Isolating Quantum Coherence using Coherent Multi-dimensional Spectroscopy with Spectrally Shaped Pulses

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Abstract: We demonstrate how spectral shaping in coherent multidimensional spectroscopy can isolate specific signal pathways and directly access quantitative details. We identify, isolate and analyse weak coherent coupling between spatially separated excitons in asymmetric double quantum-wells.
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1. Introduction

Coherent multidimensional spectroscopy (CMDS) of electronic transitions has become an increasingly useful and versatile tool for understanding complex systems, from semiconductor nanostructures[1] to light-harvesting complexes from photosynthetic organisms[2,3]. Much of the analysis of the results from these techniques, however, is qualitative and relies on fitting complex models to reproduce the experimental data. Here we describe pathway-selective coherent multidimensional spectroscopy based on spectral shaping of the excitation pulses as a means to isolate specific signal pathways and directly access quantitative details. We apply this approach to selectively excite coherent superpositions of excitons that are spatially separated and localised to different semiconductor quantum wells.

Coherent coupling between spatially separated systems has long been explored as a necessary requirement for quantum information and cryptography[4]. Recent discoveries suggest such phenomena appear in a much wider range of processes, including light-harvesting in photosynthesis[2,3]. With the approach we describe here, identification and quantitative analysis of this type of weak coherent coupling between distant quantum systems becomes simpler and more powerful.

2. Experimental and Results

We utilize a CMDS setup based on spatial light modulators, similar to the one developed by Nelson et al. [5]. With this setup we not only have precise control of the pulse delays, phase and spectral phase, but we also have control over the spectral amplitude, which allows us to spectrally shape the individual pulses to excite specific quantum pathways.

In the asymmetric double quantum wells studied here there are two GaAs quantum wells of different width separated by an AlGaAs barrier. In general, there are four exciton states that can be observed (Fig. 1(a)), two localized to the wide well and two localized to the narrow well. In this system, it is expected that coherent coupling between excitons localized in the same well is strong, while coherent coupling between excitons localized in different wells is weak. In broadband CMDS experiments clear signatures of coherence between excitons in the same well can be identified and even analyzed due to them being well-separated in a 3D spectrum. Signatures of coherence between two pairs of excitons localized in different wells can also be identified, but for these peaks little other analysis is possible because the weak coherence signal is swamped by other signal contributions.

By spectrally shaping the first two pulses so that the first is resonant only with the excitons in the narrow well and the second pulse is resonant only with excitons in the wide well, as shown in Fig. 1b, we are able to selectively excite the coherences between excitons localized in different quantum wells. The resulting 3D spectrum, Fig. 1c, shows not only peaks due to the two inter-well coherences identified in the broadband experiment, but also two due to additional inter-well coherences. All four peaks are ‘cross-peaks’ in the projection onto the (ω_1, ω_3) plane (the standard 2D spectrum) and are shifted along ω_3 by the relevant and expected energy differences for these coherent superpositions.

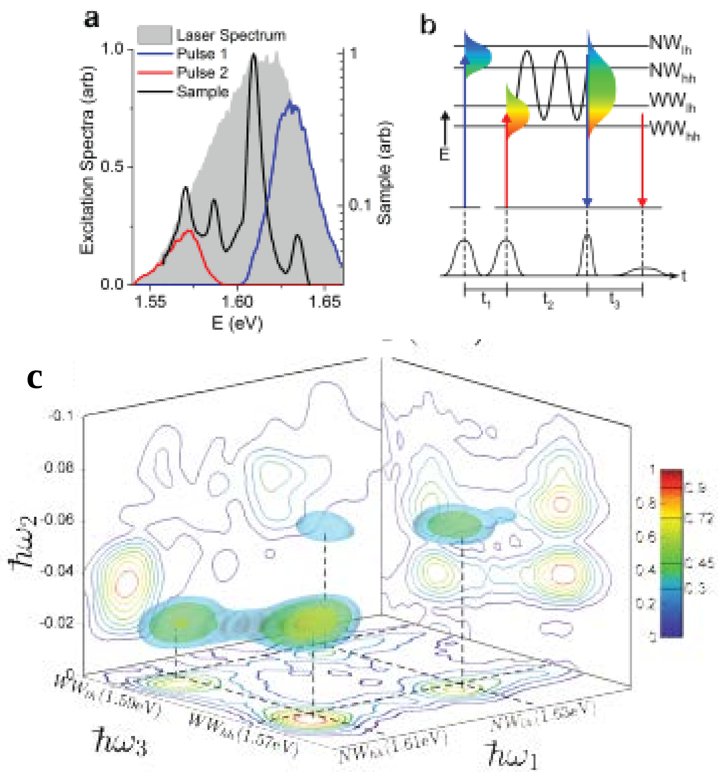

Fig1. (a) shows the spectrum of the asymmetric double quantum well sample (black line), with the four spectral peaks, together with the laser spectrum for the broadband (shaded) and the two shaped pulses (red and blue). (b) shows the coherence specific excitation scheme utilized and (c) shows the 3D plot under these excitation conditions.

The identification of all four of these inter-well coherences, where the excitons are well-separated with no spatial overlap, is a significant result in itself, however, the ability to isolate in three spectral dimensions allows further unprecedented analysis. We have developed new and enhanced existing[6] techniques to analyze such three-dimensional peak shapes and reveal details of the interactions between the different spatially separate states and the extent of any correlated broadening. Finally, with a dynamic range $>10^4$ in electric field amplitude this approach allows us to quantitavtively compare different signal pathways and in the present case precisely determine coupling strengths and there dependence on a range of parameters.

3. Conclusions

We have devised a pathway specific CMDS experiment that combines an ability to selectively excite specific quantum pathways by spectrally shaping the excitation pulses with the many of the benefits of CMDS. The ensuing ability to identify, isolate and analyse coherences, and indeed any specific signal pathway, can provide significant insight into the interactions and dynamics in a range of complex systems. In photosynthetic light harvesting complexes, for example, this type of approach has the potential to resolve important questions regarding the nature and role of quantum effects in efficient energy transfer.

4. References

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