

Three-Pulse Echo Peak Shift Spectroscopy of Disordered Semiconductor Quantum Wells and Dense Atomic Vapors

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Abstract. Three-pulse echo peak shift spectroscopy yields the correlation function of the frequency fluctuations due to acoustic phonons for excitons in disordered semiconductor quantum wells and fluctuations due to atomic motion in a potassium vapor.

The dephasing of optical transitions is often treated in the Markovian approximation, where the dephasing events are assumed to be infinitely fast and there is no memory, i.e., the phase after the event does not depend on what it was prior to the event. Physically, dephasing occurs because the transition frequency fluctuates during a dephasing event. The Markovian approximation means that the two-time correlation function of the frequency fluctuations is a delta function. Given sufficiently high time resolution, the Markovian approximation is not valid, resulting in non-exponential temporal dynamics. In the non-Markovian regime, the dephasing cannot be described by a simple rate, but rather the correlation function must be determined [1]. Three-pulse echo peak shift (3PEPS) spectroscopy is capable of measuring the correlation function [2,3]. We apply 3PEPS to two systems that display non-Markovian dynamics, a disordered semiconductor quantum well [4] and a dense atomic vapor [5].

In 3PEPS spectroscopy, a 3-pulse transient four-wave-mixing (FWM), also known as photon echo, experiment is performed. The incident pulses are approximately 100 fs in duration and have nJ energy. The signal is recorded as the delay between the first two pulses, τ , is scanned. Typically the signal is recorded in two directions, $\mathbf{k}_+ = -\mathbf{k}_a + \mathbf{k}_b + \mathbf{k}_c$ and $\mathbf{k}_+ = \mathbf{k}_a - \mathbf{k}_b + \mathbf{k}_c$ where τ is defined to be positive for k_a arriving first and negative for k_b arriving first. As these scans are symmetric about $\tau = 0$, recording both reduces error in determination of $\tau = 0$. These scans are performed as the delay between the second and third pulses, T , is varied. The echo peak shift is the τ for which the signal is the maximum. The echo peak shift as a function of T maps out the correlation function for the frequency fluctuations.

Figure 1 shows typical FWM signals from the heavy-hole exciton resonance in a disordered semiconductor quantum well and the EPS for a series of temperatures and excitation densities. The disorder localizes excitons and produces inhomogeneous broadening. Two main processes lead to decoherence, migration of excitons between localization sites and the interaction of excitons with acoustic phonons. The EPS display a sharp decrease followed by a slow increase. After ~ 4 ps, there is an exponential decay to an offset of ~ 100 fs. We attribute the exponential decay to migration of the excitons between localization sites, which results in spectral diffusion (SD). Perhaps the most intriguing feature of the EPS is the increase during the first few picoseconds. This increase cannot be explained by a SD model. To

model the non-Markovian dynamics at short times, we use an underdamped oscillator model [1] in which acoustic phonons modulate the exciton energy. This model does give an increasing EPS.

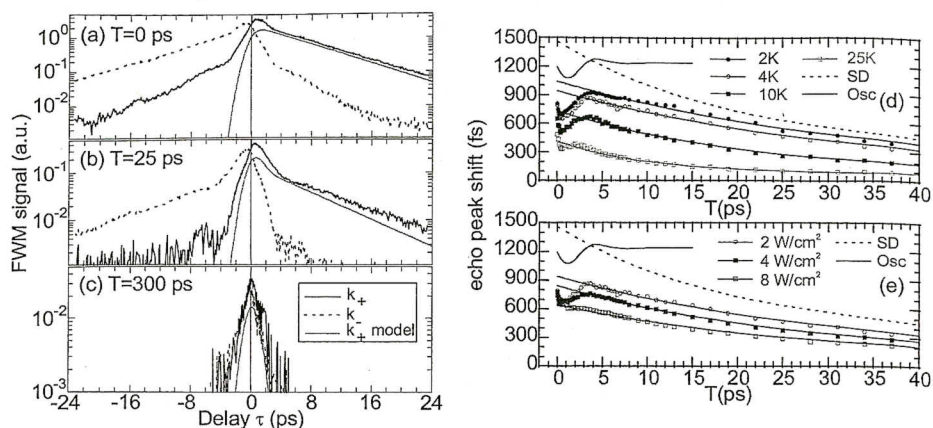


Fig. 1. Time-integrated FWM signals from a disordered GaAs quantum well as a function of τ for $T =$ (a) 0, (b) 25, and (c) 300 ps (logarithmic vertical scale). The solid (dashed) lines represent the k_+ (k_-) direction. The solid gray lines represent the calculated k_+ signal. The vertical lines at the center are at $\tau = 0$. The sample temperature is 2 K. (d) Echo peak shift as a function of T for a series of temperatures (2, 4, 10, and 25 K) at an excitation intensity of 2 W/cm². (e) EPS as a function of T for a series of excitation intensities (2, 4, and 8 W/cm²) at a temperature of 4 K. The markers represent experimental points and the lines represent exponential fits to the data for $T > \sim 5$ ps. The dashed black and solid gray lines represent the calculated EPS from the spectral diffusion (SD) model and the underdamped oscillator (Osc) model, respectively.

Figure 2 shows the transient FWM signal from a dense potassium vapor and the EPS for a series of temperatures. At these temperatures and densities, Doppler broadening is negligible and decoherence is due to resonance broadening, i.e., “collisions” between like atoms. Previous studies have shown that the signatures of non-Markovian dynamics are evident in the FWM signal as non-exponential behavior at short times and that the experimental results are well described by assuming an exponentially decaying correlation function [6]. 3PEPS removes the need to assume a correlation function. An atomic vapor is an ideal system for studying non-Markovian dynamics because there is a good separation of timescales, with the pulse being much shorter than the “collision duration” which in turn is much shorter than the decoherence rate.

These experimental results are in qualitative agreement with an exciton model combined with stochastic frequency fluctuations obtained from molecular dynamics simulations. The measured EPS from the vapor shows a single exponential decay at low temperatures (and densities) while at higher temperatures it become biexponential (Fig. 2(e)). The molecular dynamics simulations provide snapshots of the configuration of 20 potassium atoms. For each snapshot, the resonant couplings are determined from the potential energy surfaces for the potassium dimer. The couplings, and hence the eigenenergies, fluctuate as the atoms move. From the energy fluctuations, a correlation function and EPS can be determined (Fig. 2(f)). The calculations also show a biexponential behavior. By artificially turning off the long

range part of the potential, we can show that the slower component is due to long range dipole-dipole interactions [5].

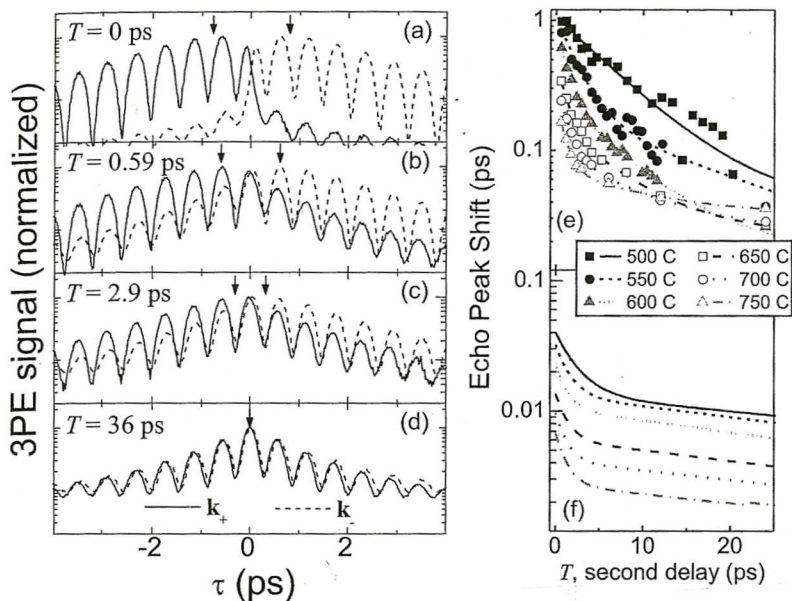


Fig. 2. (a-d) Experimental 3PE signals from a dense potassium vapor at 600 C versus the delay between the first two pulses, τ , for 4 values of the delay between the second and third pulse, T . The beats are due to simultaneous excitation of the D_1 and D_2 lines. The arrows indicate the peak of the signal without beats. (e) Experimental peak shifts for a series of temperatures and densities. (f) Calculated peak shifts from molecular dynamics simulations in an exciton picture.

3PEPS spectroscopy is a powerful tool to extract information about decoherence processes in a variety of systems. Here we have used it to study the decoherence of excitons in disordered quantum wells and in a dense atomic vapor.

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