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Polarization- and time-resolved spectroscopy of dipole-forbidden excitons

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In this talk I will present a novel experimental technique for time-resolved spectroscopy of dipole-forbidden excitons called difference-frequency generation with two-photon excitation (2P-DFG). In this method, pump laser pulses create a coherent exciton population via two-photon excitation, while probe laser pulses stimulate the emission of photons with the energy difference between the excitons and the stimulating photons. By varying the time delay between the pump and probe pulses, the 2P-DFG signal is recorded as a function of time, enabling direct measurements of excitonic coherence times. By tuning the probe photon energy, the DFG signal can be spectrally separated from both lasers and photoluminescence emissions. Additionally, as the crystal is transparent to all four photons involved in the 2P-DFG process, the signal primarily emerges in the bulk, minimizing the influence of surface irregularities.

Using this approach, we directly measure the coherence times of dipole-forbidden *S*- and *D*-type Rydberg excitons in cuprous oxide (Cu_2O) with principal quantum numbers up to $n = 4$. The coherence times are further investigated across temperatures ranging from 1.4 K to 38 K and average excitation powers between 3 and 30 mW. While the 1S exciton exhibits a coherence time of approximately 3 nanoseconds, the coherence times of Rydberg states are about 3 orders of magnitude lower.

This method is further enhanced by polarization tomography for state-selective control in both pump and probe channels, offering advantages beyond previous methods. Characteristic polarization selection rules for this process, derived through group-theoretical analysis, are experimentally confirmed by measuring the complete polarization dependence of the 2P-DFG signal. Carefully chosen linear polarization settings enable the selective probing of specific 1*S* exciton eigenstates in a magnetic field while suppressing signals from others. This approach allowed us to measure magnetic-field-induced quantum beats across different regimes, including the exponential decay of a single $M = 0$ state, as well as two- and three-level quantum beats involving the $M = -1$ and $M = +1$ states. These results establish 2P-DFG as a powerful and versatile method for probing the coherence properties of dipole-forbidden excitons in semiconductor systems.