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6. AUTHORS
Vinod Menon

7. PERFORMING ORGANIZATION NAMES AND ADDRESSES
CUNY - City College of New York
160 Convent Avenue
New York, NY 10031 -9101

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14. ABSTRACT
Under the STIR program we developed solution processed ZnO nanoparticle based microcavity polaritons lasers. Polariton lasing action under optical pumping was demonstrated at threshold power levels of ~ 40 uJ/cm2. Ultrafast time resolved measurements were carried out that clearly show a sharp reduction in the emission lifetime above the threshold. Fourier space imaging of polariton emission is also reported that shows the appearance of two distinct angles at which emission maximum occurs due to the bottleneck effect seen often in polariton lasers. Finally, we present a design for electrically pumped version of the polariton laser

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Report Title

Final Report: Solution Processed ZnO Nanoparticle Based UV Lasers

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TOTAL:

Number of Papers published in non peer-reviewed journals:

(c) Presentations

Stimulated polariton emission from ZnO-nanoparticle based microcavity," X. Liu, K. Appavoo, M. Sfeir, S. Kena Cohen and V. M. Menon, CLEO 2014, San Jose, https://doi.org/10.1364/CLEO_SI.2014.STu2O.2

Number of Presentations: 1.00

Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

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Number of Non Peer-Reviewed Conference Proceeding publications (other than abstracts):

Peer-Reviewed Conference Proceeding publications (other than abstracts):

Received Paper

TOTAL:

Number of Peer-Reviewed Conference Proceeding publications (other than abstracts):

(d) Manuscripts

Received Paper

TOTAL:

Number of Manuscripts:

Books

Received Book

TOTAL:

Received Book Chapter

TOTAL:

Patents Submitted

Patents Awarded

Awards

None

Graduate Students

NAME	PERCENT_SUPPORTED	Discipline
Zheng Sun	0.50	
FTE Equivalent:	**0.50**	
Total Number:	**1**	

Names of Post Doctorates

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Names of Faculty Supported

NAME	PERCENT_SUPPORTED	National Academy Member
Vinod Menon	0.05	
FTE Equivalent:	**0.05**	
Total Number:	**1**	

Names of Under Graduate students supported

NAME	PERCENT_SUPPORTED
FTE Equivalent:	
Total Number:	

Student Metrics

This section only applies to graduating undergraduates supported by this agreement in this reporting period

The number of undergraduates funded by this agreement who graduated during this period: 0.00

The number of undergraduates funded by this agreement who graduated during this period with a degree in science, mathematics, engineering, or technology fields:...... 0.00

The number of undergraduates funded by your agreement who graduated during this period and will continue to pursue a graduate or Ph.D. degree in science, mathematics, engineering, or technology fields:...... 0.00

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Names of Personnel receiving masters degrees

NAME

Total Number:

Names of personnel receiving PHDs

NAME

Total Number:

Names of other research staff

NAME	PERCENT_SUPPORTED

FTE Equivalent:

Total Number:

Sub Contractors (DD882)

Inventions (DD882)

Scientific Progress

See Attachment

Technology Transfer

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Report Type:	Final Report
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Report Period End Date:	05/31/2016

Abstract:

Under the STIR program we developed solution processed ZnO nanoparticle based microcavity polaritons lasers. Polariton lasing action under optical pumping was demonstrated at threshold power levels of ~ 40 μJ/cm^2. Ultrafast time resolved measurements were carried out that clearly show a sharp reduction in the emission lifetime above the threshold. Fourier space imaging of polariton emission is also reported that shows the appearance of two distinct angles at which emission maximum occurs due to the bottleneck effect seen often in polariton lasers. Finally, we present a design for electrically pumped version of the polariton laser.

Introduction:

ZnO has been touted to be one of the most attractive candidates for solid state UV lasers. In fact there have been several demonstrations of lasers and light emitters based on a variety of growth techniques and device architectures such as nanowires, microresonators, microdisks, photonic crystal cavities, random lasing in ZnO powders and nanowires[1]–[5]. ZnO has also been considered as a strong candidate to realize polariton (half-light half-matter quasiparticles) lasers that can operate at room temperature[6], [7]. In fact there have already been few demonstrations for polariton formation and lasing using ZnO microcavities [8]–[11].

Currently, the growth of crystalline ZnO materials is usually carried out using molecular beam epitaxy, metal organic chemical vapor deposition or pulsed laser deposition. An alternate low-cost approach is solution processing using ZnO nanoparticles which has the following advantages: allows large area device fabrication, mechanical flexibility and most importantly ease of fabrication. Furthermore, these nanoparticles have been found to be defect free as observed from the lack of green defect related emission and robust to high optical pump powers. Recently the PI's group in collaboration in collaboration with the Sfier group at Brookhaven National Labs demonstrated low threshold random lasing in subwavelength thick ZnO films [12]. Through this STIR program, we developed a room temperature optically pumped ZnO microcavity polariton laser. Furthermore, we carried out Fourier space imaging and time resolved measurements to characterize the lasing.

The goals of the proposed work were:

1. Demonstrated optically pumped polariton lasing
2. Study the dynamics of polariton lasing in ZnO cavities
3. Provide a design guideline for electrically pumped laser

Optically pumped polariton laser:

The schematic of the ZnO nanoparticle based microcavity is shown in Fig. 1. The micriocavity structure consists of a half wavelength thick ZnO cavity layer sandwiched between two Si_3N_4/SiO_2 distributed Bragg reflectors (DBR) grown via plasma enhanced chemical vapor deposition (PECVD). The ZnO nanoparticle layer is deposited via spin coating on the bottom DBR and then placed on a hot plate at 350C for 20 minutes to remove the solvents. The ZnO nanoparticles were obtained from Sigma Aldrich. Following the deposition of the ZnO nanoparticles, the top DBR was completed via PECVD. Prior to the fabrication of the microcavity structure, the ZnO nanoparticle thin film was characterized optically as well as structurally. Shown in Fig. 2 is the dielectric constants of the ZnO nanoparticle film obtained via ellipsometry. The inset shows the scanning electron microscope image of the ZnO nanoparticle film. The obtained dielectric constants are slightly lower than that of bulk films due to the presence of voids in the nanoparticle film. The room temperature photoluminescence (PL) does not show the defect emission that is often seen in the green spectral range. The absorption measurements at room temperature does not clearly show the different excitonic absorption features.

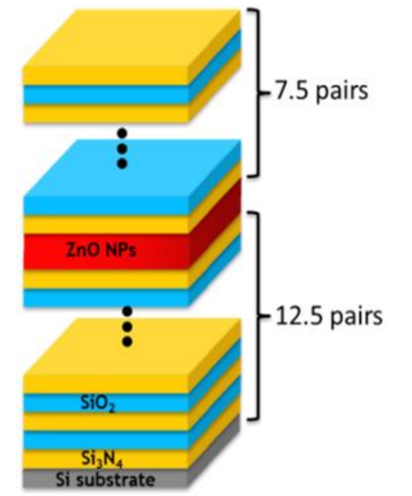

Fig. 1. Schematic of the ZnO microcavity

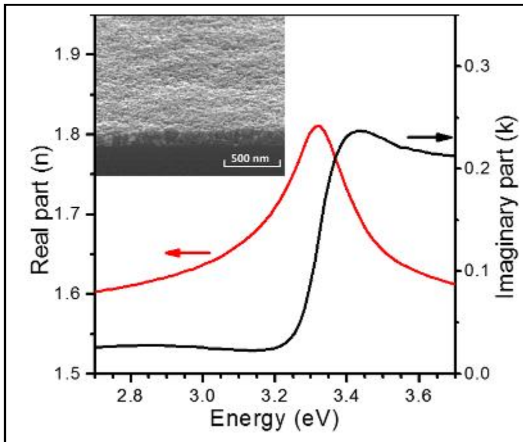

Fig. 2. Dielectric constants of the ZnO nanoparticle film obtained via ellipsometry. The inset shows the SEM image of a ZnO nanoparticle film.

Following the fabrication of the microcavity structure, we carried out pump power dependent PL experiments. Shown in Fig. 3 below is the emission spectra for various pump powers as well as the power –out and emission linewidth as a function of pump fluence.

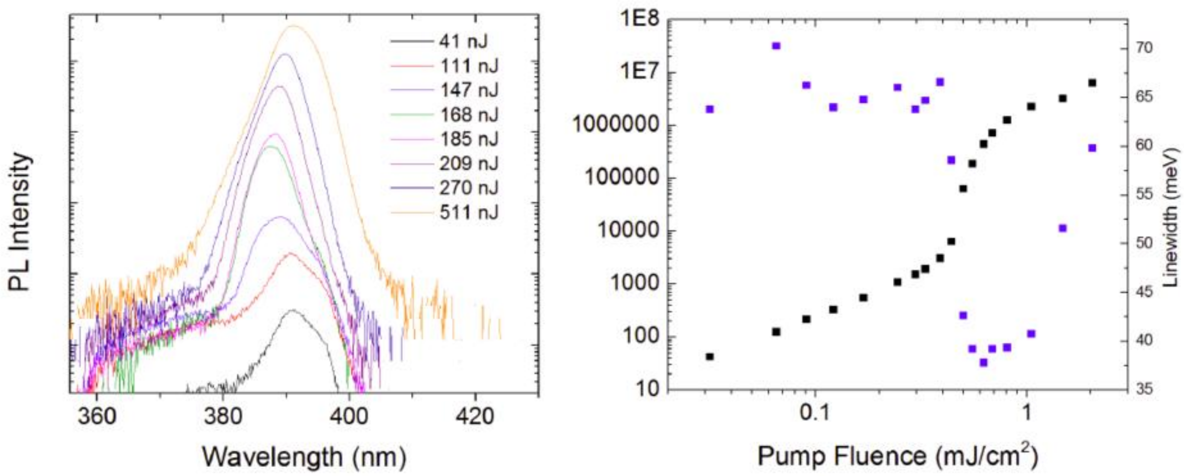

Fig. 3. (Left) Emission spectrum for different pump power levels. (Right) Power out (black cirles) and emission linewidth (blue circles) as a function of pump fluence clearly showing the lasing threshold

The emission spectrum is rather broad and consists of multiple modes. One of the intriguing aspects of the lasing demonstration here is the rather large linewidths seen. We hypothesize that we could be having cavity assisted polariton random lasing here. In addition to the pump power dependence on the emission properties, we also carried out angle resolved photoluminescence measurements using a Fourier space imaging set up. Shown in Fig. 4 is the result of the angle resolved measurements. Below threshold we observe almost uniform the emission from the lower polariton branch. However once above threshold power level, we see the emergence of two distinct emission maximums away from *k*=0. The emergence of these symmetric emission maxima on either side of *k*=0 at ~ 15 degrees is indicative of the process of polariton stimulated scattering bottleneck[13].

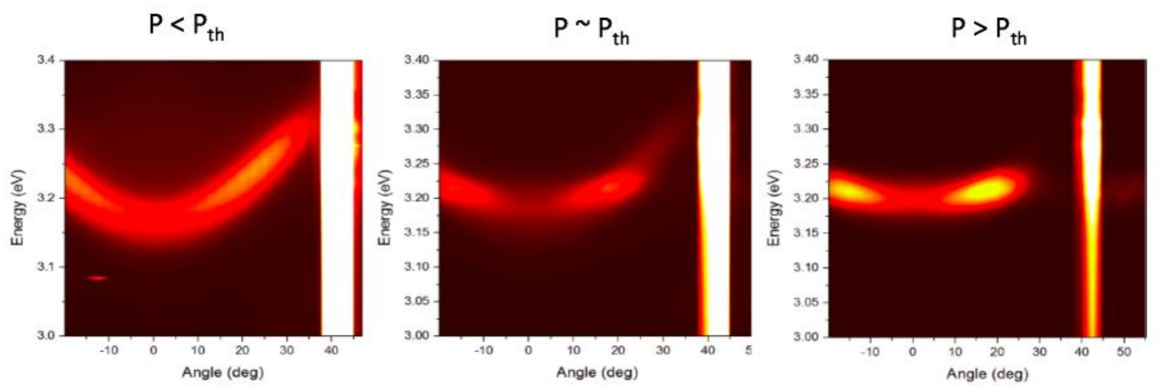

Fig. 4. Angle resolved PL from ZnO microcavity below, ay and above threshold pump power. Clearly above the threshold we see emission maximum at specific angles indicating bottleneck effect in stimulated polariton scattering as well as clear spectral narrowing and nonlinear increase in output intensity.

Finally we also carried out time resolved luminescence measurements using a Kerr gating set up at Brookhaven National labs[14]. An excitation pulse with wavelength of 280 nm, and 100 fs pulse duration was focused down to a 500 μm diameter spot size for the pump. The experiments were conducted with a Ti:Sapphire laser system (Spectra Physics) with the pump being generated using a commercial optical parametric amplifier (OPA, Light Conversion). A home built high sensitivity Kerr-gating set up was used for the transient PL experiments. Benzene was used as the Kerr medium in the low fluence limit with ~ 530 fs and quartz in the high fluence limit with a time resolution of ~220 fs. The gated emission signal was collected using a liquid nitrogen cooled CCD Camera (Symphony II) attached to a monochromator (Horiba Jobin Yvon – IHR 320).

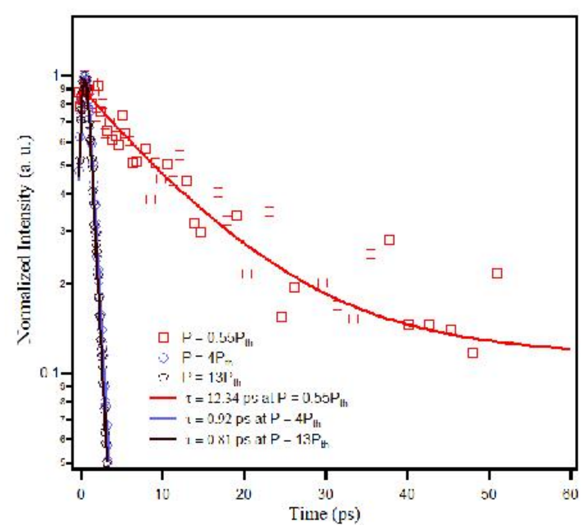

Fig. 5. Time resolved PL data on the ZnO microcavity below and above threshold sowing a clear reduction in lifetime.

The results of the time resolved measurements are shown in Fig. 5. Clearly above the threshold the emission dynamics shows sharp reduction in lifetime from 12.5

ps to 0.9 ps. Above the threshold the lifetime stays the same. In addition to just transient traces for emission, we also obtained the wavelength dependent transient emission spectrum as shown below in Fig. 6.

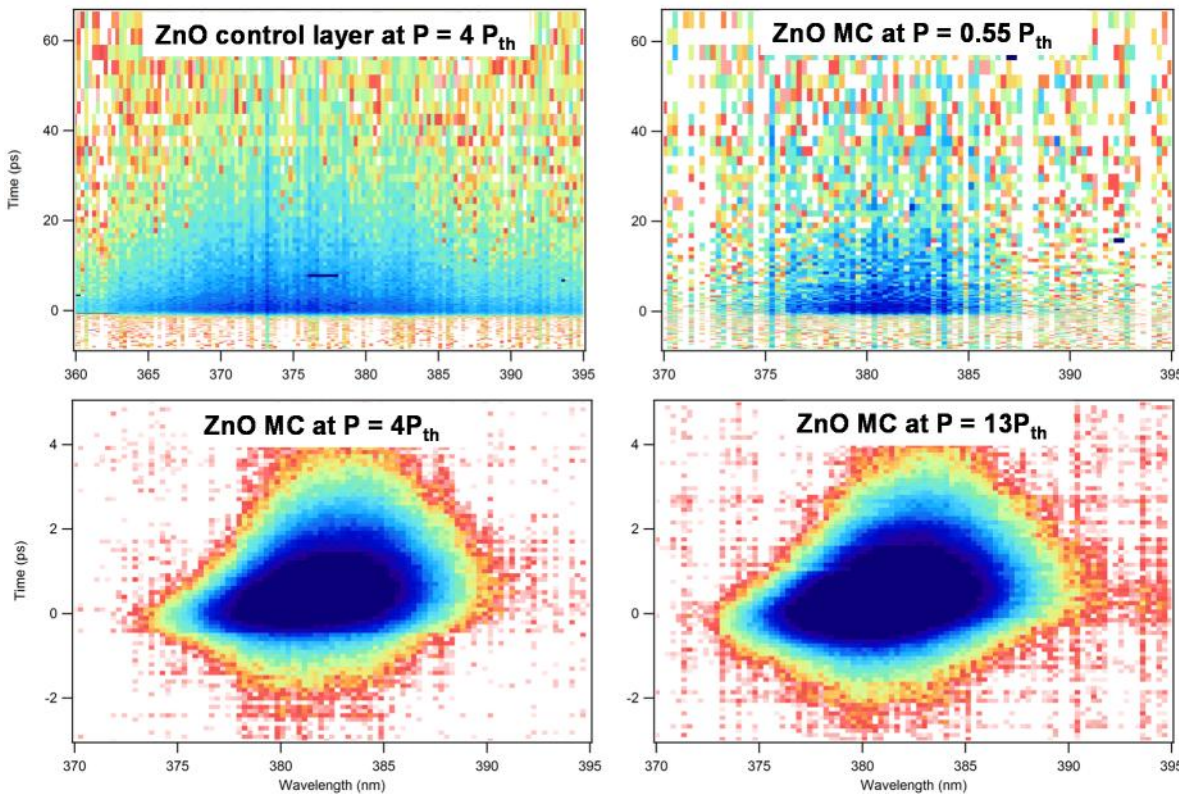

Fig. 6. Spectrally resolved transient PL measurements showing the nonlinear increase in emission intensity accompanied with abrupt spectral narrowing indicative of stimulated emission and gain.

Future plans:

Below we discuss briefly the design of an electrically pumped ZnO polariton laser. This design takes the cue from a previous demonstration of the first electrically pumped polariton laser[15]. Here a lateral geometry will be used as shown in the schematic below. One of the major hurdles to overcome is the realization of p-type ZnO nanoparticles. The ZnO nanoparticles generally tend to be n-type. Recently there has been some success on realized nitrogen doped ZnO which shows p-type conductivity[16]. If this can indeed be accomplished, then one should in principle be able to realize an electrically pumped polariton laser such as the one shown in Fig. 6. The fabrication of such a device will start with spin coating of ZnO nano particles (n-type followed by p-type) and then patterning the cavity layer. This will be followed by the realization of DBR first depositing SiO_2 and then using focused ion-beam to etch grooves in SiO_2 and sputter coating of TiO_2. During this process the ZnO layer will be protected using resist. Finally the top and bottom contacts will be patterned as shown.

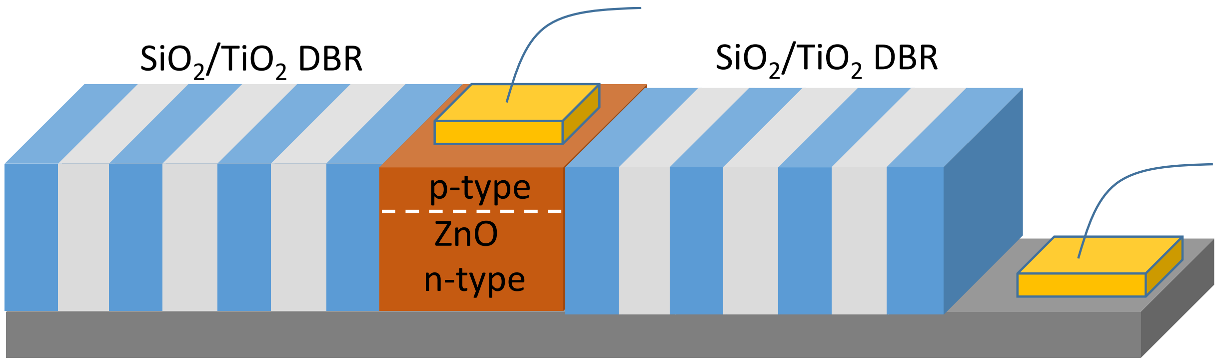

Fig. 6. Schematic of the proposed electrically pumped ZnO polariton laser with lateral DBRs.

Publication/Presentation

1. "Room temperature stimulated polariton scattering in ZnO microcavity," X. Liu, K. Appavoo, M. Sfeir, S. Kena-Cohen and V. M. Menon – under preparation
2. Stimulated polariton emission from ZnO-nanoparticle based microcavity," X. Liu, K. Appavoo, M. Sfeir, S. Kena Cohen and V. M. Menon, CLEO 2014, San Jose, https://doi.org/10.1364/CLEO_SI.2014.STu2O.2