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Ultrafast Coherent Carrier Control in Quantum Wells

J.J. Baumberg (a), A.P. Heberle (a), K. Köhler (b), and A.V. Kavokin (c)

(a) Hitachi Cambridge Laboratory, Madingley Road, Cambridge CB3 0HE, U.K. Tel.: +44 (1223) 467944; Fax: 467942; e-mail: baumberg@phy.cam.ac.uk

(b) Fraunhofer-Institut für Angewandte Festkörperphysik, D-79108 Freiburg, Germany

(c) Istituto Nazionale per la Fisica della Materia, Il Universita di Roma 'Tor Vergata', I-00133 Roma, Italy

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A fast scanning ultrafast phase-dependent reflectivity technique is used to measure the coherent properties of multiple quantum wells at carrier densities below 10^8 cm^{-2}. Signal to noise ratios in the reflected intensity >80 dB can be achieved allowing underlying coherences to be measured at time delays beyond 30 ps. An unexpected 1.5 ps rise time of the coherence is resolved before a non-exponential decay and ascribed to interference of polaritons. By time-resolving the pair of pulses reflected off the sample, a complete history of energy flow in the quantum wells can be identified. Both spontaneous and stimulated coherent reemission are observed in good agreement with models based on the Bloch equations.

Introduction. The optical response of a semiconductor depends on the exact state of the electronic system. This is well understood for nonlinear changes in absorption and refractive index induced by high light intensities. Such nonlinear effects disappear at low intensities since the photoexcited electrons are sufficiently separated that carrier–carrier interactions become negligible. This picture must be supplemented with the phase-dependent nature of the light-matter interaction. Even at low carrier densities, this implies that the optical response can depend strongly on the previous state of the semiconductor. Previously we have demonstrated that one can utilise this coherent memory of the optical phase in the interband polarisation to create and destroy excitons on ultrafast time scales [1 to 3].

In this paper we extend previous measurements of the density and polarisation of excitons injected into quantum wells (QWs), to examine the light emerging after the interaction. We use a fast scanning modification of phase-locked pulse sequences to extract both the integrated and time-resolved phase-dependent reflectivity (ΔR_{coh}) of QW excitons. At ultralow densities the decay of the coherent excitonic response becomes density independent as expected, however, an unexpected rise in the coherent signal after initial excitation is resolved which vanishes at higher densities.

Experiment. To identify the phase dependence of the sample response we focus two collinear equally-intense 100 fs pump pulses onto a 200 μm spot on the sample and record the optical response as a function of the known phase difference between the

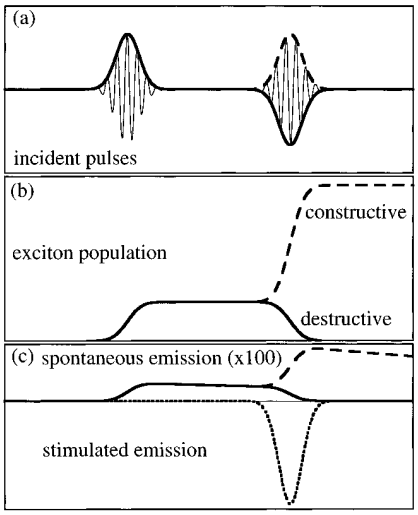

Fig. 1. Coherent control experiment. a) Incident pair of pulses are constructively- (dashed) or destructively-phased (solid). b) QW exciton density after excitation. c) Spontaneous and stimulated emission from the QW in the reflected direction as predicted by the semiconductor Bloch equations in the two scenarios

pulses. The timing of the pump pulses is controlled using a Michelson interferometer [2]. Previously the density of photoexcited excitons was investigated as the optical phase varied [1]. With the two pulses destructively configured (Fig. 1, solid), the excitons created by the first pulse are destroyed by the second laser pulse, returning the carrier density almost to zero. By contrast, changing the interpulse time delay by 1.34 fs so that the pulses now produce constructive effects, the absorption of the second pulse is quadrupled (dashed).

By modifying the experiment, extremely high signal to noise data can be resolved allowing access to a wide variety of measurements not easily reached using conventional techniques. Instead of actively lock-ing the phase between the two incident pulses, the relative phase (and hence time delay) is rapidly scanned by a vibrating mirror mounted in one arm of the Michelson interferometer (Fig. 2) [4,5]. By recording simultaneously the fringes of a collinearly-aligned HeNe laser and the total reflectivity of the double pulse train using a fast ADC computer board, the phase-dependent reflectivity component can be extracted. Since the frequency of the scanning fringes is oscillating at several Hz, this lock-in technique tracks a variable reference frequency (around 100 kHz) and rejects noise at other frequencies. Fast interpolation of the data using high-speed microprocessing means that entire time scans can be continually averaged to further improve the signal to noise. The success of this technique relies on accurately tracking the HeNe fringes as inaccuracies which miscount fringes will rapidly wash out any features in the infrared reflectivity data. To further improve the data, a balanced reflectivity geometry is adopted in which the light reflected is referenced to that incident on the sample to highlight their difference. This substantially reduces the large coherent spike at zero time delay produced by the direct temporal overlap of the two pulses and reduces contributions from laser noise at the fringe sweep rate. Collinear alignment of the

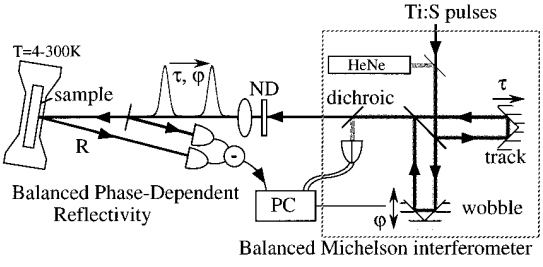

Fig. 2. Experimental scheme: pulses from an externally-recompressed mode-locked Ti:S laser are used to produce pulse pairs with variable but tracked temporal separation (±10 as)

two pulses is critical and is confirmed by test scans of the fringe visibility with the pump laser in CW mode. These precautions allow detection of coherent reflectivity amplitudes, under optimum conditions, of 1 part in 10^6 compared to the incident pulses. This extreme sensitivity emphasises contamination by satellite artefacts: any weak stray collinear pulses produced by optical components in the beam line will generate spurious transients which are also phase dependent. In the current experiments unavoidable residual satellites appeared at intensity levels of 1 ppm. In addition to coherent exciton dynamics, this optical Fourier Transform spectroscopy is also well suited for isolation of weak sharp spectral features on a large background (e.g. for single quantum dots [6]).

Time-Integrated Results. The sample which is used for these results has 5 QWs of 12 nm GaAs separated by 12 nm $Al_{0.3}Ga_{0.7}As$ barriers, and is that previously used for coherent control experiments to facilitate direct comparison. Similar features have been seen in other high-quality QW samples. The reflection spectrum at $T = 4$ K together with the photoluminescence and typical laser spectrum are shown in Fig. 3. Distinct features arise from the heavy- and light-hole QW excitons and the 1s bulk exciton in the underlying GaAs buffer.

The amplitude of the coherent reflectivity oscillations (ΔR_{coh}) records how much the reflectivity of the second laser pulse has been altered by the phase of the first laser pulse. Because ΔR_{coh} is linear in the intensity of the laser pulses, very low excitation intensities can be used. This contrasts with more conventional four-wave mixing techniques which are third-order in the intensity and thus extremely weak at such low carrier densities. Fig. 4 shows ΔR_{coh} at carrier densities around 10^8 cm^{-2} for several detunings (Δ) from the main heavy-hole exciton. When the laser is tuned to high energy ($\Delta = +4$ meV) strong light-hole−heavy-hole beating shows that coherence within the valence band remains up to 6 ps after excitation. Conversely at negative detunings ($\Delta = -15$ meV), a strong interference of the heavy-hole exciton with the bulk exciton is seen which persists for extremely long time delays >30 ps (not shown). This signal reflects the fact that at late times the coherent signals originate only from regions of the semiconductor optical spectrum that retain longlived memory of the original laser phase. At optimal detuning for the heavy-hole exciton ($\Delta = -4$ meV), a signal with minimal beating is seen which rises for the first 1.5 ps before decaying non-exponentially with a decay time of ≈3.5 ps. The decay time speeds up at higher laser energy reflecting the faster phase scattering during carrier relaxation. Also shown (dashed) is the interferogram of the excitation laser pulses which exhibits a much faster fall-off than the rise in the signal. This rise in the signal at early times is extremely unusual since it is expected that phase scattering persistently acts to *decrease* the coherent memory of the exciton.

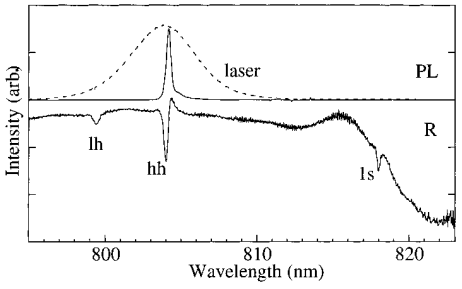

Fig. 3. Reflection (R) and luminescence (PL) of the 5 QW sample (see text) at $T = 4$ K

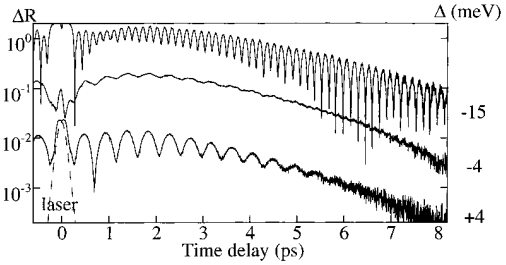

Fig. 4. Phase-dependence of the time-integrated reflection of the double pulse as a function of interpulse delay. The amplitude of the 2.67 fs period oscillations has been extracted and shown as a function of the detuning of the laser from the heavy-hole exciton (Δ). The upper two curves are shifted for clarity. Also shown (dashed) is the amplitude of the direct interference of the laser pulses

Fig. 5 shows that the density regime probed is indeed below that in which most nonlinear processes act. The decay time of the signal only reduces at carrier densities above a few 10^9 cm^{-2} as expected from carrier–carrier scattering processes. The early rise in the coherent reflectivity begins to wash out at carrier densities of mid-10^8 cm^{-2}. At lower carrier densities no change is seen in the signal amplitude. However, Fig. 6b resolves changes in the exciton *phase* (corresponding to sub-Å energy shifts) produced by even the weakest excitation used. Such changes are hard to resolve using traditional spectroscopy, because they integrate over all time, masking the dynamics. There is some evidence to suggest that such lowest order nonlinearities in quantum wells are due to acoustic phonon emission and not Coulomb mediated scattering which dominates at high densities [6].

A complete explanation for the rise in the coherent reflectivity seen is still lacking. The feature is found to be independent of polarisation, angle of reflection, quantum-well width (similar for 25 nm QWs) and pulsewidth (from 60 to 200 fs). A rise in the strength of scattered light due to resonant Rayleigh scattering has been recently observed [7,8]. However, only decays are expected for the directly reflected beam and indeed our signal is independent of the position on the sample as opposed to [7]. A simplistic interpretation of the data would imply that the coherent oscillator strength increases for the first few ps after carrier injection. Rising signals have been seen in time-resolved four-wave mixing experiments and attributed to inhomogeneous broaden-

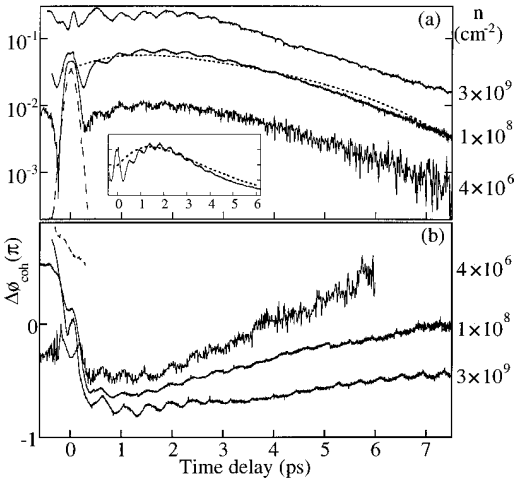

Fig. 5. a) Phase-dependent reflection of the double pulse at different incident intensities showing the rise and decay of the exciton coherence. The inset is for medium densities, plotted on a linear scale. The dotted curve is a fit from polaritonic interference in this sample using a purely radiative homogeneous linewidth, and an inhomogeneous linewidth of 0.2 meV. b) The extracted phase of the coherent reflectivity at different densities

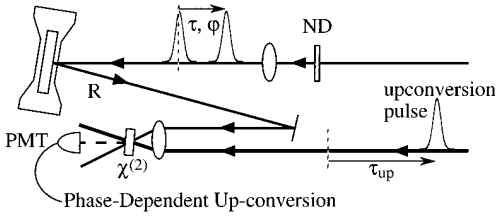

Fig. 6. Experimental scheme for up-conversion of the reflected light from the sample

ing [9] and exciton–exciton Coulomb correlations at higher densities [10]. In addition, coupling between polaritons has been shown theoretically to produce a rise in the emerging signal from quantum wells [11,12]. Finally non-Markovian effects might also be expected to play a role on these time scales. Here we consider the scenario based on the combination of polariton effects and inhomogeneous broadening [12]. As soon as two bright polariton modes are present, it becomes possible to excite their superposition, *which is initially not in phase*. This is because each mode has an associated phase spectrum, which modifies the phase of the local field exciting the other mode. Inhomogeneous broadening is then responsible for eliminating additional rephasings at later times, and causing the non-exponential, Gaussian decay of the coherent reflectivity which is observed in a fit derived from [12] (Fig. 5a, dotted curve). Because the homogeneous exciton linewidth is extremely small (being predominantly radiative), polaritons are produced by optical coupling between resonant regions of *different* quantum wells. The inhomogeneous collection of these polaritons is probed here. At higher densities the homogeneous linewidth increases, destroying the polaritons and washing out the early rise. Further analysis is in progress to confirm this explanation, which suggests the importance of polariton effects in coherent spectroscopies on band-engineered semiconductor heterostructures.

Time-Resolved Reflection. Modelling of the coherent control experiments using the semiconductor Bloch equations implies that the second laser pulse is either amplified or deamplified by the coherently excited semiconductor [3]. This can be verified by time-resolving the pulses reflected off the sample. The reflected light is focused into a nonlinear crystal and mixed with an upconversion pulse derived from the same laser (Fig. 6). By measuring the incoherent (i.e. total) and coherent (phase-dependent) upcon-

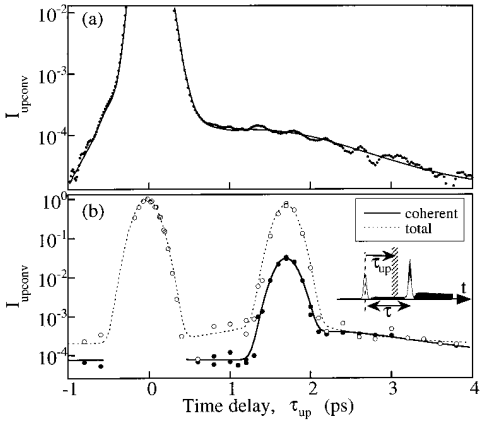

Fig. 7. a) Time-resolved reflection of a single weak pulse. b) Phase dependent (solid), and phase-independent (dashed), contributions to the time-resolved reflection of the pulse pair, which are separated by a fixed delay, $\tau = 1.8$ ps

verted blue light, the complete emission dynamics of quantum wells can be extracted. Fig. 7a shows the direct upconversion of a single incident pulse demonstrating the difficulty for conventional time-resolved techniques in resolving the short-time behaviour. The QW emission is compatible with the coherent reflectivity data in Fig. 5 although an initial rise is not resolved due to the strong reflected pulse from the sample surface. Fig. 7b shows both the incoherent and coherent time-resolved signals from the reflected pulse pair at a fixed separation of 1.8 ps. Before the second pulse arrives, the reflection is independent of the interpulse phase as expected. However, the reflection of the second pulse is seen to have a strong component (4%) which depends on the phase of the first pulse. As seen in Fig. 1c, coherent destruction of the excitons is connected with the stimulated emission of their energy back into the second pulse, clearly visible in the data. After the second pulse, the re-emission from the quantum well becomes completely dependent on the phase difference between the pulses. This shows that all of the carriers which were injected in the quantum well can be removed by the second pulse thus turning off any further spontaneous emission in the forward direction.

The stimulated reemission by the second laser pulse can be tracked as the first pulse is further separated in time (Fig. 8a). The upconversion window is now fixed coincident with the second laser pulse, and the delay between the two pulses is varied. The total light collected quadruples when the two pulses are simultaneous, due to upconversion of the direct pulse interference. If the first pulse arrives after the second pulse then no coherent contribution to the reflectivity of the second pulse is seen, whereas in the reverse order a rise and subsequent decay of the coherent stimulated reemission is recorded, as expected. If the upconversion time window is shifted to occur *after* the second pulse (Fig. 8b) the same rise and decay is recorded in the spontaneous emission from the QW, although the coherent component now decays on a faster time scale than the total.

The data allows the complete reconstruction of the fate of light impinging on the QW. Because the second pulse completely removes the excitons from the sample, the amount of stimulated emission measures the total energy which was stored in the sample. The rate of spontaneous coherent reemission of this energy in the absence of a second pulse is measured here to be a factor of 4×10^{-3} smaller than the stimulated rate which implies that the reradiation in the absence of other scattering processes would take 24 ps, in agreement with theory [12]. Hence the 3.5 ps decay observed is due to dephasing. In this sample, a single resonant 100 fs laser pulse deposits about 1% of its energy into the exciton, of which only 15% of this energy is subsequently reradiated and the remaining 85% is scattered irrevocably. With a second laser pulse arriving a few ps later and phased appropriately, all the absorbed energy can be reextracted from the quantum well.

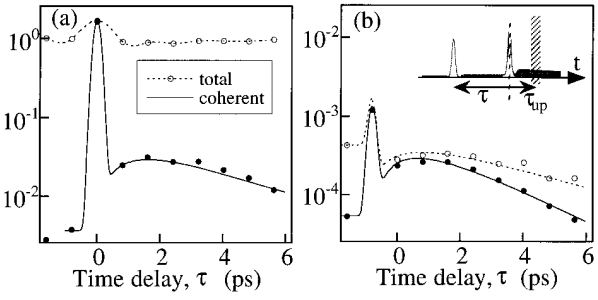

Fig. 8. Time resolved reflection at fixed real time delays, τ_{up}, as a function of the separation of the pulse pair, τ, at a) second pulse maximum and b) 800 fs after second pulse

Conclusion. Time-resolved coherent spectroscopies with ultrafast pulses shorter than the material coherence times provide a powerful tool to probe the optical interaction of semiconductors with light. High signal to noise background-free data is obtained using fastscan techniques which complement traditional linear spectroscopies. These techniques are extremely suitable for application to microcavities and active devices [6]. Time-resolving the emission from the quantum well confirms the model of phase-dependent stimulated emission and the complete fate of light impinging on the sample is recovered. We are currently extending experiments to synthesise both the amplitude and phase of the entire optical waveform for the investigation of novel coherent phenomena.

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