Coherently Coupled Optical Stark Shifts Provide Evidence For Intervalence Band Coherences in Semiconductor Quantum Wells

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The optical Stark shift in semiconductors is an important transient nonlinearity that helps researchers to understand fundamental ultrafast microscopic processes. Studies of similarities and differences between optical Stark shifts in atomic two-level systems and those observed in semiconductors led to the understanding of semiconductor-specific many-body effects. We recently investigated the semiconductor counterparts of coherent atomic three-level effects utilizing the coherent coupling between heavy-hole (hh) and light-hole (lh) excitons. By analyzing the coherently coupled optical Stark effect on hh and lh exciton resonances we showed[1] that our data provide evidence for the existence of hh-lh Raman coherences in semiconductor quantum wells. In atomic three-level systems, Raman coherences are the foundation of important nonlinear optical effects such as electromagnetically induced transparency and lasing without inversion.

Figures 1(a) and 1(b) show the transient absorption changes measured at both hh and lh exciton resonances in an InGaAs quantum-well structure. The two degenerate three-band systems are effectively reduced to a single three-band system by driving a single hh transition with a circularly polarized pump and probing with either co- or counter-circular polarization. For co-circular polarization we observed the well-known transient blueshift of the hh exciton resonance. We did not observe any nonlinear response at the lh exciton resonance indicating negligible direct coupling between the pump field and the lh exciton. With a counter-circularly polarized probe pulse a definite blueshift appears at the coherently coupled lh exciton resonance.

Our theoretical analysis is based on a full many-body approach within the third-order nonlinear optical regime.[2] We considered a semiconductor quantum-well analog for the atomic three-level system, that is, the three-band system that consists of a conduction band as well as hh and lh valence bands. While in the band picture the analogy holds for each in-plane momentum state, the Coulomb interaction prevents the analogy from being completely accurate. Exciton scattering and biexcitonic effects have no counterparts in three-level systems. Nevertheless, as long as those processes do not dominate the Stark shift, an approximate three-level versus semiconductor Stark shift analogy is possible, and the definition of an excitonic Raman coherence (excitonic intervalence band coherence) is appropriate.

Figure 1(c) shows theoretical results for the ratio of hh-lh Stark shifts with and without intervalence band coherences. When these coherences are included, the lh shift is much larger than without coherences and the hh-lh shift ratio is approximately two. Deviations by a factor of 2 come from the hh and lh mass differences, especially at small detunings, from correlation contributions. At large detunings the ratio behaves essentially like a three-level system: it is two (infinity) for the case with (without) Raman coherences. Clearly, the experimental observations (squares) provide direct evidence for the presence of an intervalence band Raman coherence, because the observed hh-lh shift ratio is close to the ideal value of two at large detunings.

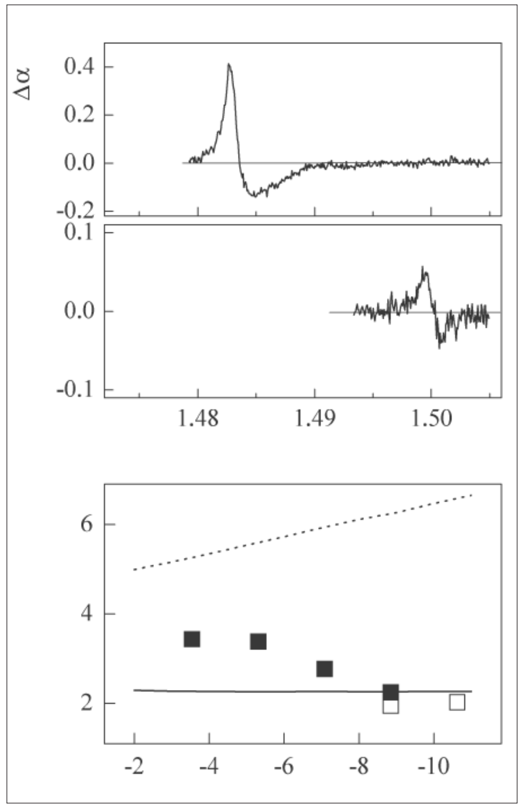

Figure 1. (a) Differential absorption spectrum of InGaAs quantum wells (T = 4 K) measured at maximum temporal pump-probe overlap with co-circularly polarized pump and probe pulses. The 1.2-ps pump pulse had an intensity of 21 MW/cm^2 and was detuned at 8.8 meV below the linear hh exciton resonance. The weak, broadband probe pulses were approximately 100 fs long. (b) Same as (a) counter-circular polarization of pump and probe pulse. (c) Ratio of the hh exciton Stark shift over the coherently coupled lh exciton Stark shift versus detuning between pump pulse and hh exciton energy. Solid and dotted lines show the theoretical results with and without intervalence band coherences, respectively. The squares correspond to the experimental values for 3.7-W/cm^2 (solid) and 21-MW/cm^2 (open) pump pulses.

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References

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