NSF FINAL REPORT
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Collaborative Research: Optically Created Metastable Mesoscopic Nuclear Spin States: Glassy Transitions and Properties Beyond Electron Decoherence in Quantum Dots

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This program has been aimed at furthering our understanding coherent optical control of electron spins in solid state structures for application to quantum computing and quantum devices. The bulk of the work has been done on InGaAs quantum dots provided by our collaborators at NRL. Our experimental work was supported extensively by my long time collaborator Prof. L-J Sham at the University of California at San Diego. However, we recently began studies on transition metal dichalcogenides. In their intrinsic two dimensional form, the coherent nonlinear optical response is dominated by exciton-exciton interactions. Prof. Mackillo Kira and his group are supporting this work with theoretical modeling. This is the final report on this work as the program ended on 7.31.2021.

1. Single Photon Raman Scattering Measurements of Mesoscopic Metastable Nuclear States in InGaAs Quantum Dots

Over the course of this program, we have detailed our progress on understanding the complexities of the mesoscopic metastable nuclear spin states we first discovered some years ago on an earlier program. The main results of this work are now published Physical Review B, **102**, 235425 (2020). The paper shows the clearly two distinct mesoscopic nuclear spin configurations. The work sets the stage now for using a microwave pulse or possibly optical control to create a coherent state between the two spin states that might ultimately serve as a long lived nuclear spin qubit. Both nuclear spin states are characterized by reduced nuclear spin fluctuations that has resulted in long lived spin coherence, as we reported earlier. We also found that the nuclear spin states lived longer than 1 second in the dark, which is encouraging in terms of a potential mesoscopic nuclear spin qubit.

2. Experiments on Transition Metal Dichalcogenides

In the last report, we detailed our first studies of coherent nonlinear optical response of this system. This response is key to the use of these materials for some types of quantum devices. In this period, we have now been able to redo all the measurements on a higher quality material. We initially used a simple extension of the master equations to fit the data, but have found that a more complete theory is required. We are collaborating with Prof. Mackillo Kira at Michigan. His initial work on this problem with Stephen Koch in narrow bandgap III-V materials has defined the field.

Figure 1 shows the narrow band photoluminescence of h-BN encapsulated $MoSe_2$ photoluminescence. The primary peak is arises from the exciton and the smaller peak is from the trion. The exciton peak is fit reasonably well with a Lorentzian function, and the resulting linewidth is about 3 meV. The high quality of the sample is clearly illustrated by the reduced inhomogeneous broadening of the exciton peak, and the relatively weaker trion peak indicating low doping levels. Both of these features are expected and result from encapsulating the MoSe2 monolayer with h-BN.

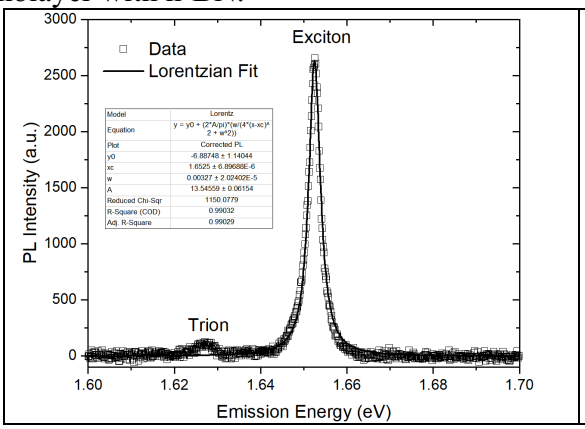

Figure 1: Photoluminescence spectrum of the h-BN encapsulated $MoSe_2$ monolayer at 4 K. The excitation was a linearly polarized HeNe laser with an energy of 1.96 eV. The PL spectrum of the MoSe2 monolayer shows two peaks: a trion peak at about 1.628 eV and an exciton peak at about 1.653 eV. The exciton peak was fit with a Lorentzian function and the resulting linewidth is about 3.3 meV. A relatively low trion peak and narrow linewidth are both expected results from the encapsulation of the MoSe2 monolayer with h-BN.

Figure 2 shows the nonlinear optical spectrum measured in reflection and transmission and the resulting nonlinear absorption profile. In the simplest picture, first developed by our group in collaboration with Stephan Koch, U. Marburg (Phys. Rev. Lett. **71**, 1261-1264 (1993)) shows that exciton-exciton interactions are the origin of the nonlinear optical response. This is quite distinct from the behavior in two-level systems where nonlinear optical response is dominated by competition between absorption and stimulated emission on long time scales and Rabi behavior on short time scales (compared to the decoherence time). The exciton-exciton interactions lead to a broadening (excitation induced dephasing) and a frequency shift (excitation induced shift). The relative contribution depends on the details of the interaction.

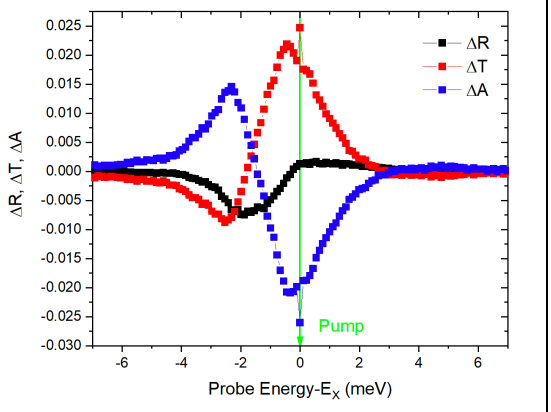

Figure 2: Typical absolute differential reflection, transmission, and absorption spectra of the h-BN encapsulated MoSe2 monolayers. The absolute differential spectra were obtained by characterizing the optical losses in the measurement apparatus to accurately estimate the amount of probe power reflected from and transmitted through the sample relative to the incident probe power. The pump and probe lasers were co-circ e7890ularly polarized.

The data in Fig. 2 shows that in this case in $MoSe_2$, the excitation induced shift appears to dominate the imaginary part of the nonlinear optical susceptibility, reflected in the differential absorption spectrum which shows evidence of a red shift. Given that the shift was red, we

confirmed that this shift was due to intrinsic physics and not simple thermal effects by showing the response time of the basic structure was much faster than thermal relaxation times.

Figure 2 also shows a single data point in the reflection and transmission spectrum which is real but not resolved because of the step size in the digitization of the data. However, Fig. 3 shows the data, reflected in the absorption spectrum, at much higher resolution. The width is exceeding narrow and is the result of was originally terms by Willis Lamb and population pulsations. These are oscillations of the exciton population driven at the difference frequency between the pump laser and the probe laser. The width reports on the electron-hole recombination time. We note that Fig 3 shows an additional narrow feature. This feature is the result of a small leakage of pump light in the direction of the probe beam which mixes with the probe beam in the detector. The effect is frequently used to measure the response time of detectors.

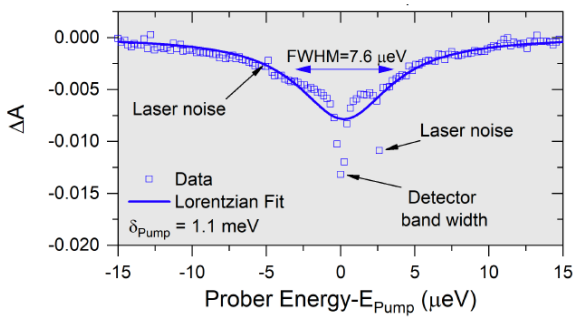

Figure 3. High resolution differential absorption spectra showing the population pulsation resonances that appear at zero pump-probe detuning in the blue curve in Fig. 2q1. The sub-μeV features at zero pump-probe detuning on the high resolution spectra correspond to the electrical bandwidth of the photodetector and is the result of mixing of the optical fields in the detector. The additional sub-μeV features on the high resolution spectra correspond to noise coming from the laser electronics. The pump and probe lasers were co-circularly polarized.

Students: Two students were supported at different times on this grant have since defended their work and were awarded the PhD. in physics and applied physics, respectively.