REPORT DOCUMENTATION PAGE	Form Approved OMB NO. 0704-0188

1. REPORT DATE (DD-MM-YYYY) 20-04-2022	2. REPORT TYPE Final Report	3. DATES COVERED (From - To) 5-Jun-2018 - 31-May-2021

4. TITLE AND SUBTITLE Final Report: Quantum Optical Spectroscopy System for Materials in Extreme Environments	5a. CONTRACT NUMBER W911NF-18-1-0224
	5b. GRANT NUMBER
	5c. PROGRAM ELEMENT NUMBER 611103
6. AUTHORS	5d. PROJECT NUMBER
	5e. TASK NUMBER
	5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAMES AND ADDRESSES University of Pennsylvania Office of Research Services 3451 Walnut Street, 5th Floor Philadelphia, PA 19104 -6205	8. PERFORMING ORGANIZATION REPORT NUMBER
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS (ES) U.S. Army Research Office P.O. Box 12211 Research Triangle Park, NC 27709-2211	10. SPONSOR/MONITOR'S ACRONYM(S) ARO
	11. SPONSOR/MONITOR'S REPORT NUMBER(S) 72106-MS-RIP.1

12. DISTRIBUTION AVAILIBILITY STATEMENT

Approved for public release; distribution is unlimited.

13. SUPPLEMENTARY NOTES

The views, opinions and/or findings contained in this report are those of the author(s) and should not contrued as an official Department of the Army position, policy or decision, unless so designated by other documentation.

14. ABSTRACT

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:			17. LIMITATION OF ABSTRACT	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON
a. REPORT	b. ABSTRACT	c. THIS PAGE			Lee Bassett
UU	UU	UU	UU		19b. TELEPHONE NUMBER 215-573-7565

RPPR Final Report
as of 20-Apr-2022

Agency Code: 21XD

Proposal Number: 72106MSRIP **Agreement Number: W911NF-18-1-0224**

INVESTIGATOR(S):

Name: Ph.D Lee Bassett
Email: lbassett@seas.upenn.edu
Phone Number: 2155737565
Principal: Y

Organization: **University of Pennsylvania**
Address: Office of Research Services, Philadelphia, PA 191046205
Country: USA
DUNS Number: 042250712 EIN: 231352685
Report Date: 31-Aug-2021 Date Received: 20-Apr-2022
Final Report for Period Beginning 05-Jun-2018 and Ending 31-May-2021
Title: Quantum Optical Spectroscopy System for Materials in Extreme Environments
Begin Performance Period: 05-Jun-2018 **End Performance Period:** 31-May-2021
Report Term: 0-Other
Submitted By: Ph.D Lee Bassett Email: lbassett@seas.upenn.edu
Phone: (215) 573-7565
Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees: **STEM Participants:**

Major Goals: This project supported the acquisition of a unique optical spectroscopy system designed to probe quantum effects of individual defects in semiconductor nanomaterials under carefully controlled environments. The system specifically includes (1) a widely-tunable, continuous wave, narrow-linewidth, visible and near-infrared optical parametric oscillator for coherent photoexcitation spectroscopy and coherent quantum control along with (2) a closed-cycle optical cryostat providing an ultrahigh vacuum (UHV) sample environment, temperature control between 6 and 450 K, and a short optical working distance to facilitate diffraction-limited confocal fluorescence microscopy with sub-micron resolution.

The unique system acquired through this grant will support detailed investigations of the electronic and chemical structure of quantum defects in solid state materials in the absence of surface contamination, including pristine diamond surfaces, nanodiamond particles for quantum sensors, quantum emitters in hexagonal boron nitride, and other emerging host materials such as thin films, colloidal nanoparticles, and nanowires.

Accomplishments: Please see the attached PDF.

Training Opportunities: This project directly supported the acquisition of new instrumentation as described in Major Goals and Accomplishments. However, the project indirectly provided substantial opportunities for education, training, and professional development of individuals working in the PI's laboratory on the design, installation, testing, and use of the instrumentation. In particular, the UHV optical cryostat was custom designed by a team of graduate students, postdocs, and the PI over a period of many months, working with the engineers of the cryostat supplier (ARS) as well as with other companies to source the optical cell, vacuum pumps, vacuum gauges, automated mechanical stages, and optical components. Students in the PI's lab also became familiar with the operation, testing, and detailed alignment procedures of the CWAVE optical parametric oscillator, through in-person visits by service technicians and (during the pandemic) remote debugging session via zoom.

Results Dissemination: Nothing to Report

Honors and Awards: Nothing to Report

Protocol Activity Status:

Technology Transfer: Nothing to Report

Partners

,

I certify that the information in the report is complete and accurate:
Signature: Lee C Bassett
Signature Date: 4/20/22 11:43AM

Final Report
Army Research Office Agreement W911NF-18-1-0224
Funding period: 6/5/2018-5/31/2021

PI: Lee C. Bassett
Associate Professor of Electrical & Systems Engineering,
University of Pennsylvania, PA

This DURIP project supported the acquisition of a unique optical spectroscopy system designed to probe quantum effects of individual defects in semiconductor nanomaterials under carefully controlled environments. The system specifically includes (1) a widely-tunable, continuous wave, narrow-linewidth, visible and near-infrared optical parametric oscillator for coherent photoexcitation spectroscopy and coherent quantum control along with (2) a closed-cycle optical cryostat providing an ultrahigh vacuum (UHV) sample environment, temperature control between 6 and 450 K, and a short optical working distance to facilitate diffraction-limited confocal fluorescence microscopy with sub-micron resolution.

This report lists the instrumentation that was acquired with support of this grant, including special details regarding the design, construction, and operation of the customized system. It also describes the research projects on which this equipment has been or will be used, as described in the proposal and as will be of interest to the DoD.

List of equipment acquired

Description	Item Number	Vendor	Price	Purchase Date	Any special circumstances
RF Signal Generator, 25 MHz-12 GHz	SG6000LD	DS Instruments	$967.00	8/3/2018	
RF Amplifier, 600-4200 MHZ, 15W	ZHL-15W-422S+	Minicircuits	$2,295.00	8/3/2018	
Pulse generator, digital + analog	Pulse Streamer 8/2	Swabian Instruments	$4,768.50	10/19/2018	
Optical Components	Various	ThorLabs	$791.34	8/10/2018	
Power supply	A32HT900	Acopian	$540.00	8/24/2018	
Photon counting modules (2)	COUNT-100T-FC	Laser Components	$12,330.00	8/3/2018	

C-WAVE tunable laser	C-WAVE LP VIS IR	Cobolt/Huebner	$168,475.50	8/29/2018	
Fast steering mirror		Optics In Motion	$3,900.00	1/14/2019	
Closed cycle optical cryostat, final payment	CS210SF-GMX-20B-Custom	Advanced Research Systems	$49,105.66	4/27/2021	Final payment at time of delivery, delayed due to COVID
Closed cycle cryocooler, first installment	CS210SF-GMX-20B-Custom	Advanced Research Systems	$52,290.00	2/15/2019	Customized UHV optical cryostat, payment issued after design completed
Small optics and electronics	Various	Various	$4,133.00	various	
Total			$299,596.00		

Optical parametric oscillator system

The optical parametric oscillator (OPO) laser system acquired is the tunable, continuous-wave C-WAVE model from Hübner Photonics (Fig. 1). The C-WAVE uses OPO and second harmonic generation (SHG) technology to achieve tunable single-frequency output through automatic optimization of cavity temperature and length. The CWAVE can output single-frequency laser light from 450 - 650nm within 1 nm accuracy in the visible and 900-1300nm within 2nm in the infrared with a narrow spectral linewidth below 1MHz. The internal 1.5W pump laser provides power for up to >200mW output in the visible and >400mW in the IR. Once optimized for a specific wavelength, fine adjustments between 0.1-0.5nm can be achieved without the need for further optimization allowing for high-resolution photoluminescence excitation measurements.

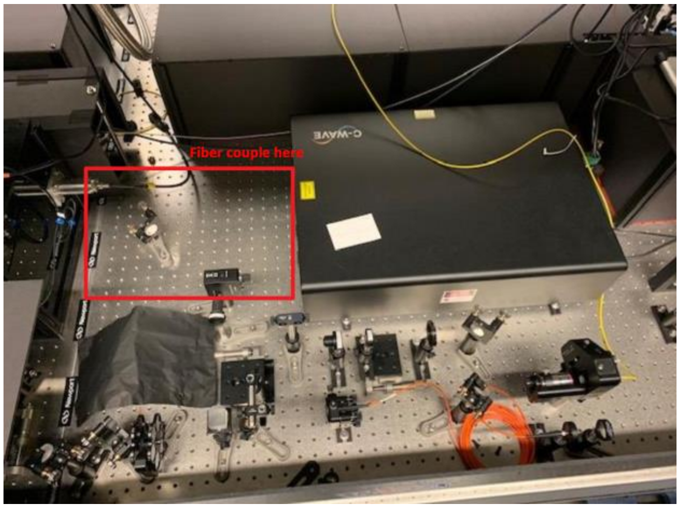

Figure 1. C-WAVE laser system on optics table in Bassett lab. The fiber-coupling optics used to route the emission to the optical cryostat on another table are temporarily removed for diagnostic experiments.

Design and operation of a customized UHV optical cryostat

Basic specifications

The customized UHV optical cryostat (Fig. 2) from Advanced Research Systems (ARS) is a closed-cycle helium cryostat able to reach temperatures between 6-450 K at the sample. The design of the sample chamber was modified to accommodate a load lock and cold finger that can protrude into a glass optical cell through which the sample will be imaged. The cryostat uses a He exchange gas interchange that minimizes vibrations to below 50 nm peak-to-peak, which is crucial to resolve and image diffraction-limited spots on the sample.

Ultrahigh vacuum operation

The cryostat design incorporates a load lock for sample loading to allow the UHV chamber to remain shielded from contaminants during operation. Sample loading involves mounting the sample in atmosphere and loading it into the UHV chamber through a series of gate valves. The sample is mounted vertically on an adjustable sample holder, which allows for positioning of the sample within 1mm of the window surface. A turbo station (Edwards) is used to pump down the load lock and UHV chamber to high vacuum (~10^{-6} torr). A gamma ion pump (Leybold) and a non evaporable getter (SAES) are then used to pump down the UHV chamber to UHV pressures (~10^{-10} torr). The getter assists with absorption of residual gases, such as hydrogen, which are less efficiently evacuated by the ion pump. A heating stage at the sample can reach temperatures of 450°C, allowing for *in situ* sample surface cleaning.

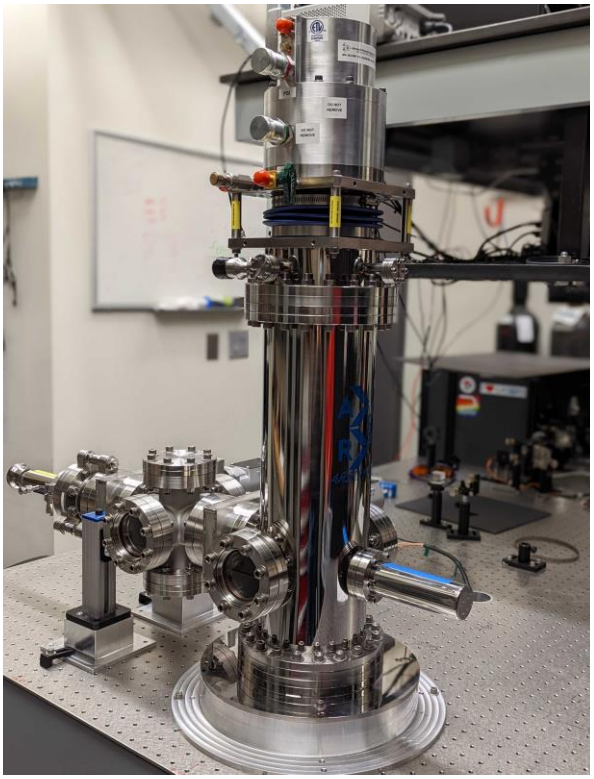

Figure 2. ARS cryostat on optical table in Bassett lab. The vertical tower is part of the UHV chamber, in which a cold plate is couple to the cold head using He exchange gas. The load lock extends to the left from the UHV chamber, and the glass optical cell (not pictured) will be attached in the place of the steel blank shown extending to the right from the UHV chamber.

Optical design

Samples are imaged through an all glass, hand blown, optical cell. The optical cell was custom made by Precision Glassmaking from quartz and UV grade silica. The cell has a -mm-thick fused silica window to allow for short-working distance, high NA confocal imaging with an external objective. The window is etched with a broadband, nano-textured antireflection coating. The cell's diameter of 1.5in with a 0.75in taper (Fig. 3) allows magnet access to the sample, making it possible to study magneto-optical effects and conduct spin resonance experiments.

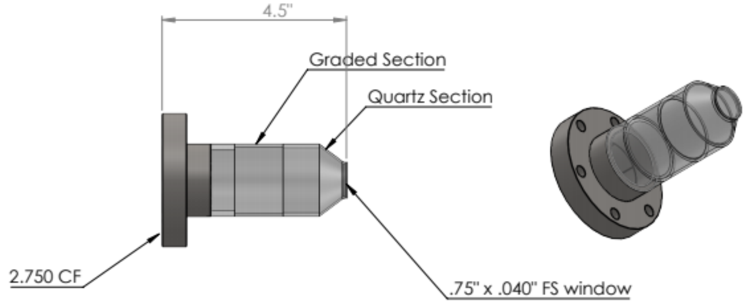

Figure 3. Schematic drawings of the custom glass optical cell.

Operation

In order to achieve ultra-high vacuum, the UHV chamber must be baked out, and kept free of contaminants. The bakeout procedure involves covering the cryostat in heating tape and heating the UHV chamber to ~200°C for >24 hours to allow for desorption of water from the steel. Once the chamber has been baked out, the load lock reduces exposure of the chamber to atmosphere during sample loading to decrease the need for repeated bakeouts and increase the chance of reaching UHV pressures. To use the load lock, the sample is mounted on the sample mount outside the cryostat, while the UHV chamber is pumped down to high vacuum through the load lock. A gate valve separating the load lock and UHV chamber is closed to isolate the UHV chamber, and the sample is loaded through into the load lock chamber at atmosphere. The load lock is then pumped back to HV and the sample is loaded into the UHV chamber, where the load lock rod is disconnected and removed. Finally, the UHV chamber is pumped down to UHV with the gamma ion pump and getter that are attached to the UHV chamber.

Optical system design

The confocal microscope is designed with a fixed sample and a moving objective outside the cryostat. The objective is mounted on a periscope (Fig. 4) consisting of three linear stages from PI (Physik Instrumente), which translate the excitation beam in x, y, and z onto the back of the objective as it moves. A fast-steering mirror from Optics in Motion modifies the angle of excitation incident on the back of the objective, allowing for acquisition of photoluminescence scans across tens of microns. To shield the excitation source's stability from vibrations of the cryostat, the C-WAVE OPO laser is kept on a separate optical table and will be fiber-coupled to the cryostat table using a high power, polarization maintaining single mode fiber. A noise eater and feedback loop will be used to monitor and control the power post-fiber and attenuate high frequency noise. A variable optical-density wheel and a wide band half-wave plate will provide power and polarization control respectively. Linearly variable dichroic and long pass filters can accommodate the wide excitation range of 450-650nm. The all glass optical cell contributes aberrations that require correction due to the window thickness, tilt, and deformation when under vacuum. An objective correction collar can compensate for the 1mm thick window, which causes the most significant aberrations. However, to further correct for the window's tilt and deformation, in the future, we may incorporate adaptive optics through a deformable mirror and feedback loop.

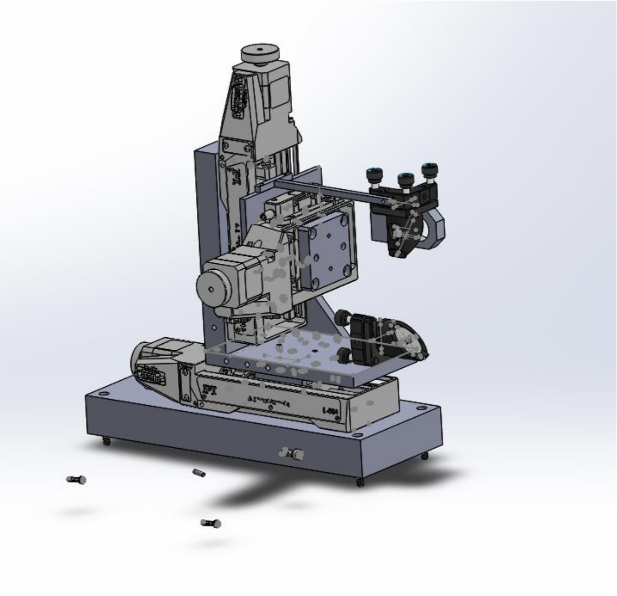

Figure 4. Design for periscope consisting of two elliptical mirrors and an objective (not pictured) mounted on x, y and z stages respectively. Beam is incident to the lower elliptical mirror from the right and is translated onto the back of the objective.

Conclusion

We are grateful to the ARO for supporting the acquisition of this unique research instrumentation in our laboratory. We look forward to utilizing this system to probe new physical phenomena associated with quantum defects and to explore the crucial role of surface chemistry on their performance as quantum sensors, single-photon sources, and nanoscale quantum processors. In addition to the impact on emerging quantum technologies, the research capabilities enabled by this grant will increase our foundational understanding of materials, chemistry, and quantum-mechanical dynamics at the smallest scales.