Quantum coherence induces pulse shape modification in a semiconductor optical amplifier at room temperature

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Coherence in light–matter interaction is a necessary ingredient if light is used to control the quantum state of a material system. Coherent effects are firmly associated with isolated systems kept at low temperature. The exceedingly fast dephasing in condensed matter environments, in particular at elevated temperatures, may well erase all coherent information in the material at timescales shorter than a laser excitation pulse. Here we show for an ensemble of semiconductor quantum dots that even in the presence of ultrafast dephasing, for suitably designed condensed matter systems quantum-coherent effects are robust enough to be observable at room temperature. Our conclusions are based on an analysis of the reshaping an ultrafast laser pulse undergoes on propagation through a semiconductor quantum dot amplifier. We show that this pulse modification contains the signature of coherent light–matter interaction and can be controlled by adjusting the population of the quantum dots via electrical injection.
Cohherent control of the quantum state of matter by light is an established technique, in particular for isolated material systems. The nature of coherent interactions allows to reversibly change the state of matter and the state of the electromagnetic field on an ultrafast timescale without energy loss. A particularly illustrative realization of such a process in a resonantly driven two-level system are Rabi oscillations, showing the complete and reversible periodic excitation of a two-level system. The qubits of quantum computing, for example, are based on Rabi oscillations in single two-level systems. Harnessing coherent processes, the deterministic nature of which allows steering the energy flow in the coupled light–matter system, opens up new avenues for applications also in mesoscopic and macroscopic devices. For a semiconductor device operating in the coherent regime based on Rabi oscillations, the comparatively slow gain recovery process via replenishing the active carriers from a reservoir, hitherto limiting the performance of devices to data rates of about 200 Gb s−1 (refs 11,12), could be avoided. Implementing coherent sub-picosecond pulses with population lifetime limited processes, for example, ultrafast switching, opens up a new field of coherent photonics.

Rabi flopping has been observed in various semiconductor systems of different dimensionality at low temperature, in particular in isolated centres like quantum dots (QDs), QDs are zero-dimensional structures formed by self-organization, and show an atom-like delta-function density of states. They are zero-dimensional structures formed by self-organization, and show an atom-like delta-function density of states. They support localized excitations, although they are immersed in a solid-state environment. With this unique combination of single-particle behaviour and condensed matter characteristics, they are ideally suited for studying the response of a complex system to excitation by a light pulse. In addition, self-assembled QDs are readily integrated into electrically pumped device structures, with the electrical injection allowing to control the initial population of their quantum states.

For QD structures, decoherence times of 50–500 fs, depending on the injection current, have been predicted and determined, making it hard to directly observe Rabi oscillations, as scattering rapidly scrambles the phases, reducing a reversible oscillation to an irreversible equilibration. The coherent light–matter interaction, however, still does leave its traces on the electromagnetic field of the exciting pulse as it leaves the sample, opening an alternative method of measurement. The only necessary condition in this case is that the pulse induces a complete Rabi flop during the time within which the polarization in the medium is not completely damped. Since the Rabi frequency scales with the field strength, strong optical pulses are required.

Signatures of coherent light–matter interaction on the electromagnetic field at room temperature have been predicted in numerical simulations for quantum well (QW) structures. Indeed, Rabi oscillations in a quantum cascade structure with two-dimensional (2D) confinement—at low temperature—have recently been observed by analysing the change in pulse shape of a strong laser pulse propagating through the material. Also, a pulse in a quantum-dash-based device has been shown to exhibit signatures of Rabi oscillations.

Here we show that an intense laser pulse interacting with an ensemble of QDs can drive Rabi oscillations fast enough to be observable even in a solid-state environment at room temperature. The signature of Rabi oscillations is retained even under conditions of electrical pumping, adding the QD inversion as an additional control parameter. Adjusting the QD population via the electrical current allows for switching the modulations of the pulse shape, thus controlling coherent effects through population. These effects have on one hand the potential to be exploited for ultrafast modulation of optical signals. On the other hand, for telecommunications applications it is advised to keep the light intensity at levels low enough to avoid a distortion of the bit sequence due to unwanted coherent modulation.

**Results** Experimental observation of laser pulse reshaping. In our experiment, we observe the changes a strong Gaussian laser pulse undergoes when propagating at room temperature through an In(Ga)As-based QD-semiconductor optical amplifier (SOA). The device offers the possibility of electrical current injection. Laser pulses with a central wavelength of 1,280 nm, resonant to the QD ground-state (GS) transition, are generated in an optical parametric oscillator (OPO) pumped by a Ti:sapphire laser with a repetition rate of 75.4 MHz (Coherent MIRA 900-HP). The pulse profile is a Gaussian function with a temporal and spectral full width at half maximum (FWHM) of 230 fs and 15 nm, respectively. We analyse the pulse shape in amplitude and phase in a heterodyne cross-correlation experiment. A scheme of the set-up is shown in Fig. 1. This frequency-resolved optical short-pulse characterization by heterodyning (FROSH) is capable of tracing the oscillations of the complex electric field convolution with a

![Figure 1 | Experimental method. Schematic of the FROSH set-up for electric field cross-correlation measurement of fs pulses. To illustrate the time resolution of the method, the inset shows a zoom into a representative set of data, the real (blue) and imaginary (red) part of the lock-in signal and the interference pattern of the reference diode with a wavelength of λ = 780 nm (green).](image-url)
resolution better than $\pi/20$, corresponding to 0.1 fs. Being a linear method, and analysing the electric field rather than the intensity, it offers an intrinsically high sensitivity even at low signal levels. A more detailed description of the experimental procedure is given in the Methods section.

The experimentally determined pulse shape modification observed on the output facet of the device for a Gaussian input pulse for different pulse powers and injection currents is displayed in Fig. 2. The experimental results are given as a convolution of the electric field of the probe pulse passing the device with a Gaussian reference pulse. The black lines in the top row and centre row represent the measured convolution in terms of amplitude and phase, respectively. An alternative visualization of the time-dependent information contained in the exciting pulse is a Gabor transformation. This transformation basically yields time-resolved power spectra, as for example, also known from wavelet theory, and is thus discussed in frequency–time space. The bottom row of Fig. 2 shows the results, with the Gabor transform of the experimental signal $S(\tau)$ defined as

$$G(\tau', \lambda) = \int_{-\infty}^{\infty} S(\tau) \exp \left(-i \frac{2\pi\epsilon_0}{\lambda} \tau \ln \left(\frac{\tau' - \tau}{\sigma^2}\right)\right) d\tau$$  (1)

Here, $\epsilon_0$ is the speed of light and $\sigma = 106$ fs is the half-width at half maximum of an elementary minimum uncertainty wavelet. In our experiment, a weak pulse passes the unpumped device without a significant change in the pulse shape (Fig. 2a). Increasing the pulse intensity leads to the appearance of interesting modifications, see Fig. 2b–d, as a result of the fact that the frequency of Rabi oscillations increases linearly with the pulse area that the frequency of Rabi oscillations increases linearly with the pulse area. For a suitable choice of the coupling parameters, the oscillation energy is flowing from light pendulum to QD pendulum and back during the time permitted by the time-dependent amplitude of excitation (during the pulse). The envelopes and phases of the pendula relative to the phase of the excitation are displayed in Fig. 3b,c, respectively, so the fast oscillations (frequency $\omega_1$) are removed for clarity. The time traces of the dynamics of the light pendulum and the QD pendulum both show points in time at which the oscillation amplitude is reduced to a minimum, and a phase jump occurs. From basic mechanics, it is known that in a forced oscillation, we simulate the quantum device by a zero-dimensional two-level system (QD) coupled to a reservoir of charge carriers in a 2D reservoir (QW/wetting layer), being in non-equilibrium as a result of electrical carrier injection (see refs 10,31,32). The model includes 0D–2D in- and out-scattering processes as detailed in the Methods section. The results of the numerical simulations are displayed as red convolution amplitude and phase curves in Fig. 2. The calculated pulse modification is in excellent agreement with the experimental results, in particular it reproduces the coincidence of amplitude dip and phase jump consistently observed in the experiment. Further, it can be seen that our modelling is able to correctly describe the pulse modifications for all the different electrical pump currents and thus allows for a deep understanding of the underlying microscopic processes as will be detailed below.

**Discussion**

To make the complex system of coupled differential equation accessible to physical intuition, we first condense the essence of the dynamic expressions to a basic skeleton of a dipole coupled to a light field, which results in equations formally analogous to the well-known coupled oscillators of classical mechanics, see Methods section. The dynamics of this system is displayed in Fig. 3. We consider a light-field pendulum triggered by an excitation with a frequency $\omega_1$ and a Gaussian amplitude in time, coupled resonantly to a QD pendulum (Fig. 3a). Comparing with the Maxwell–Bloch equations without propagation, the light pendulum is identified with the real part of the electric field amplitude $E(t)$ and the QD pendulum with the real part of the polarization $P(t)$. For a suitable choice of the coupling parameters, the oscillation energy is flowing from light pendulum to QD pendulum and back during the time permitted by the time-dependent amplitude of excitation (during the pulse). The envelopes and phases of the pendula relative to the phase of the excitation are displayed in Fig. 3b,c, respectively, so the fast oscillations (frequency $\omega_1$) are removed for clarity. The time traces of the dynamics of the light pendulum and the QD pendulum both show points in time at which the oscillation amplitude is reduced to a minimum, and a phase jump occurs. From basic mechanics, it is known that in a forced oscillation,
there is a phase difference of about $\pi/2$ between the driving force and the oscillator coupled to it. Thus, the time instants at which the oscillation stops and the phase rapidly changes can be qualitatively understood as the instants at which the role of the respective pendulum changes from driving the oscillation to being driven by the partner pendulum. In a convolution of light pendulum oscillations with excitation oscillations, corresponding to the convolution of the electric fields of probe pulse $E_p(t)$ and reference pulse $E_r(t)$ in our experiment, the signature of switching the pulse envelope modulation is still present (Fig. 2e). This behaviour is consistent with the assumption of a coherent energy transfer between QDs and optical field. Depending on the initial population of the QDs, the Rabi oscillations are phase shifted by half a cycle, in one case starting with absorption of light energy and in the other case starting with amplification by stimulated emission, controlled in the experiment by setting the appropriate injection current $J$. A net effect is thus only visible if the QDs are predominantly either populated with carriers or unpopulated (Fig. 2b,c), in the case of transparency the two anti-phase oscillations are expected to cancel, as they do in the experimental data of Fig. 2c. The dynamics of the coupled QD polarization and population is clearly seen via its back action on the exciting light field.

Figure 4 displays the results of the numerical simulations regarding the dynamics of the inversion and the polarization inside the QDs. In a two-level system, this is usually visualized using the Bloch sphere, based on a coordinate system spanned by the degree of population inversion and complex microscopic polarization, that is, the off-diagonal elements of the density matrix containing the time-dependent terms of the coherent dynamics. For resonant light–matter interaction, the dynamics in the Bloch sphere is restricted to the plane spanned by inversion and imaginary part of the polarization. For a better visualization,
the figure shows the total polarization but only the population dynamics of the QD sub-ensemble resonant to the driving laser pulse (the dynamics of the other QD subgroups is discussed later). The trajectories shown in the Bloch sphere in Fig. 4 correspond to the experimental conditions of Figs 2a–e and reflect the interaction with the modified laser pulse. At zero current (Fig. 4a), no charge carriers populate the QDs, and the Rabi oscillations start from the south pole of the Bloch sphere with an amplitude governed by the pulse area $\Theta$ of the driving laser pulse. Charge-carrier scattering processes as well as phase shifts of the polarization within the inhomogeneously broadened ensemble lead to a damping of the oscillations along the device (inset in Fig. 4a). For a partially or fully inverted system, the starting point of the coherent population oscillations is in the Northern Hemisphere of the Bloch sphere (Fig. 4b, coherent dynamics at the input facet of the device). The pulse amplification results in the rotation of the Bloch vector becoming increasingly fast during the pulse propagation, reflected in the number of cycles completed by the population in Fig. 4c at the output facet. In the intermediate case of partially filled QDs, the pulse drives at the same time two anti-phase Rabi oscillations, one starting from the lower and the other from the upper state of the two-level system with effectively disappearing signature on the pulse envelope. At high injection current, we see the impact of the injection current and corresponding high carrier density in the 2D reservoir as a reduced dephasing time $T_2$ leading to a weaker contrast in the pulse modulation.

It may seem surprising that clear pulse shape modifications are present even in the inhomogeneously broadened QD ensemble of our QD-SOA, but it will become clear by the following discussion of the dynamics of all different QD subgroups that are separately included in our modelling and displayed in Fig. 5b–f. The red and black lines in Fig. 5a and d represent the calculated electric field envelope and the convolution of the electric field with a reference field equal to the input pulse, at the input and output facets of the device, respectively. Note that the electric field (red line) of the pulse propagating through the sample is significantly distorted. The convolution with the Gaussian reference is causing a smoothing of this pulse shape modification, and translates the electric field reshaping into a dip in the tail of the black convolution curve. The colour-coded contour plots in the second and third row of Fig. 5 visualize the time evolution of the microscopic polarization amplitude of the QD ground states (GSs) for all subgroups of the inhomogeneously broadened ensemble (distinguished by their detuning in Fig. 5b,c), as well as the corresponding GS inversion for all contributing subgroups (Fig. 5c,f). Inspecting the simulated results of Fig. 5b, it becomes visible that at first a polarization is built up by the incoming optical pulse for all subgroups, but with a much higher amplitude for the group in resonance with the impinging light field (see bright yellow structure at $t = 0$). All polarizations then decay with the dephasing time $T_2$ and thus the bright structure centre around the initial detuning of 6 meV disappears. The polarization, in turn, reduces the GS inversion of the QDs by stimulated emission. In our case, the input pulse power is strong enough to drive the carrier occupations of the central subgroups below transparency, as evident from the blue area where the sign of the inversion is reversed in Fig. 5c. This zero crossing reflects a partial Rabi flop. At the output facet (Fig. 5f), the Rabi oscillations are faster because of the increased field amplitude and thus the blue area with negative inversion is compressed in time. The faster Rabi oscillations can also be seen in the polarization dynamics in Fig. 5e. Note that the dynamics of the polarization of the negatively and positively detuned subgroups is not symmetric around the initial detuning of 6 meV owing to the additional frequency components and chirp of the pulse induced along the propagation, which leads to a non-uniform excitation of the individual subgroups.

The electric field is driven by the total polarization of the inhomogeneously broadened QD ensemble, and thus by a superposition of the polarization of all the different subgroups. Consequently, the spectral broadening translates into an additional dephasing of the polarization, which, for the case of a uniform occupation of all QDs, would resemble a Gaussian decay with a time constant of 73 fs. Thus, by introducing an additional dephasing process, the observed coherent signature could in principle also be reproduced with a simple two-level system using effective parameters. Certainly, as verified with our numerics, such an approach cannot accurately describe the observed pump-current dependence of the pulse modulation, for example, the reappearing and disappearing dips in the convoluted signal as well as the gradual loss of coherence at high injection currents. Here a pure dephasing process associated with the current-dependent process in the 2D reservoir accelerates the macroscopic polarization decay and yields $T_2$ times below 100 fs (ref. 23). The limit of instantaneous dephasing is illustrated in Fig. 5d. The blue-dashed line in Fig. 5d represents the calculated electric field convolution at the output facet for the approxima-
tion of adiabatically eliminated polarization dynamics, yielding an incoherent rate equation approach. Clearly, without the coherent polarization dynamics, the observed pulse shape cannot be reproduced.

**Methods**

**Experimental methods.** The device contains 15 undoped QD-in-a-well layers with a nominal QD density of $10^{11}$ cm$^{-2}$ separated by 33–35 nm thick GaAs spacers.
The shallow etched waveguide provides gain guiding on current injection, with the top contact defining an active region with a lateral extension of 2 μm. The length of the waveguide is 1.25 mm. The injection current I is varied from 0 to 200 mA, corresponding to 4.4 kA cm⁻². When pumped electrically, the device shows amplified spontaneous emission (ASE) from the QD GS centered at 1.281 nm (0.967 eV), which is well described by a Gaussian with a FWHM of 0.03 eV.

From pump-probe experiments, the GS transparency current is determined to be Iₜ ≈ 5 mA. At 200 mA, ASE data show that at injection currents > 100 mA (80 mA), the GS reaches the maximal inversion and the GS luminescence intensity saturates.

In the FROSCH experiment, the laser pulse is split into a probe (E_p) and a reference pulse (E_r). A fast lock-in amplifier (Perkin Elmer DSP 7280) detects the lock-in signal spectrally. The beating generated by probe and reference pulse is recorded on two photodetectors in a balanced detection scheme. The detected intensity is filtered by a fast lock-in amplifier (Perkin Elmer DSP 7280). The lock-in signal S(t) of FFT(E_p(t)−E_r(t)) is the convolution of the electric fields of reference pulse and probe pulse.

**Numerical simulations.** The Maxwell–Bloch model consists of coupled coherent optical Bloch equations for the QD inter-band polarizations p_m and the carrier occupation probabilities p_j for each subgroup j of QDs inside the inhomogeneously broadened QD ensemble. The dynamical equations in the slowly varying envelope and rotating wave approximation are given by:

\[ \frac{dp_m}{dt} = -\Delta p_m + \frac{Q}{2}(\epsilon_m^{\dagger} + \epsilon_m) - \frac{1}{2} T_m \]

\[ \frac{dp_m}{dt} = -\text{Im}(\epsilon_m^{\dagger} + \epsilon_m) - \text{Im}(\epsilon_m^{\dagger} + \epsilon_m) \]

The subscript b (c) labels electrons and holes, respectively, and m (GS, ES) labels the confined QD levels, GS and excited state, respectively (with degeneracy v_m). We assume Gaussian distributions of the QD subgroups around the GS and ES centre frequencies, respectively, with a spectral FWHM of AE = 30 meV in agreement with experimental ASE results. The QD states are coupled to a 2D carrier reservoir, represented by a QW-like wetting layer, by means of Auger recombination in the rate 10–1 ps⁻¹ denoted by \( \Gamma_{\text{decay}} \).

The source for light propagation described by equation (5) is the macroscopic polarization defined by \( P_m(\Omega) = 2N QD \sum_j \varphi_j(\Omega)(QD)_j \), where \( QD \) is the QD density, \( \varphi_j(\Omega) \) denotes the probability to find a QD in the jth subgroup. The environmental noise \( x(t) \) is a random Gaussian process with zero mean and spectral density \( S_\text{x}(f) \).

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### Author contributions

M.K., N.O. and Y.K. designed and performed the experiments. J.K. and B.L. carried out simulations. D. B. provided the device. U.W., E.S. and K.L. initiated and coordinated the project. All authors contributed to data analysis, interpretation and conclusions, and writing the manuscript.

### Additional information

Competing financial interests: The authors declare no competing financial interests.

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