Journal of Materials Chemistry C

Accepted Manuscript



This is an *Accepted Manuscript*, which has been through the Royal Society of Chemistry peer review process and has been accepted for publication.

Accepted Manuscripts are published online shortly after acceptance, before technical editing, formatting and proof reading. Using this free service, authors can make their results available to the community, in citable form, before we publish the edited article. We will replace this Accepted Manuscript with the edited and formatted Advance Article as soon as it is available.

You can find more information about *Accepted Manuscripts* in the **Information for Authors**.

Please note that technical editing may introduce minor changes to the text and/or graphics, which may alter content. The journal's standard <u>Terms & Conditions</u> and the <u>Ethical guidelines</u> still apply. In no event shall the Royal Society of Chemistry be held responsible for any errors or omissions in this *Accepted Manuscript* or any consequences arising from the use of any information it contains.



www.rsc.org/materialsC

HIGHLIGHT

Cite this: DOI: 10.1039/c0xx00000x

www.rsc.org/xxxxx

Polariton Laser Can Now Be Pumped Electrically

Cong Wei and Yong Sheng Zhao*

Received (in XXX, XXX) Xth XXXXXXXX 20XX, Accepted Xth XXXXXXXX 20XX DOI: 10.1039/b000000x

⁵ Exciton-polaritons arising from the strong coupling between excitons and photons are composite bosons which are half light and half matter. Due to their relatively light effective mass (seven orders of magnitude less than the hydrogen atom mass), they can potentially condense at temperatures much higher than those required for atom Bose-Einstein condensations (tens of nanokelvins), which make them not only a perfect model for fundamental studies of dynamical Bose-Einstein condensates but also a suitable

¹⁰ system to design the novel optical components. Various polaritonic devices such as polariton parametric amplifiers, optically pumped polariton lasers and polariton light-emitting diodes have been achieved. With the recently reported electrically pumped polariton lasers added to the list, the polaritons seem to have made their way out of the laboratory and have a bright, white future ahead.

Introduction

- ¹⁵ Lasers are desired in many areas ranging from on-chip optical communication to biomedical imaging, high sensitive sensors and big gamut full colour display, as they present the means to deliver powerful, coherent and directional high-frequency electromagnetic wave. Conventional laser based on the stimulated
- ²⁰ emission of photons sets stringent requirements on the minimum amount of energy necessary for its operation. In comparison, exciton-polaritons, arising from the strong coupling between excitons and photons,¹ can undergo Bose-Einstein condensation and emit the coherent light directly from their condensate state.
- ²⁵ This process, called "polariton lasing", requires no conventional population inversion scheme and offers a more effective way to generate the coherent light at very low threshold.²

By now, most of the polariton lasers were achieved under optical pumping in a microcavity built by placing a ³⁰ semiconductor layer between two mirrors.³⁻⁵ The optical field of the microcavity strongly couples with the excitons, forming two modes: the lower polariton (LP) and the upper polariton (UP). Exciton-polaritons with large transverse momentum (k_{ll}) created

- by nonresonant laser excitation, overcome the relaxation ³⁵ bottleneck resulted from the reduced density states of the small in-plane momentum LPs⁶ and condense into the ground state via polariton-phonon and polariton-polariton scattering (Figure 1a).³ Coherent light is subsequently achieved through radiative decay of the condensate polaritons.
- ⁴⁰ Unfortunately, the optically driven devices cannot be integrated into the current electronic circuits directly, which is disadvantageous for practical applications. Moreover, in a microcavity under electrical injection, due to the strong chargedipole interaction with electrons, the polaritons with large
- ⁴⁵ transverse momentum can efficiently pass their excess energy and wave vector to the injected electrons and overcome the relaxation

polaritons from their ground state by hot electrons,⁷ the Bose-Einstein condensation of the exciton-polaritons under electrical 55 injection is still beyond the reach until very recently, two independent groups reported the first electrically injected polariton laser in Nature¹⁴ and Physical Review Letters,¹⁵ where they presented a reservoir of cold electrons in the semiconductor layer that could be used to cool the hot electrons rapidly. а b exciton-polariton exciton-polariton cavity photor ⊖electron exciton energy cavity photon polariton-electron scattering pumping Е exciton energy



bottleneck more easily (Figure 1b).7 Therefore, developing an

electrically pumped polariton laser has attracted great scientific interest during the past couple of years. Polariton light emitting

50 diodes have been realized in various systems range from organic⁸

to inorganic semiconductors^{9, 10} even near the room

temperature.¹¹⁻¹³ However, because of the ionization of the

in (a) optically pumped microcavities, and (b) electrically pumped microcavities, respectively. The optical field of the microcavity strongly couples with the excitons, forming two modes: the lower polariton (LP)
65 and the upper polariton (UP). Under optical pumping, hot polaritons with large transverse momentum (k_{||}) could dissipate their excess energy and condense to the ground state of the LP branch via inefficient multiple phonon emission processes (dashed arrows in a) or via the polariton-polariton scattering (solid arrows in a) at a higher polariton density. While 70 under electrical injection, due to the strong charge-dipole interaction, the polariton-electron scattering process (solid arrows in b) would offer a more effective way to drive the exciton-polaritons to the bottom state even at very low polariton density. Therefore, the threshold for operating the polariton laser would be remarkablly decreased with the participation 75 of the injected electrons.

Condensation process

The two groups realized their electrically pumped polariton lasers in the similar p-i-n junction (Figure 2a) with InGaAs quantum wells in an intrinsic GaAs cavity sandwiched by gradually doped

- ⁵ distributed Bragg reflectors (DBRs). Two-dimensional electron gases were introduced into this cavity by the delta-doped layers included at every second interface, which would dissipate heat by lateral thermal diffusion over the entire sample as a reservoir of cold electrons.^{7, 15} Therefore, the hot electrons could relax quickly
- ¹⁰ avoiding the ionization of polaritons. The current-densitydependent energy-momentum dispersions of the system (Figure 2b-d) showed three different regimes, corresponding to the incoherent polariton emission (Figure 2b), polariton laser operation (Figure 2c) and cavity mediated laser operation (Figure
- ¹⁵ 2d) respectively, which demonstrated that the Bose-Einstein condensation of exciton-polaritons was achieved before the microcavity system switched to the weak coupling regime under electrical injection.



- Fig. 2 (a) Schematics of the exciton-polariton laser diode structure adapted by Höfling et al. Four integrated InGaAs quantum wells in an intrinsic GaAs one-wavelength-thick cavity were sandwiched by gradually doped distributed Bragg reflectors (DBRs) with a ring-shaped electrode on the top DBR. (b-d) Energy-momentum dispersions (k₀, in-25 plane wavevector) with falsecolour intensity profiles corresponding to different current densities. Below threshold, the polaritonic system was
- different current densities. Below threshold, the polaritonic system was characterized by a thermal distribution of particles (b). Polariton laser was observed at higher injection currents (c). At even higher pump rates, photonic lasing occured (d). Figures are adapted from ref. 14.

30 The bright future of the exciton-polaritons

In order to reveal the emission nature of their polariton laser, Höfling et al.¹⁴ utilized the magnetic-field-dependent circular polarization spectra to identify whether their system was in the strong coupling regime under different injection currents. Photons ³⁵ originating from polaritons exhibit a specific magnetic-fielddependent Zeeman splitting that has a clear correlation with the excitonic fraction of the polaritons,^{16, 17} whereas the cavity emission in the weak coupling regime is unaffected by a magnetic field. Unlike the common way based on the transition of energy-⁴⁰ momentum dispersions and the nonlinearity in input-output characteristics, the method not only provides a clear-cut criterion to distinguish between a polariton-mediated laser and a cavitymediated laser, but also helps to probe the emission character of the polariton system in real time, which will offer a powerful tool ⁴⁵ for the studies of dynamical Bose-Einstein condensation.

Due to its ultralow threshold, the exciton-polariton laser diodes prepared by the two groups are very promising to play the important role of coherent signal source in the nanophotonic circuits. Driven by the injection current, this light source could ⁵⁰ convert an electrical signal into a coherent optical signal, which is an ideal candidate for bridging the gap between the current microelectronic circuits and their nanophotonic counterparts. Moreover, compared with the photons, exciton-polaritons have anomalously low group velocity, leading to a substantially large ⁵⁵ refractive index of the media, which will enable them to be propagated and manipulated at subwavelength scale.^{18, 19} The polariton laser and other polariton-based components,²⁰ occupying very small volumes, open the way to the miniaturization of integrated photonic devices.

60 Conclusions

The electrically pumped polariton laser, a new generation of coherent light source, not only affords a model system for the fundamental understanding of strong-coupling cavity quantum electrodynamics but also promotes the development of 65 nanophotonics to a very great extent. Up to now, the reported electrically pumped exciton-polariton laser can only operate at cryogenic temperatures due to the small binding energy (~ 10 meV) of the extended Wannier-Mott excitons, which is unavailable for practical applications. Excitons with broader 70 spectral tunability and higher binding energy, such as the Frenkel type excitons originated from organic semiconductor materials,²¹ might be considered to participate in the lasing diode to extend the emission wavelength to the whole visible range, and to enable the devices to work at higher temperature, especially room 75 temperature. Combining the rational design of the device configuration with the wide selection of materials, the polaritonic devices seem to have a bright, colourful future ahead.

Acknowledgements

This work was supported by the National Natural Science ⁸⁰ Foundation of China (No. 21125315), the Chinese Academy of Sciences, and the Ministry of Science and Technology of China (2012YQ120060).

Notes and references

Beijing National Laboratory for Molecular Sciences (BNLMS), CAS Key 85 Laboratory of Photochemistry, Institute of Chemistry, Chinese Academy of Sciences, Beijing 100190, China. Fax: (+) 86-10-62652029; E-mail: yszhao@iccas.ac.cn

- C. Weisbuch, M. Nishioka, A. Ishikawa and Y. Arakawa, *Phys. Rev. Lett.*, 1992, 69, 3314-3317.
- H. Deng, G. Weihs, D. Snoke, J. Bloch and Y. Yamamoto, *Proc. Natl Acad. Sci. USA*, 2003, **100**, 15318-15323.
- 5 3. H. Deng, G. Weihs, C. Santori, J. Bloch and Y. Yamamoto, *Science*, 2002, **298**, 199-202.
- S. Christopoulos, G. B. H. von Högersthal, A. J. D. Grundy, P. G. Lagoudakis, A. V. Kavokin, J. J. Baumberg, G. Christmann, R. Butté, E. Feltin, J. F. Carlin and N. Grandjean, *Phys. Rev. Lett.*, 2007, 98, 126405.
- 5. S. Kéna-Cohen and S. R. Forrest, Nat. Photonics, 2010, 4, 371-375.
- A. I. Tartakovskii, M. Emam-Ismail, R. M. Stevenson, M. S. Skolnick, V. N. Astratov, D. M. Whittaker, J. J. Baumberg and J. S. Roberts, *Phys. Rev. B*, 2000, 62, R2283-R2286.
- 15 7. G. Malpuech, A. Kavokin, A. Di Carlo and J. J. Baumberg, *Phys. Rev. B*, 2002, **65**, 153310.
- J. R. Tischler, M. S. Bradley, V. Bulović, J. H. Song and A. Nurmikko, *Phys. Rev. Lett.*, 2005, 95, 036401.
- 9. D. Bajoni, E. Semenova, A. Lemaître, S. Bouchoule, E. Wertz, P. ²⁰ Senellart and J. Bloch, *Phys. Rev. B*, 2008, **77**, 113303.
- A. A. Khalifa, A. P. D. Love, D. N. Krizhanovskii, M. S. Skolnick and J. S. Roberts, *Appl. Phys. Lett.*, 2008, **92**, 061107.
- S. I. Tsintzos, N. T. Pelekanos, G. Konstantinidis, Z. Hatzopoulos and P. G. Savvidis, *Nature*, 2008, 453, 372-375.
- 25 12. S. I. Tsintzos, P. G. Savvidis, G. Deligeorgis, Z. Hatzopoulos and N. T. Pelekanos, *Appl. Phys. Lett.*, 2009, 94, 071109.
 - T. C. Lu, J. R. Chen, S. C. Lin, S. W. Huang, S. C. Wang and Y. Yamamoto, *Nano Lett.*, 2011, 11, 2791-2795.
 - 14. C. Schneider, A. Rahimi-Iman, N. Y. Kim, J. Fischer, I. G. Savenko,
- M. Amthor, M. Lermer, A. Wolf, L. Worschech, V. D. Kulakovskii, I. A. Shelykh, M. Kamp, S. Reitzenstein, A. Forchel, Y. Yamamoto and S. Höfling, *Nature*, 2013, 497, 348-352.
- P. Bhattacharya, B. Xiao, A. Das, S. Bhowmick and J. Heo, *Phys. Rev. Lett.*, 2013, **110**, 206403.
- ³⁵ 16. T. A. Fisher, A. M. Afshar, M. S. Skolnick, D. M. Whittaker and J. S. Roberts, *Phys. Rev. B*, 1996, **53**, R10469-R10472.
 - A. Armitage, T. A. Fisher, M. S. Skolnick, D. M. Whittaker, P. Kinsler and J. S. Roberts, *Phys. Rev. B*, 1997, **55**, 16395-16403.
- 18. K. Takazawa, J.-i. Inoue, K. Mitsuishi and T. Takamasu, *Phys. Rev. Lett.*, 2010, **105**, 067401.
 - C. Zhang, C.-L. Zou, Y. Yan, R. Hao, F.-W. Sun, Z.-F. Han, Y. S. Zhao and J. Yao, *J. Am. Chem. Soc.*, 2011, **133**, 7276-7279.
- B. Piccione, C.-H. Cho, L. K. van Vugt and R. Agarwal, Nat. Nanotechnol., 2012, 7, 640-645.
- 45 21. Y. Yan and Y. S. Zhao, Adv. Funct. Mater., 2012, 22, 1330-1332.