Tuning the Energy of a Polariton Condensate via Bias-Controlled Rabi Splitting

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(Received 21 May 2014; published 2 July 2014)

We introduce an electrically driven scheme to tune the polariton condensate energy in a high-finesse GaAs microcavity. In contrast to the conventional redshift observed in semiconductor quantum wells (QWs) under applied electrical bias arising from the quantum-confined Stark effect (QCSE), we report here the blueshift of a polariton condensate caused by controlled reduction of the Rabi splitting due to tunneling-induced charge buildup and fractional bleaching of QWs. At larger electrical bias, the QCSE becomes dominant, leading to a redshift in the linear regime, while in the nonlinear regime to the eventual quenching of the condensate emission. This ability to tune the polariton condensate energy brings within reach the realization of voltage-controlled polariton condensate devices and variable-wavelength sources of coherent light.

DOI: 10.1103/PhysRevApplied.2.014002

I. INTRODUCTION

Following the observation of Bose-Einstein condensation of exciton polaritons [1], half-light half-matter quasiparticles, attention has shifted towards their diverse quantum phenomena and the realization of polariton-based quantum information devices [2–7]. Despite recent progress in optical on-chip manipulation of polariton condensates in both planar and patterned microcavities [3–5,8,9], electrically programmable potentials for polariton condensates essential to future polariton devices have yet to be demonstrated. One critical element in this endeavor is the ability to imprint a potential energy landscape beyond conventional lateral confinement (which leads to strain and polarization pinning) or a built-in detuning gradient [5]. Optical imprinting relies on the injection of additional carriers to modify the potential energy landscape through a density-dependent blueshift of the polariton energy. These optically excited carriers can interact undesirably with the condensate, for example, by carrier heating, and have led to the breakdown of prototype polariton devices at high control powers [8,10]. As electric field control does not require the creation of additional carriers, it is a highly attractive candidate for polariton condensate control. Furthermore, electric-field-controlled polariton logic circuits have recently been suggested as the preferred route to the realization of universal polariton logic devices [11].

Here we introduce an electrically driven scheme to tune the energy of a polariton condensate in a high-finesse GaAs microcavity. In contrast to the expected redshift of the lower polariton branch due to the quantum-confined Stark effect (QCSE), application of electrical bias across the structure [12,13] blueshifts the polariton branch via controlled fractional reduction of the Rabi splitting [14].

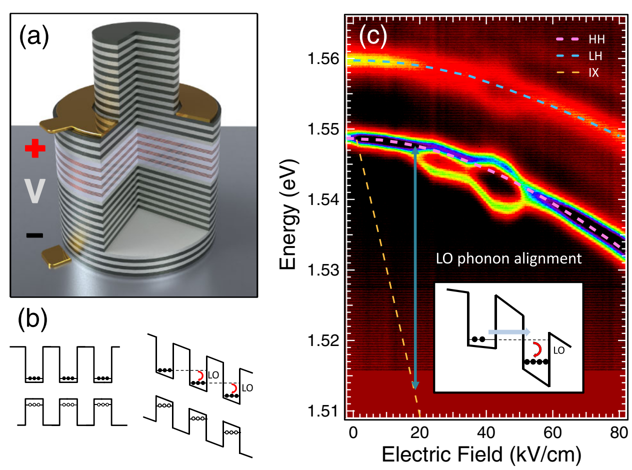

FIG. 1. (a) Cutaway schematic of a high-Q microcavity with four sets of triple QWs embedded in the cavity region. An electric field normal to the QW plane is applied through a top annular and bottom contact on the n-doped substrate. (b) Energy levels of one of the four sets of triple QWs under the applied field showing the LO-phonon-assisted tunneling process. (c) Reflectivity spectra showing bare HH and LH exciton tuning with the electric field. The inset shows modification of the local electric field at the QWs due to accumulation of carriers following irreversible LO-phonon-assisted tunneling.

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Reflectivity and photoluminescence (PL) measurements on the bare triple quantum well (QW) system used in the microcavity [Fig. 1(b)] reveal that the energy blueshift arises at electric fields at which longitudinal optical (LO)-assisted coupling of direct excitons to the same reservoir of indirect excitons occurs [15]. Such dissipative coupling through a nonreversible tunneling process induces charge buildup in the QWs, which modifies the local electric field, splitting the initially degenerate exciton energy into two bands. The presence of long-lived excitons in the rightmost QW introduces broadening to the excitonic resonance leading to fractional bleaching of QWs resulting in reduction of Rabi splitting and ultimately to the blueshift of the lower polariton branch.

II. SAMPLES DESCRIPTION

A schematic of the electrically contacted high-($Q \sim 16\,000$) finesse microcavity device is shown in Fig. 1(a). The $5/2\ \lambda$ microcavity comprises four sets of three 10-nm $Al_{0.3}Ga_{0.7}As/GaAs$ QWs incorporated between the top (bottom) distributed AlAs/$Al_{0.15}Ga_{0.85}As$ 32 (35) period Bragg reflectors (DBRs). The DBRs are undoped to circumvent optical losses that lower the cavity finesse [16]. An initial etch down to the n-doped substrate forms 200-μm diameter mesas and exposes the substrate for the deposition of the bottom contact. A second etch down to the last few layers of the DBR mirror before the cavity is used to produce an annular recess around the mesas onto which the top contact is evaporated. This process allows minimization of both the contact separation and the voltage required to produce the necessary electric fields. The high out-of-plane resistivity at the interface of the remaining DBR layers between the top contact and the cavity ensures the uniformity of the applied field across the mesa. A second sample having identical QWs without the top mirror is grown for a comparative study of QW exciton electric-field tuning in the absence of the strong coupling regime. The reflectivity stop band of the bottom DBR mirror is centered at the energy of the excitonic transitions to enhance absorption, creating pronounced dips in the reflectivity spectra.

III. EXPERIMENTAL RESULTS AND DISCUSSION

We initially characterize the triple QW sample under application of an electric field. The contour plot of the reflectivity spectra at normal incidence with increasing field [Fig. 1(c)] shows clear QCSE tuning of both light-hole (LH) and heavy-hole (HH) excitonic resonances with an applied field. Strikingly, at an applied field of 17 kV/cm the exciton peak splits into a double "bubble" structure which collapses back into a single exciton peak above 50 kV/cm. Furthermore, the excitonic bubble appears at a bias at which the electronic levels in neighboring QWs are aligned exactly with the energy difference of one LO phonon, as shown in Fig. 1(b). We attribute the appearance of such bubbles to the onset of resonant tunneling which redistributes electric charge across QWs, thus modifying the electric field locally at each QW as seen in the inset in Fig. 1(c). With an increasing applied field, the tunneling rates and total accumulated charge continuously readjust to preserve the one LO phonon energy difference. A second bubble opens up when electronic levels in all three QWs are aligned, inducing additional charge transfer. In this bias range, even larger local electric fields are produced, pulling further apart the exciton energies in different wells. PL measurements on the same structure reveal a significant broadening of the lowest excitonic resonance from initial 1.5 to 5 meV caused by free carriers scattering. In contrast, the upper bubble branch is minimally affected and retains both its oscillator strength and linewidth. Such an abrupt electric-field-controlled broadening of exciton transitions in only a fraction of QWs can be utilized to tune on demand the microcavity Rabi splitting producing energy blueshifts of the lower polariton branch.

To test this idea, we perform steady-state measurements, on a negatively detuned (approximately −2 meV) microcavity mesa under nonresonant excitation at 1.649 eV by a continuous-wave Ti:sapphire laser focused through a microscope objective $NA = 0.55$ to produce a 40-μm diameter spot. The far-field PL emission in the condensate regime is collected by imaging the Fourier plane onto the slits of a spectrometer coupled to a liquid-nitrogen-cooled CCD.

Figure 2 shows the steady-state far-field emission for optical pumping at double the condensation threshold with an increasing electric field (left to right). At zero applied field [Fig. 2(a)], the PL is dominated by polariton condensate emission normal to the sample plane (polariton wave vector $k = 0$). The superimposed lower-intensity lower polariton branch emission originates from outer parts of the excitation spot where carrier densities are significantly smaller, producing a minimal pump-induced blueshift. Notably, with increasing field and for the chosen sample detuning, there is a distinct shift of the condensate to higher energy, with this shift reaching 0.85 meV at an applied field of 27 kV/cm. Contrary to the normally expected redshift of the lower polariton branch caused by the lowering of the exciton energy due to QCSE, we observe the polariton condensate energy blueshift in Fig. 2. At larger electrical bias, the QCSE becomes dominant, leading to the redshift of the lower polariton branch and the eventual loss of the condensation regime.

To investigate further the origin of this energy blueshift, PL emission collected normal to the sample surface ($k = 0$) is plotted for different electric fields in both the linear [Fig. 3(a)] and condensation regimes [Fig. 3(b)]. The energies of the lower polariton branch (LPB) and cavity mode (CM) are marked by the solid and dashed lines, respectively. The polariton condensate (PC) emission is

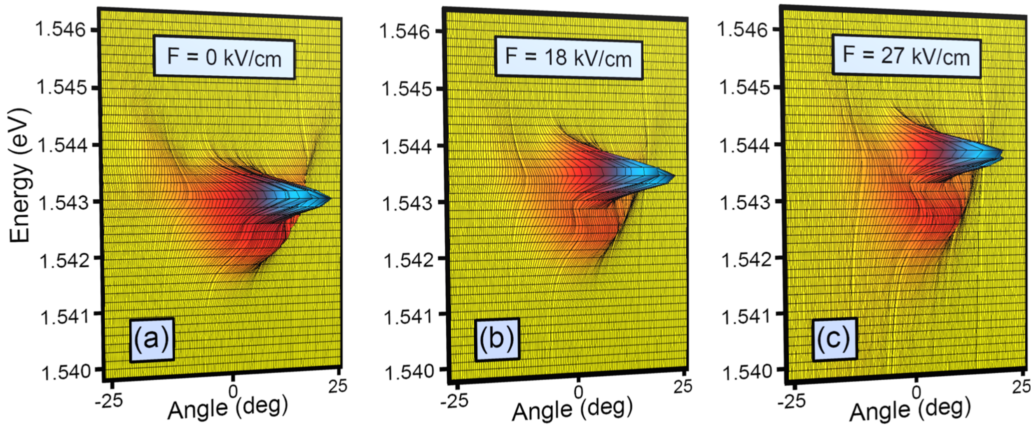

FIG. 2. Angle-dependent polariton far-field emission under nonresonant continuous wave excitation recorded on the logarithmic scale. A gradual blueshift of polariton condensate energy is observed when the electric field is applied: (a) zero bias, (b) 15 kV/cm, and (c) 24 kV/cm. Superimposed weak emission arising from noncondensed low-excitation density areas of the sample surrounding the central spot shows a similar energy blueshift.

also marked with a solid line. In the linear regime [Fig. 3(a)] with increasing electric field, the lower polariton branch first blueshifts by 0.8 meV and then undergoes a redshift by 1.5 meV at 37 kV/cm. Similar results are obtained in the condensation regime [Fig. 3(b)] with an observed blueshift of the condensate energy at low fields and subsequent loss of the polariton condensation regime at higher fields. The energy shift under applied bias is unique to laser emission from a microcavity in the strong coupling regime as opposed to vertical-cavity surface-emitting laser-type lasing where emission arises from the cavity mode and is not tunable with the application of electric fields. The eventual loss of the polariton lasing regime is caused by the increase of the condensation threshold at higher fields. Increasing the excitation density reinstates the lasing regime; however, it results in smaller overall shifts due to screening of the bias field.

This detailed tuning of the normal incidence emission at different electric fields below the condensation threshold shows a number of distinct features (Fig. 4). For convenience, the bare exciton electric-field tuning curves extracted from reflectivity spectra of Fig. 1(c) as well as the cavity mode energy are superposed on the same graph. Because of inhomogeneities in the microcavity QW growth (most likely caused by QW monolayer fluctuations), not all four sets of triple QWs have the same energy but are split into two subsets separated by 1.5 meV as seen by the dashed curves DX1 and DX2. Excellent fits to the polariton emission spectra for varying electric field are obtained by using a coupled harmonic oscillator model, with the only input parameters being the measured bare exciton energy positions and linewidths of each excitonic sub-branch at different bias. Notably, at an electric field of 17 kV/cm, a clear reduction of the Rabi splitting can be seen predominately associated with the broadening of the low-energy excitonic line due to excess carrier buildup

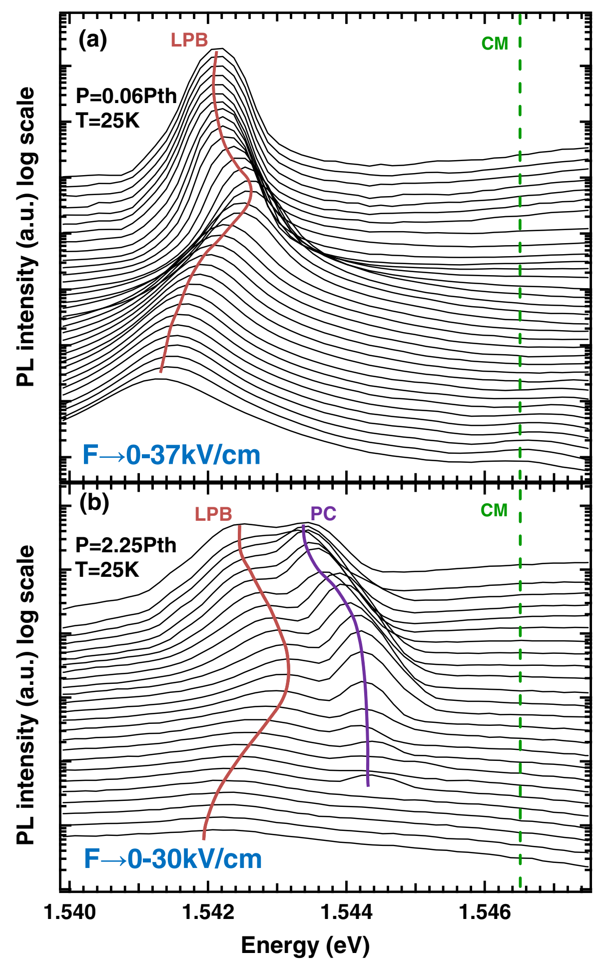

FIG. 3. PL spectra recorded at normal incidence to the sample plane for an increasing electric field (a) below and (b) above the condensation threshold, respectively. Low-density lower polariton branch emissions arising from the outer part of excitation spot are marked with (LPB), whereas bias-dependent condensate energy is marked by a polariton condensate (PC) line.

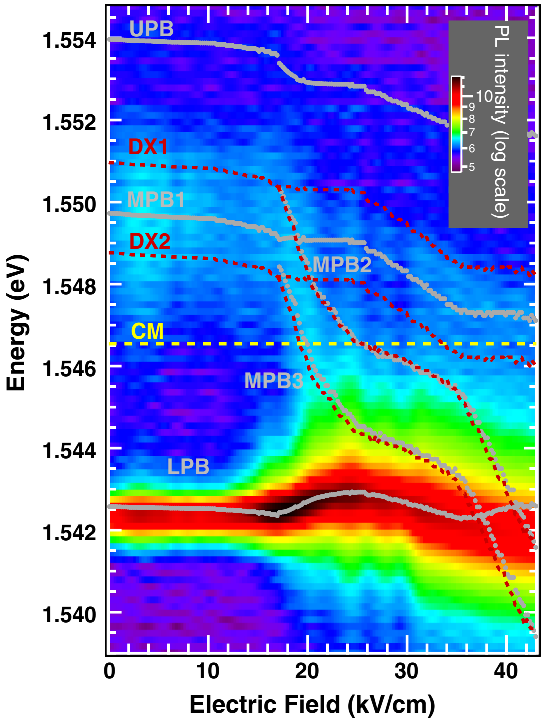

FIG. 4. Normal incidence PL emission with varying applied bias. Bare uncoupled exciton branches are shown by dotted line. LPB, MPB, and UPB polariton emission peaks are fitted by using the coupled harmonic oscillator model using the exciton energy and linewidths measured from PL measurements on bare QWs. A LPB energy blueshift occurs due to Rabi-splitting reduction caused by free-carrier-induced broadening of the low-lying exciton branch at the onset of LO-phonon-assisted tunneling.

caused by LO-phonon-assisted tunneling. Because of the dissipative nature of this tunneling process, the charge transfer is irreversible and accumulation of free carrier occurs only in the right QWs. This lifts degeneracy in exciton energy, oscillator strength, and nonradiative broadening between three QWs embedded in the microcavity. It eventually leads to the ensemble of peculiar phenomena we observe, including the bubble formation and the Rabi-splitting drop. Thus, the number of QWs participating directly in the strong coupling reduces from 12 to 9, with a reduction in Rabi splitting of $\sqrt{9/12} = 85\%$ matching the experiments well. This result demonstrates how the growth design is able to precisely set the field-tuning blueshifts, by providing the ratio of QWs contributing to strong coupling before or after the tunneling switches on. In the same way that field drops across superlattices can be nonuniform, providing a panoply of nonlinear switching phenomena [17], the nonuniform fields induced by appropriate QW engineering of the microcavity allow us to select the field-induced tuning of microcavities.

IV. CONCLUSIONS

In conclusion, we show clear energy tuning of the exciton-polariton condensate under application of electric bias. The observed surprising initial energy blueshift arises from the reduction of microcavity Rabi splitting due to a field-dependent LO-phonon-assisted tunneling process which causes charging and subsequently broadening of the excitonic transition in electron-accumulating QWs. Such an adjustable bias-dependent switching of only a fraction of QWs in a microcavity allows tuning of polariton condensate energies via the bias-controlled Rabi splitting. The observed energy shift is comparable with the blueshift required in polariton devices currently operating under optical addressing [8]. The scheme developed here will thus allow direct gating of polariton condensate flows and will find application in bias-controlled polariton condensate transistors and circuit devices [18,19]. Furthermore, utilization of such schemes in large and controllable Rabi-split systems based, for example, on organic or GaN materials could lead to the realization of widely tunable polariton laser diode devices.

ACKNOWLEDGMENTS

We acknowledge EU ITN Grant No. INDEX 289968, Greek GSRT ARISTEIA program Apollo, and POLAFLOW ERC Starting Grant. P. S. gratefully acknowledges financial support from the Leverhulme Trust. A. K. thanks the EPSRC Established Career Program. P. T. acknowledges the EU Social Fund and Greek National Resources EPEAEK II-HRAKLEITOS II.

[1] J. Kasprzak, M. Richard, S. Kundermann, A. Baas, P. Jeambrun, J. M. Keeling, F. M. Marchetti, M. H. Szymanska, R. Andre, J. L. Staehli, V. Savona, P. B. Littlewood, B. Deveaud, and Le S. Dang, Bose-Einstein condensation of exciton polaritons, Nature (London) **443**, 409 (2006).

[2] A. Amo, D. Sanvitto, F. P. Laussy, D. Ballarini, E. Del Valle, M. D. Martin, A. Lemaitre, J. Bloch, D. N. Krizhanovskii, M. Skolnick, C. Tejedor, and L. Vina, Collective fluid dynamics of a polariton condensate in a semiconductor microcavity, Nature (London) **457**, 291 (2009).

[3] K. G. Lagoudakis, T. Ostatnicky, A. V. Kavokin, Y. G. Rubo, R. Andre, and B. Deveaud-Pledran, Observation of half-quantum vortices in an exciton-polariton condensate, Science **326**, 974 (2009).

[4] E. Wertz, L. Ferrier, D. D. Solnyshkov, R. Johne, D. Sanvitto, A. Lematre, I. Sagnes, R. Grousson, A. V. Kavokin, P. Senellart, G. Malpuech, and J. Bloch, Spontaneous formation and optical manipulation of extended polariton condensates, Nat. Phys. **6**, 860 (2010).

[5] G. Tosi, G. Christmann, N. G. Berloff, P. Tsotsis, T. Gao, Z. Hatzopoulos, P. G. Savvidis, and J. J. Baumberg, Sculpting oscillators with light within a nonlinear quantum fluid, Nat. Phys. **8**, 190 (2012).

[6] S. I. Tsintzos, N. T. Pelekanos, G. Konstantinidis, Z. Hatzopoulos, and P. G. Savvidis, A GaAs polariton

light-emitting diode operating near room temperature, Nature (London) **453**, 372 (2008).
[7] A. Amo, T. C. H. Liew, C. Adrados, R. Houdre, E. Giacobino, A. V. Kavokin, and A. Bramati, Exciton-polariton spin switches, Nat. Photonics **4**, 361 (2010).
[8] T. Gao, P. S. Eldridge, T. C. H. Liew, S. I. Tsintzos, G. Stavrinidis, G. Deligeorgis, Z. Hatzopoulos, and P. G. Savvidis, Polariton condensate transistor switch, Phys. Rev. B **85**, 235102 (2012).
[9] D. Ballarini, M. De Giorgi, E. Cancellieri, R. Houdre, E. Giacobino, R. Cingolani, A. Bramati, G. Gigli, and D. Sanvitto, All-optical polariton transistor, Nat. Commun. **4**, 1778 (2013).
[10] H. S. Nguyen, D. Vishnevsky, C. Sturm, D. Tanese, D. Solnyshkov, E. Galopin, A. Lemaitre, I. Sagnes, A. Amo, G. Malpuech, and J. Bloch, Realization of a double-barrier resonant tunneling diode for cavity polaritons, Phys. Rev. Lett. **110**, 236601 (2013).
[11] T. Espinosa-Ortega and T. C. H. Liew, Complete architecture of integrated photonic circuits based on AND and NOT logic gates of exciton polaritons in semiconductor microcavities, Phys. Rev. B **87**, 195305 (2013).
[12] G. Bastard, E. E. Mendez, L. L. Chang, and L. Esaki, Variational calculations on a quantum well in an electric field, Phys. Rev. B **28**, 3241 (1983).
[13] D. A. B. Miller, D. S. Chemla, T. C. Damen, A. C. Gossard, W. Wiegmann, T. H. Wood, and C. A. Burrus, Band-edge electroabsorption in quantum well structures: The quantum-confined Stark effect, Phys. Rev. Lett. **53**, 2173 (1984).
[14] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevApplied.2.014002 for an angle-dependent far-field emission video with a varying electric field recorded on the logarithmic intensity scale.
[15] T. H. Wang, X. B. Mei, C. Jiang, Y. Huang, J. M. Zhou, X. G. Huang, C. G. Cai, Z. X. Yu, C. P. Luo, J. Y. Xu, and Z. Y. Xu, LO-phonon-assisted tunneling in asymmetric double-well structures with thick barriers, Phys. Rev. B **46**, 16160 (1992).
[16] P. Tsotsis, P. S. Eldridge, T. Gao, S. I. Tsintzos, Z. Hatzopoulos, and P. G. Savvidis, Lasing threshold doubling at the crossover from strong to weak coupling regime in GaAs microcavity, New J. Phys. **14**, 023060 (2012).
[17] A. Wacker, Phys. Rep. **357**, 1 (2002).
[18] C. Anton, T. C. H. Liew, G. Tosi, M. D. Martn, T. Gao, Z. Hatzopoulos, P. S. Eldridge, P. G. Savvidis, and L. Vina, Dynamics of a polariton condensate transistor switch, Appl. Phys. Lett. **101**, 261116 (2012).
[19] C. Anton, T. C. H. Liew, J. Cuadra, M. D. Martn, P. S. Eldridge, Z. Hatzopoulos, G. Stavrinidis, P. G. Savvidis, and L. Vina, Quantum reflections and shunting of polariton condensate wave trains: Implementation of a logic AND gate, Phys. Rev. B **88**, 245307 (2013).