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Gate Tunable Coupling of Epsilon-Near-Zero and Plasmonic Modes

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In this work, an active tuning of the coupling strength in a strongly coupled system comprised of a thin epsilon-near-zero material and gold nanorods as plasmonic resonators is demonstrated. A novel gating scheme is developed where an ionic liquid is employed to bias the coupled system and tune the coupling in transmission mode. A significant tuning of the coupled resonance up to 30 nm is observed by changing the bias voltages from 0 to 4.5 V. This control mechanism on strong coupling opens exciting opportunities for various disruptive applications by offering advanced control and tunability on strongly coupled systems.

1. Introduction

The strong-coupling phenomenon has been exploited in different systems as it enables the hybrid modes with an increased degree of freedom. Metallic structures are widely employed to achieve strong coupling owing to their ability to confine the electromagnetic field to deep subwavelength volumes.[1,2] The strong coupling manifests itself as splitting of the resonance into two hybrid states in the spectral domain where the separation defines the coupling strength.[3,4] However, the low-quality factor of metallic nanostructures put the limitation to achieve strong coupling. Fortunately, this limitation can be lifted by employing epsilon-near-zero (ENZ) materials that offer subwavelength field enhancement[5,6] or quantum heterostructures that offer large dipole transitions.[7] ENZ materials such as highly doped transparent conducting oxide (TCO) can confine the electromagnetic fields into the deep subwavelength volume at the wavelength where the real part of the dielectric permittivity crosses zero. Furthermore, it has been demonstrated that ultrathin ENZ films support ENZ modes[8] with strong electric field confinement and excitation of the enhanced local density of optical states. Integration of metallic resonators with such materials to realize strong coupling provides advantages in spectral scalability and device miniaturization. The combination of ENZ film with plasmonic nanostructures has shown striking performance in enhanced nonlinearity,[9–11] subwavelength tunneling, beam shaping, and steering.[12,13] In a similar direction, previous studies with ENZ-plasmonic nanostructures hybrid system have shown to exhibit strong coupling.[14,15] Moreover, the strongly coupled mode is modified passively by changing the geometrical parameters of the antenna[5] and changing the doping levels in quantum well-plasmonic resonator system.[7]

In recent studies, various applications where ENZ material is integrated with different photonics platforms[16,17] has been demonstrated. For example, the inclusion of ENZ material with metallic structures exhibited diverse functionalities like tunable absorbers,[18,19] beam steering,[20] waveguides[21] and integrated plasmonic devices.[22] It is highly desired to achieve an active tuning in a strongly coupled system that offers the coupling of light–matter interactions to uplift the application capability and achieve advanced functionality. Although, the permittivity of ENZ materials is utilized in several studies to tune the optical properties by electrical biasing,[18,20,22] so far, the tuning of strong coupling has been demonstrated only by passive means. While on the other hand, most of the electrical biasing schemes are limited to operating in reflection mode due to the presence of a back reflector.

Motivated by the exciting possibilities and opportunities opened up by the strong coupling of ENZ modes and plasmonic modes for different applications, in this work, we introduce the dynamic tuning of this coupling with a novel gating scheme using ionic liquid. We experimentally demonstrated the electrical tuning of the coupling strength in the hybrid structure consisting of a gold nanorod array on a thin film of indium tin oxide (ITO). Besides, all the results presented here are compared with numerical simulations, demonstrating good agreement with the design and experiments. The hybridization of ENZ mode on the ITO thin film and localized surface plasmon (LSP) mode on the nanorods manifests as splitting in the transmission spectra that signifies the strong coupling regime. In our work, we employed a novel gating scheme that operates in transmission mode without the requirement of a back metallic gating layer. Notably, same design can be operated to achieve the tuning in reflection mode which can be enhanced further introducing a back reflector. This diversifies the application range of our gating scheme.

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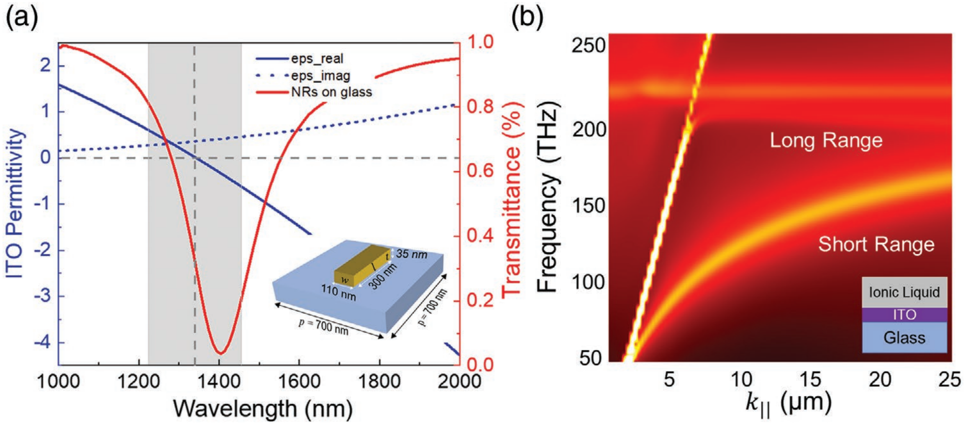

Figure 1. a) Experimentally recorded complex permittivity of 40 nm thin ITO film (blue solid and dotted line). Simulated transmittance spectra of the optimized nanorods (NRs) array on a glass substrate (red solid line) with the ionic liquid background and the inset shows schematic of the unit cell consisting of gold nanorod fabricated on a glass substrate. b) Dispersion relation of the ionic liquid-ENZ-glass three-layered system.

2. Results and Discussion

ITO is compatible with semiconductor processing and has been widely used in commercial devices such as displays, solar panels, etc. ITO is adopted as an active material for this study due to its remarkable properties like tunable permittivity,[23–25] lower optical losses in combination with a large electrical conductivity. To characterize the dispersion of ITO, we performed the ellipsometry measurement on 40 nm ITO thin film and extracted the complex permittivity. In **Figure 1**a, the real and imaginary parts of the ITO thin film's permittivity are shown as the blue solid and dotted line, respectively. The zero permittivity ($\varepsilon \approx 0$) region that spans from 1230 to 1450 nm is highlighted in Figure 1a. The dispersion relation for ITO thin film embedded in ionic liquid (inset Figure 1b) was calculated using the finite-difference time-domain (FDTD) technique. The simulation was performed using Bloch boundary conditions and experimentally recorded permittivity of ITO is fed into the simulation. The cloud of dipoles with different phase and orientations were placed in the simulation region. To obtain the dispersion graph (Figure 1b), we swept different values of wave vector k_x (i.e., $k_{||}$) and calculated the corresponding eigen-frequencies. In this process, the simulation time was kept sufficiently long enough to attenuate the modes which are not supported by our structure. In Figure 1b, the short-range and long-range surface plasmon modes of ITO thin films are marked. The dispersion of long-range mode is nearly flat for a large range of transverse wave vectors ($k_{||}$) near 220 THz frequency. This is consistent with the ellipsometry result shown in Figure 1a, which identifies the ENZ wavelength at 1340 nm (≈223 THz). Therefore, the long-range surface plasmon is referred as the ENZ mode.[14] However, due to the impedance mismatch at the interface between the ionic liquid and the ENZ medium, the ENZ modes' excitation is inefficient.

Whereas the plasmonic nanostructures confine the incident electromagnetic field at a deep subwavelength scales due to excitation of LSP which leads to a remarkable enhancement of the local field. Therefore, in order to enable high electromagnetic fields inside the ENZ film and generate the LSP mode, we have used Au nanorods in this study. To optimize Au nanorod's dimension such that their plasmonic resonance is in the ENZ region of the ITO film, we performed FDTD numerical simulation using Ansys Lumerical FDTD Solutions. The linearly polarized plane wave source of wavelengths 1000–2000 nm was utilized to excite the nanorod with polarization along the nanorod's long edge. The gold (Au) was modeled by using Johnson and Christy material data[26] from the in build material library of Lumerical. In the optimization process, we kept the background refractive index to 1.41 to emulate ionic liquid ambiance. As will be shown, the ionic liquid is needed for tunability. The simulated transmission plot for the optimized nanorod is shown in Figure 1a (solid red line). The transmission spectra for various other dimensions are presented in Figure S4a, Supporting Information. The dimensions of the optimized nanorod antenna are length (l) = 300 nm, width (w) = 110 nm, and thickness (t) = 35 nm as marked in the inset of Figure 1a. We kept the square periodicity (p) of 700 nm to maximize the density of the nanorod while avoiding the cross-talks between them.

Once we identified the modes, the strong coupling of these modes is investigated by bringing them to the same platform. We experimentally recorded the ENZ-integrated plasmonic hybrid system's transmission spectra with (solid red line) and without ionic liquid (solid black line), as shown in **Figure 2**a. The presence of the ionic liquid red-shifts the overall spectra due to an increase in background refractive index ($n_{\text{air}} = 1$ to $n_{\text{ionicliquid}} \approx 1.41$). In the optimization process of the plasmonic antenna, the background refractive index is included as 1.41. As a result, its resonance position is red-shifted to the ENZ region. Therefore, in Figure 2a, both upper and lower polaritons are clearly visible, which shows that in the presence of ionic liquid, the system exhibits strong coupling that manifests as apparent spectral splitting (solid red line). In contrast, the system without ionic liquid is only weakly coupled. The inset of Figure 2a represents the scanning electron microscope (SEM) image of the plasmonic structures (top view), showing the geometry of the gold antennas (square lattice Au nanorods). Note that fabricated nanoantennas are slightly different from the simulated

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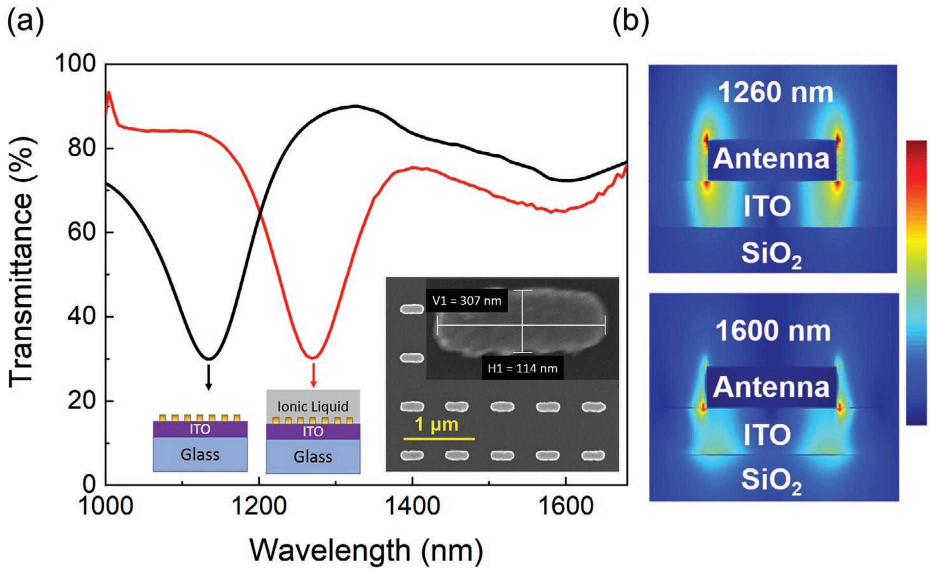

Figure 2. a) Experimentally recorded transmission spectra of ITO integrated plasmonic antenna hybrid system with and without ionic liquid and inset shows SEM image of the nanorods (top view) showing the gold antennas (square lattice Au nanorods). b) Electric field (E_x) distribution at lower polariton (1260 nm) and upper polariton (1600 nm) for nanorod–ITO integrated system.

ones due to the fabrication imperfections, which results in antenna edges being rounded off.

The electric field confinement in the plasmonic structure and ENZ layer of the strongly coupled system shows the hybrid characteristics' features. The antennas placed on the ITO film allow the electric field to be concentrated inside the ITO layer resulting in enhancement of the field intensity at polaritonic wavelengths. Figure 2b shows electric field distribution of the ENZ-integrated system, calculated at lower polariton (1260 nm) and upper polariton (1600 nm) that display a mixture of localized plasmons and ENZ mode confinement. The resonance mode of the nanorod used in our study is electric in nature, as the calculated surface currents (Section S2, Supporting Information) show the dipole like oscillations of the modes.

To achieve the dynamic tunability of ENZ-LSP coupling, we designed a novel electrical gating scheme that allows transmission measurements providing an extra dimension of performance. In previous studies to achieve the tunability, the gating schemes require the metallic plate to be present at the backside of metadevices which severely limits the mode of operation to be only in reflection.[18,20,22] In contrast, our designed gating is versatile in nature, which is compatible with operating in both the transmission and reflection modes. The schematic of the designed architecture to achieve the electrical gating in transmission mode is shown in **Figure** 3a. The gating assembly consists of two plates: bottom plate includes the substrate with 40 nm ITO thin film followed by a square array of gold nanorods as shown in Figure 2a inset, and the top plate is a bare glass. On both of the substrates, we created a gold periphery window in order to connect copper wires with good ohmic contact. To avoid a short circuit, these two substrates are separated from each other by insulating tapes. The ionic liquid[27] is sandwiched between these two plates, which serve as media to accumulate ions at ITO and ionic liquid interface upon electrical biasing.

The active tuning of strongly coupled ENZ-LSP modes is achieved by electrical biasing. The experimentally acquired spectra for various bias voltages are shown in Figure 3b. For clarity, in this figure, we have shown only the lower polariton, and the individual spectrum for different voltages is offset vertically. By increasing the bias voltage from 0 to 4.5 V, the lower polariton shifts from 1260 to 1288 nm, while there is a subtle change in the upper polariton as shown in Section S3, Supporting Information. As the electric field confinement at lower polariton (as shown in Figure 2b) is more intense than the upper polariton, the lower polariton shows better tuning with respect to upper polariton. Additionally, the change in the permittivity with different external bias voltages is more effective closer to the ENZ wavelength where we observe the lower polariton.

Since the splitting between the upper and lower polariton is proportional to the coupling strength,[28] change in the lower polariton's wavelength represents the change in the coupling strength of the strongly coupled system. Further, this coupling's tunability was verified using different nanorod dimensions, as shown in Figure S4, Supporting Information. The nanorods' size is considered so that the plasmon resonance still resides inside the ENZ region (gray shaded region of Figure 1a), which ensures the system remains in the strong coupling regime.

The achieved tunability lies under the occurred modification in the permittivity of the ITO film as a response to the applied DC voltage. Under the presence of the electrical bias, the ions in an ionic liquid get polarized and accumulate at both the top and bottom interfaces. The increase in the voltage results in an increment of charge concentration at the interface of ITO and ionic liquid. These assembled ions at the interface induce charge increment that in turn changes the permittivity of the ITO. By increasing the bias voltage, the density of the ions increases and saturates after a threshold voltage which

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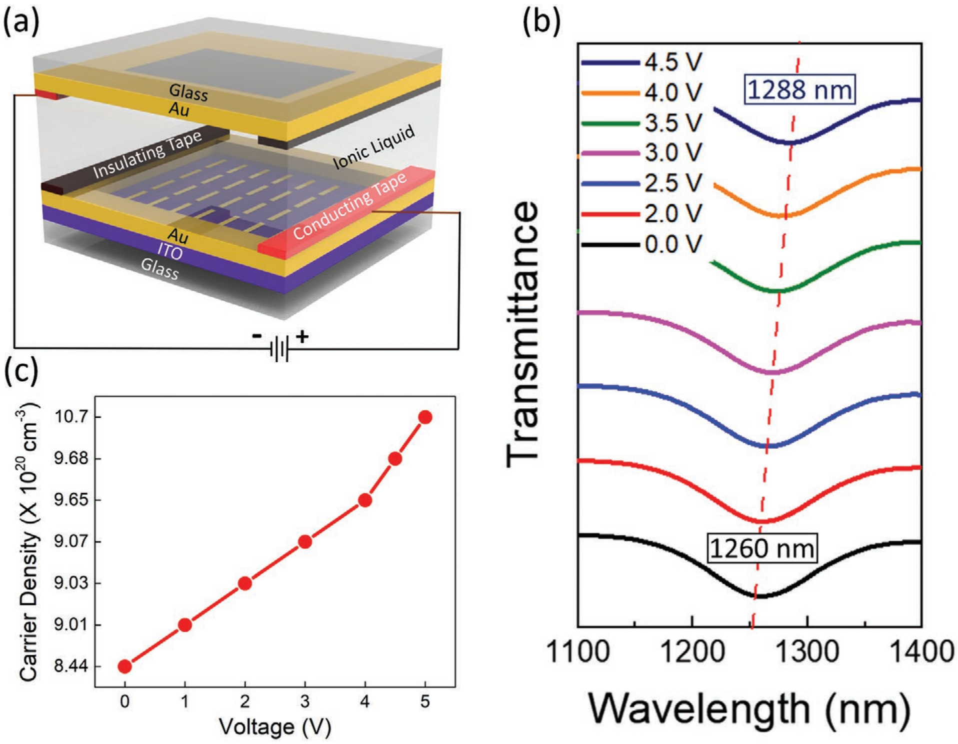

Figure 3. a) The schematic of a novel gating architecture for the electrical gating in transmission mode. b) Experimentally recorded transmission spectra near lower polariton wavelength at different bias voltages (for clarity, individual spectra are offset vertically). c) Experimentally recorded carrier densities in ITO for various voltages using Hall measurement system.

is 4.5 V in our case. To validate this point, we performed the Hall measurement on the ITO gated system (Figure S6a, Supporting Information) with developed biasing architecture. Figure 3c shows the measured carrier concentration in ITO for various voltages using the Hall measurement system. It is evident that by increasing the voltage, the carrier concentration changes due to the polarized ionic liquid. This enhanced carrier density changes the ITO permittivity, allowing us to tune the strong coupling between plasmonic nanorods and the ENZ layer. Thus, changing the bias voltage offers electrical tunability of ENZ-LSP hybrid modes' coupling strength that manifests as a change in the spectral splitting, as shown in Figure S3, Supporting Information.

3. Conclusion

We developed an electrically tunable strongly coupled ENZ-LSP system based on a novel gating scheme. The strong coupling between optimally designed plasmonic nanorods and thin ITO film manifests as splitting in transmission spectra of the structure. The developed ionic liquid based gating system allows tuning the permittivity of the ITO film. Subsequently, the coupling strength between ENZ and LSP mode gets changed by changing the applied DC voltage. We observed up to 30 nm of shift in lower polariton. Since the lower and upper polaritons' frequency difference quantifies the coupling strength, the spectral shift of the lower polariton via applied voltage signifies tuning the coupling strength. Dynamically tunable strongly coupled systems are essential for exploring fundamental physics and developing applications. Therefore, our study opens up a new avenue to explore many new phenomena and enhance integrated nanodevice performance by offering advanced control and tunability on strongly coupled systems. Additionally, the system exhibits reversible behavior and our gating scheme can be implemented in both transmission and reflection mode thus broadens the application horizon of photonic devices toward tunable flat lens, beam steering, and transmissive spatial light modulators which is vital for LIDAR and wireless communication.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Keywords

epsilon-near-zero mode, epsilon-near-zero materials, gating, indium tin oxide, plasmonic mode, strong coupling

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[1] A. Benz, S. Campione, S. Liu, I. Montaño, J. F. Klem, A. Allerman, J. R. Wendt, M. B. Sinclair, F. Capolino, I. Brener, *Nat. Commun.* **2013**, *4*.

[2] G. Scalari, C. Maissen, D. Turčinková, D. Hagenmüller, S. De Liberato, C. Ciuti, C. Reichl, D. Schuh, W. Wegscheider, M. Beck, J. Faist, *Science* **2012**, *335*, 1323.

[3] S. D. Liberato, C. Ciuti, I. Carusotto, *Phys. Rev. Lett.* **2007**, *98*, 103602.

[4] P. Törmä, W. L. Barnes, *Rep. Prog. Phys.* **2014**, *78*, 013901.

[5] S. Campione, J. R. Wendt, G. A. Keeler, T. S. Luk, *ACS Photonics* **2016**, *3*, 293.

[6] B. C. Yildiz, H. Caglayan, *Phys. Rev. B* **2020**, *102*, 165303.

[7] A. Benz, S. Campione, M. W. Moseley, J. J. Wierer, A. A. Allerman, J. R. Wendt, I. Brener, *ACS Photonics* **2014**, *1*, 906.

[8] S. Campione, I. Brener, F. Marquier, *Phys. Rev. B* **2015**, *91*, 121408.

[9] M. Z. Alam, S. A. Schulz, J. Upham, I. De Leon, R. W. Boyd, *Nat. Photonics* **2018**, *12*, 79.

[10] J. Bohn, T. S. Luk, C. Tollerton, S. W. Hutchings, I. Brener, S. Horsley, W. L. Barnes, E. Hendry, *Nat. Commun.* **2021**, *12*, 1017.

[11] Y. Yang, J. Lu, A. Manjavacas, T. S. Luk, H. Liu, K. Kelley, J.-P. Maria, E. L. Runnerstrom, M. B. Sinclair, S. Ghimire, I. Brener, *Nat. Phys.* **2019**, *15*, 1022.

[12] X. Niu, X. Hu, S. Chu, Q. Gong, *Adv. Opt. Mater.* **2018**, *6*, 21951071.

[13] Y.-W. Huang, H. W. H. Lee, R. Sokhoyan, R. A. Pala, K. Thyagarajan, S. Han, D. P. Tsai, H. A. Atwater, *Nano Lett.* **2016**, *16*, 5319.

[14] V. Bruno, C. DeVault, S. Vezzoli, Z. Kudyshev, T. Huq, S. Mignuzzi, A. Jacassi, S. Saha, Y. D. Shah, S. A. Maier, D. R. S. Cumming, A. Boltasseva, M. Ferrera, M. Clerici, D. Faccio, R. Sapienza, V. M. Shalaev, *Phys. Rev. Lett.* **2020**, *124*, 043902.

[15] K. Manukyan, M. Z. Alam, C. Liu, K. Pang, H. Song, Z. Zhao, M. Tur, R. W. Boyd, A. E. Willner, *Appl. Phys. Lett.* **2021**, *118*, 241102.

[16] C.-W. Lee, H. J. Choi, H. Jeong, *Nano Convergence* **2020**, *7*, 3.

[17] A. Forouzmand, M. M. Salary, G. K. Shirmanesh, R. Sokhoyan, H. A. Atwater, H. Mosallaei, *Nanophotonics* **2019**, *8*, 415.

[18] J. Park, J.-H. Kang, X. Liu, M. L. Brongersma, *Sci. Rep.* **2015**, *5*, 15754.

[19] J. R. Hendrickson, S. Vangala, C. Dass, R. Gibson, J. Goldsmith, K. Leedy, D. E. Walker, J. W. Cleary, W. Kim, J. Guo, *ACS Photonics* **2018**, *5*, 776.

[20] G. Kafaie Shirmanesh, R. Sokhoyan, R. A. Pala, H. A. Atwater, *Nano Lett.* **2018**, *18*, 2957.

[21] L. Tao, A. Anopchenko, S. Gurung, J. Zhang, H. W. H. Lee, *Sci. Rep.* **2019**, *9*, 2789.

[22] E. Feigenbaum, K. Diest, H. A. Atwater, *Nano Lett.* **2010**, *10*, 2111. pMID: 20481480.

[23] A. Calá Lesina, D. Goodwill, E. Bernier, L. Ramunno, P. Berini, *IEEE J. Sel. Top. Quantum Electron.* **2021**, *27*, 4700116.

[24] A. Howes, W. Wang, I. Kravchenko, J. Valentine, *Optica* **2018**, *5*, 787.

[25] G. T. Papadakis, H. A. Atwater, *Phys. Rev. B* **2015**, *92*, 184101.

[26] P. B. Johnson, R. W. Christy, *Phys. Rev. B* **1972**, *6*, 4370.

[27] O. Ozdemir, A. M. Aygar, O. Balci, C. Kocabas, H. Caglayan, E. Ozbay, *Carbon* **2016**, *108*, 515.

[28] C. Ciuti, G. Bastard, I. Carusotto, *Phys. Rev. B* **2005**, *72*, 115303.

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