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RPPR Final Report
as of 27-Aug-2019

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Final Report for Period Beginning 05-Jun-2018 and Ending 31-May-2019

Title: A Cryogenic Magneto-Optical System for Probing Correlated Physics in Covalent Organic Frameworks

Begin Performance Period: 05-Jun-2018 | **End Performance Period:** 31-May-2019

Report Term: 0-Other

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Major Goals: The ARO DURIP funding is to establish a low temperature magneto-optical spectroscopy apparatus for probing novel phenomena in two-dimensional covalent organic framework materials and other two-dimensional heterostructures.

Accomplishments: Using the ARO DURIP funding, we purchased a magneto-optical cryostat (OptiCool) from Quantum Design and have integrated it with existing lasers and optical equipment. Initial characterization of the cryostat indicates that it will be an excellent system for conducting optical and electrical measurements of novel materials under large magnetic fields and at low temperatures.

Training Opportunities: Nothing to Report

Results Dissemination: Nothing to Report

Honors and Awards: Nothing to Report

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Technology Transfer: Nothing to Report

PARTICIPANTS:

Participant Type: PD/PI
Participant: feng wang
Person Months Worked: 1.00 **Funding Support:**
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Other Collaborators:

RPPR Final Report
as of 27-Aug-2019

Using the ARO DURIP funding, we purchased a magneto-optical cryostat (OptiCool) from Quantum Design and have integrated it with existing lasers and optical equipment. Initial characterization of the cryostat indicates that it will be an excellent system for conducting optical and electrical measurements of novel materials under large magnetic fields and at low temperatures. With this equipment, we will conduct optical experiments towards understanding correlations and magnetism in two-dimensional (2D) covalent organic frameworks (COFs), transition metal dichalcogenides (TMDCs), and heterostructures of both material systems.

The closed-loop OptiCool system (Fig. 1a) creates an environment for low-noise optical and electrical measurements under large magnetic fields and at low temperatures, which is ideal for studying magnetic and quantum properties of novel two-dimensional materials. The OptiCool system's superconducting magnet offers magnetic fields of +/- 7 Tesla perpendicular to the optical table. The field uniformity of +/- 0.03% over a 1 cm spherical volume allows for reliable measurements of both large and small samples. Furthermore, the excellent vibrational stability of <10 nm peak-to-peak horizontal noise and < 4 nm peak-to-peak vertical noise allows us to study small samples that are common in the initial stages of materials development. The sample temperature can be as low as 1.5 K, which is achieved through a closed-loop helium design that supports high-quality, continuous measurements without the use of additional helium.

A unique and particularly exciting property of this system is the convenient optical access through eight top and side windows. Typically, magneto-cryostats require that the sample is far away from the optics to reduce vibration and ensure thermal and magnetic stability, leading to inflexible and cumbersome methods for coupling light to the sample. Instead, the OptiCool has eight convenient ports for direct and flexible optical access, which has allowed us to integrate our lab's diverse optical tools with low-temperature and high-magnetic-field experiments (Fig. 1b). This unique combination of convenient optical access, large magnetic fields, and low temperatures is perfect for studying correlated physics in novel low-dimensional materials.

a **b**

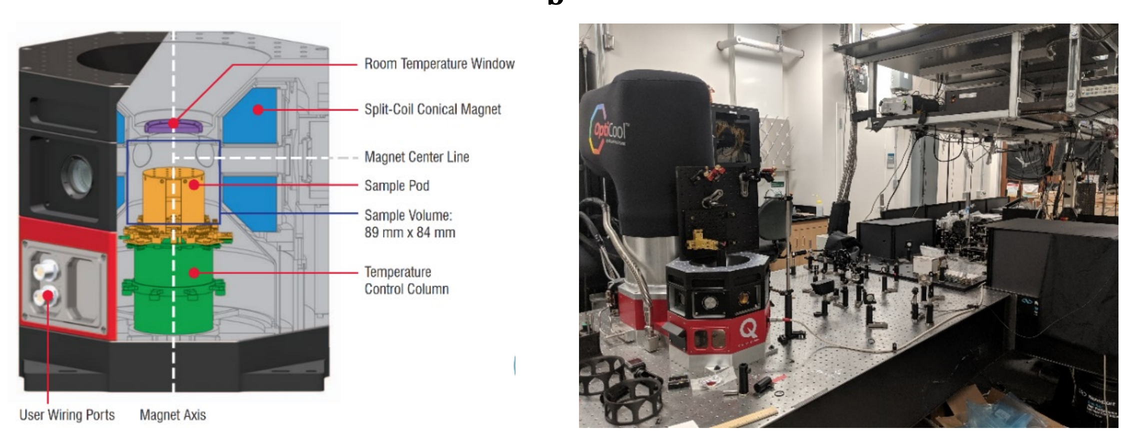

Figure 1. (**a**) Schematic of the OptiCool cryostat showing the sample pod, superconducting magnet, and several optical ports for easy integration with existing optical tools. Image from Quantum Designs. (**b**) Picture of the OptiCool system installed in our lab and integrated with optical equipment.

Several graduate students and postdoctoral scholars have installed the system and integrated it with existing light sources, including a helium-neon laser, a supercontinuum laser, a broadband lamp, and an ultrafast Ti-Sapphire-pumped optical parametric oscillator. With this combination of light sources and other optical components, we can perform broadband polarization-resolved measurements, including the magneto-optical Kerr effect (MOKE), and a diverse set of ultrafast measurements to probe correlations and magnetism in 2D COFs and TMDCs. We have performed initial characterization of the OptiCool system using a TMDC heterostructure of $MoSe_2$ and WSe_2 to test vibrational stability and optical quality using all non-magnetic optical components. The optical absorption at 1.5 K is shown in figure 2.

Our future plans for the OptiCool include demonstrating flat-band magnetism in 2D COF materials through optical investigations of hole-doped COF-1000 layers. We will use acceptor-type organic molecules, such as F4-TCNQ or $Ni(tfd)_2$ to introduce holes to the valance band. We will measure the MOKE signal as a function of the temperature to determine the Curie transition temperature. Then we will monitor the MOKE signal as a function of the applied magnetic field and measure the magnetic hysteresis loop of the 2D COF material. This study, if successful, will provide the first experimental observation of the flat band ferromagnetism, which was predicted in 1991 and was the first rigorous example of ferromagnetism arising purely from electron-electron interactions.

Following the demonstration of flat-band magnetism, we will study how to engineer and control magnetism in different ways, such as by tuning the carrier concentration. We will systematically study the change of Curie temperature and magnetic hysteresis loop in COF-1000 as a function of carrier concentration. The critical hole density beyond which the ferromagnetism starts to emerge corresponds to a quantum critical point that separates the non-magnetic and magnetic phase space in the COF material. This study will enable electrically switchable 2D magnetism, which will offer exciting opportunities for both fundamental studies and device applications. These measurements are enabled by the OptiCool cryostat and make excellent use of the unique properties of the system.

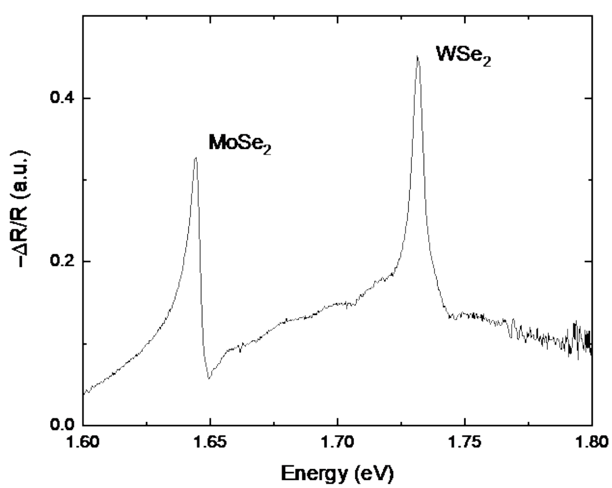

Figure 2. Optical absorption spectrum of a $MoSe_2/WSe_2$ heterostructure measured at 1.5 K in the OptiCool cryostat, indicating that we can obtain high-quality optical data of small (~ 5 um) samples while tuning temperature, magnetic field, and electrostatic gating.