\cdot\mathbf{r}} | n', k' \rangle = e^{-\frac{|q|^2 \ell_0^2}{4}} e^{-i\frac{q_y(k+k')\ell_0^2}{2}} \mathcal{G}_{n,n'}(\mathbf{q}) \delta_{k,k'-q_x}, \quad (\text{B16})$$

with

$$\mathcal{G}_{n,n'}(\mathbf{q}) = \sqrt{\frac{n'!}{n!}} \left[\frac{(q_x - iq_y)\ell_0}{\sqrt{2}} \right]^{n-n'} L_{n'}^{n-n'} \left(\frac{|q|^2 \ell_0^2}{2} \right). \quad (\text{B17})$$

For $n \geq n'$, $L_{n'}^{n-n'}(q)$ is the generalized Laguerre polynomial. For $n < n'$, $\mathcal{G}_{n,n'}(\mathbf{q})$ can be evaluated by using the identity $\mathcal{G}_{n,n'}(\mathbf{q}) = \mathcal{G}_{n',n}^*(-\mathbf{q})$.

The bosonized version of the single-particle and light-matter interaction Hamiltonian and Hamiltonian with diamagnetic term, i.e., $\mathcal{H}'_L$, $\mathcal{H}'_{\text{int}}$, and $\mathcal{H}'_{\text{dia}}$, can be obtained by evaluating the commutation relation between these Hamiltonians and the bosonic operator $d_\eta(d_\eta^\dagger)$ i.e., $[\mathcal{H}'_L, d_\eta(d_\eta^\dagger)]$, $[\mathcal{H}'_{\text{int}}, d_\eta(d_\eta^\dagger)]$, and $[\mathcal{H}'_{\text{dia}}, d_\eta(d_\eta^\dagger)]$.

For example, after a lengthy derivation, $\mathcal{H}'_L$ is written as

$$\begin{aligned} \mathcal{H}'_L = & 2\hbar\omega_0 \sum_{n,k} a_n b_n \sqrt{n(n-1)} c_{n,k}^\dagger c_{n,k} \\ & - \frac{U}{2} \sum_{n,k} (a_n^2 - b_n^2) c_{n,k}^\dagger c_{n,k} \\ & + \frac{U}{2\gamma_1} \hbar\omega_0 [a_n^2(2n-2) - b_n^2(2v+2)] c_{n,k}^\dagger c_{n,k}. \end{aligned} \quad (\text{B18})$$

The commutation relation between $\mathcal{H}'_L$ and $d_\eta(d_\eta^\dagger)$ is

$$[\mathcal{H}'_L, d_\eta] = -\omega_L d_\eta, \quad (\text{B19})$$

$$[\mathcal{H}'_L, d_\eta^\dagger] = \omega_L d_\eta^\dagger. \quad (\text{B20})$$

Then the bosonized version of the single-particle Hamiltonian is written as

$$\mathcal{H}_L = \sum_\eta \hbar\omega_L d_\eta^\dagger d_\eta, \quad (\text{B21})$$

where

$$\begin{aligned} \omega_L = & 2\omega_0 [a_v b_v \sqrt{v(v-1)} - a_{v-1} b_{v-1} \sqrt{(v-1)(v-2)}] \\ & - \frac{U}{2\hbar} [(a_v^2 - b_v^2) - (a_{v-1}^2 - b_{v-1}^2)] \\ & + \frac{U}{2\gamma_1} \omega_0 [a_v^2(2v-2) - b_v^2(2v+2) \\ & - a_{v-1}^2(2v-4) + b_{v-1}^2(2v)]. \end{aligned}$$

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