Optical coherence in semiconductor quantum structures

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Work in the last year has revealed new coherent behaviours of coupled modes of excitons and light, including the unexpected influence from disorder. New experiments studying the scattered light from semiconductors demonstrate our incomplete understanding of localisation in quantum wells. Semiconductor coherence now enables directional control of electron currents.

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Abbreviations

PL	photoluminescence
QW	quantum well
VCSEL	vertical cavity surface emitting laser

Introduction

The investigation of coherent interactions between light and matter is fundamental in understanding and manipulating the absorption and emission of photons from semiconductors. It draws strong interest from researchers in a wide variety of fields from ultrafast physics to optoelectronics (discussed in a recent special issue of the *IEEE Journal of Selected Topics on Quantum Electronics* [1•]), and continues to produce new surprises. Work in this field can be divided into two areas in which firstly, the coherence is stored in static excitations of the semiconductor; and secondly, the coherence is stored in a circulating optical field within a semiconductor cavity. Coherence phenomena involving polaritons (mixed photon-exciton modes) are complicated and enhanced by both areas simultaneously. Interest in long-lived coherences in solids has recently been further stimulated by suggestions for quantum computing applications. The following review concentrates on work published in 1997, including relevant papers from shortly before that time where appropriate.

Observation of coherent optical effects in semiconductors implies that the phase of light entering a sample is imposed on and transiently retained in the electronic excitations. Generally, for direct gap semiconductors, the process of optically exciting electrons from the valence to the conduction band necessarily produces an interband polarisation creating a population of electron-hole pairs at the same time. Because the photoexcited carriers suffer considerable scattering processes, both with each other and from acoustic and optic phonons, this coherent interband polarisation decays to zero, typically on timescales faster than 10 ps. For this reason, many of the experiments are performed using laser pulses shorter than 1 ps so that the coherent interactions can be resolved before they decay.

Coherent control

This is an emerging new field for semiconductor coherence where the electronic wavefunctions are specifically manipulated by optical pulses. Several results offer some encouraging progress. The ability to excite electrons in GaAs with a net velocity in a particular direction was recently demonstrated using simultaneous laser pulses at 775 nm and 1550 nm [2••]. Impinging light creates equal numbers of electrons travelling in opposite directions, however, interference with a laser field at twice the fundamental band gap is able to coherently destroy electrons moving in one direction, and enhance electrons going in the opposite direction, producing a measurable photocurrent in an unbiased contact at 300K.

The author's own coherent destruction experiments now extend to the control of exciton spin as well as exciton density [3•], while Marie *et al.* [4•] have shown the ability to fully characterise the different coherent relaxation times in GaAs quantum wells (QWs). Thus the optical dephasing, the quantum spin coherence and spin decay times can be determined in the same arrangement. Various new possibilities have been envisaged, such as the manipulation of carrier localisation in semiconductor heterostructures using optical and microwave electromagnetic fields [5].

The possibility of using coherent control for ultrafast optical switching has been critically evaluated [6] suggesting that coherence times must be sufficiently long and that waveguide geometries may be needed for switiching action. One possible solution is to amplify the effect using square pulses to coherently control semiconductor microcavities [7•]. However, I believe this will prove impossible in practice due to the polaritonic modifications to the phase scattering in microcavities.

Interferometry and coherent scattering

Extensions to previous techniques have dramatically improved the measurement of coherence memory in semiconductors. Likforman and colleagues [8•] have demonstrated the use of Fourier Transform spectral interferometry on photon echoes in GaAs QWs while other groups have used similar techniques to examine the four-wave-mixing emission [9]. The general spectral method utilises the interference of a reference pulse with the nonlinear emission from a sample excited by a second pulse, detected using a spectrometer with a multichannel readout, which is fast compared to the phase jitter between the two pulses. Although the technique is efficient at extracting both the amplitude and phase, it potentially suffers a compressed dynamic range restricting its use in many experiments. An alternative technique uses the temporal interference of the emitted light with a delayed phase-tracked reference pulse which enables detection of emitted intensities down to 10^{-12} of the incident pulse, and detailed phase dynamics to be studied [10•]. Similar techniques have been used to study higher order nonlinearities in GaAs [11].

A related method has been used to discriminate between the coherent and incoherent luminescence emitted from a resonantly excited QW [12•]. Using a pulse sequence equivalent to coherent control, and detecting the emitted light through a narrowband filter produces characteristic interference fringes attributable to either resonant Rayleigh scattering or incoherent photoluminescence (PL).

One of the most intriguing recent puzzles involves the role of Rayleigh scattering following excitation with a femtosecond pulse resonant with the QW exciton [13••]. In this case the light emitted away from the reflected direction is time-resolved by upconversion, resolving a slow several picosecond rise at low densities. The experiments by Haacke *et al.* [13••] show coherent polarisations existing within the sample during the Rayleigh process, while coherence with the incident light is under intensive investigation. Further Rayleigh scattering experiments on heterostructures in which there is some control of the lateral disorder length scale [14] are also under coherent investigation.

Two sub-100 fs experiments have added to the discussion about whether it is possible to observe breaking of energy conservation on sufficiently short timescales in semiconductors, a property known as 'quantum kinetics'. In the first, the dominant scattering is by emission of longitudinal optical phonons [15•] whereas in the second the scattering is from electron–electron scattering [16]. In both cases, coherence is invoked to describe how relaxation can be be driven by the memory of the system. However, the results depend heavily on detailed theoretical analyses and it seems difficult to extract a clear signature of this phenomenon.

Lower dimensional structures

Current technology has allowed realisation of quantum wire and quantum dot nanostructures. The reduced dimensionality is expected to retard phase scattering by reducing the number of energy levels available for the carriers to be scattered into. This is indeed seen to be the case from the much narrowed PL spectra taken on isolated single quantum dots [17]. Further work in this area has examined near-uniform collections of dots [18] (which, however, show no absorption), and theoretical work has managed to treat realistically the crucial Coulomb interactions between electrons and holes in wire structures [19•].

A variety of other experiments are also noted here. Improvements in the description of nonlinear optical driving of semiconductors continue [20,21], implicating, as previously, the Coulomb-induced local fields in the response. Coherent experiments in the band-to-band transitions, of GaAs, show signals beating in time which allows the dispersion of the hole bands to be roughly mapped [22•]. Further work on double quantum wells highlight Fano resonances which can result in coherent modifications of carrier tunneling rates [23]. Four-wave-mixing continues to be used as a probe of optical coherence in new material systems such as GaN [24].

Optical memory

Experiments in laser cavitites are generally less concerned with the fundamentals of semiconductor coherence. However, a few papers are worth mentioning in the present context. One theoretical idea under investigation uses the nonlinear properties of a semiconductor sandwiched between Bragg mirrors to produce bistable devices. The novel component in this scheme is the discovery that spatial solitons can be formed from lateral confinement by the nonlinear refraction [25•]. Another beautiful manifestation of coherence in vertical cavity surface emitting lasers (VCSELs) showed that the electron Larmor precession induced by a magnetic field can oscillate the circularity of the polarised laser emission [26•]. Finally an excellent example of the possibilities afforded by arrays of VCSELs is seen in their potential use for investigating spatial coherence and photon localisation [27].

Polaritons

A large body of new work has involved the properties of polaritons, known for many years, but only now being exposed in detail in semiconductors. The coupling between coherent semiconductor polarisations and electromagnetic transport leads to new polariton modes of the sample. One excellent demonstration of these properties is given by suitably adjusting the spacing between quantum wells to produce superradiant emission [28••]. Polaritonic effects may also be observed in conventional multiple quantum well stacks [10•] as well as in the propagation of optical pulses through bulk CdS crystals [29]. In addition there is a large body of theoretical work on this problem trying to analyse the physics in terms of photon propagation [30] or in terms of the polariton normal modes of the structure [31]. The latter treatment produces an extremely surprising result when inhomogeneous broadening is taken into account, in that even a single quantum well can exhibit beats in time resolved reflection [32••]. Recent experiments in quantum wells show coherent reemission but no beating though this depends extremely sensitively on the exciton wavelength [33]. The lateral transport of excitons seems to be retarded by this coherent reemission. There is also some discussion as to how, in the limit, a stack of 2D quantum wells transforms into an optically 3D material [34], as always, this is complicated by the need to consider exciton-free boundary layers. Additional models including bipolaritons suggest rather unconventional explanations for PL emission in GaAs QWs due to interface polaritons [35].

When polariton structures are folded up, enclosing a single (or few) QWs in a wavelength-scale cavity, the resulting microcavity can display extremely strong polariton modes. Recent results have indicated that sample disorder can also be averaged out by the more extended polariton states, an effect known as 'motional narrowing' and further theory builds this idea more rigorously from microscopic models [36•] (D Whitakker, personal communication). Work is energetically underway on the Rayleigh scattering signals from such structures, and theory indicates that there should be a strong link between the 'motional narrowing' and the rise in the Rayleigh signal [37]. Microscopic theory has also shown how femtosecond pulses build up inside the cavities [38]. Coherent nonlinearities studied in these systems show the destruction of the polaritons by saturation of the excitonic transition at high powers [39•]. Four wave mixing in QWs [40•] and bulk [41] microcavities demonstrates the persistent influence of the polariton quasiparticles in all optical properties, both linear and nonlinear. Further theory has been developed to examine the possibility of intersubband enhanced emission in similar structures [42].

Interest in Bloch oscillators appears to have become more subdued though direct measurements of the oscillating wavepacket have been neatly devised [43•]. Theory now grapples with the correct treatment of phonon damping and there are predictions of its influence on the terahertz emission ([44]; also see the review by Sherwin, this issue, pp 191–197).

Finally the possibility for clearly observing Bose–Einstein condensation in quantum well structures is still thought to be possible, and has been reviewed positively in [45•].

Conclusions

Optical coherence is strongly represented in several current areas of semiconductor spectroscopy. It is at the root of the quantum electrodynamical behaviour of semiconductor microcavities and propagating polaritons, in which it is responsible for producing entirely new properties. The coupling between light and confined electrons allows the engineering of new linear and nonlinear features, promising the prospect of novel devices. New physics has also emerged from consideration of inhomogeneous carrier distributions, which are now found to even enhance electromagnetic interactions, and produce unexpected transport behaviours. Coherence also enables coherent control of photoexcited photocurrents inside semiconductors. Further developments in this area may, however, require exploitation of new long coherence decay materials such as active devices. The study of the very earliest coherent emission from semiconductor quantum structures challenges our description of absorption and luminescence. Progress on this problem seems certain to be successful in the near future. Finally, there is considerable effort in eliciting new coherent physics from semiconductors along the path to creating macroscopic coherent states.

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