Laser-Induced Quantum Coherence in a Semiconductor Quantum Well

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(Received 31 August 1999)

The phenomenon of electromagnetically induced quantum coherence is demonstrated between three confined electron subband levels in a quantum well which are almost equally spaced in energy. Applying a strong coupling field, two-photon resonant with the 1-3 intersubband transition, produces a pronounced narrow transparency feature in the 1-2 absorption line. This result can be understood in terms of all three states being simultaneously driven into "phase-locked" quantum coherence by a single coupling field. We describe the effect theoretically with a density matrix method and an adapted linear response theory.

PACS numbers: 78.47.+p, 42.50.Hz, 78.66.Fd

The phenomenon of electromagnetically induced transparency (EIT) [1] typically involves a three-level system of well-defined parity, having two dipole-allowed transitions ($|1\rangle \rightarrow |2\rangle$ and $|2\rangle \rightarrow |3\rangle$) and a third ($|1\rangle \rightarrow |3\rangle$) dipole-forbidden (Raman) transition. Driving this with a "coupling field," of angular frequency ω_c, resonant with the $|2\rangle \rightarrow |3\rangle$ dipole transition establishes a coherent superposition of the probability amplitudes and makes quantum interference possible. In this case $|1\rangle \rightarrow |2\rangle$ transitions occur through two coherent paths ($|1\rangle \rightarrow |a\rangle$ and $|1\rangle \rightarrow |b\rangle$, where the "dressed states" are given by $|a\rangle = \{|3\rangle - |2\rangle\}/\sqrt{2}$ and $|b\rangle = \{|3\rangle + |2\rangle\}/\sqrt{2}$, whose transition probability amplitudes interfere destructively and cancel the original absorption at energy difference E_{12}. By allowing operation close to a transition resonance (where the material would otherwise absorb strongly), EIT enables the exploitation of highly efficient nonlinear optical processes [2]. Moreover, adding a further incoherent pump source allows the creation of lasing without inversion (LWI) [3].

EIT absorption cancellation depends on the phase of the overall $|1\rangle \rightarrow |3\rangle$ coherence; its dephasing rate (γ_{13}) must be sufficiently small that $|1\rangle \rightarrow |a\rangle$ and $|1\rangle \rightarrow |b\rangle$ paths can interfere. In, e.g., atoms, this happens when the lifetime of $|3\rangle$ is much shorter than collisional dephasing times, but if this condition is not met, quantum interference is not seen and the absorption at $\approx E_{12}$ is described only by Autler-Townes splitting [4]. In the ideal case of continuous monochromatic coupling fields, the incorrectly phased component of the $|1\rangle \rightarrow |3\rangle$ coherence damps out within several Rabi periods but pulsed measurements typically require "adiabatic preparation" techniques to prepare and maintain its phase [1].

To our knowledge observations of EIT have so far been restricted to comparatively sharp transitions in atomic vapors [1,5] and defect levels in crystals [6]. In these the condition that the Rabi frequency $\Omega_{\mathrm{Rabi}} = \mu E/\hbar$ (where μ is a transition dipole matrix element and E is an amplitude of the coupling electromagnetic field) is greater than the dephasing rates is readily achieved. Although there is much theoretical interest [7,8] in EIT, enhanced nonlinear processes and LWI in intersubband semiconductor quantum wells (QW's), the observation of coherent phenomena in these systems is complicated by their large ($\sim$100 fsec^{-1}) dephasing rates. However, QW's have the intriguing property that transition energies, dipoles, and symmetries can be engineered at will and, because of the light conduction band effective masses, intersubband dipole elements are large.

We note that Fano interference schemes, which use strong and appropriately phased tunneling coupling between the QW states and an adjacent continuum to establish the required coherences, have recently demonstrated the viability of quantum interference and transparency in a QW system [9].

Here we study a novel three-level QW "ladder"-type system, in which the states are close, compared with the transition linewidths, to being equally spaced in energy. In this case a single coupling field can simultaneously drive all three states into coherence with their quantum phases "locked" (Fig. 1) so as to produce an enhanced coherent transparency feature in the absorption spectrum. The purpose of this Letter is to report both the first observation of EIT in a QW system and the first observation of "phase-locked" quantum coherence.

The QW sample consists of 40 symmetric 10 nm n-doped ($n_s = 6 \times 10^{11}$ cm^{-2})$In_{0.47}Ga_{0.53}As$ wells with

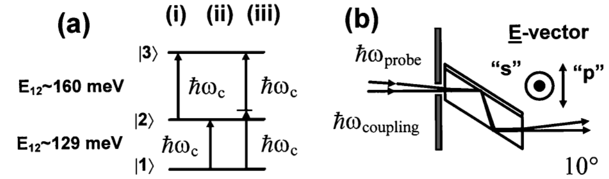

FIG. 1. (a) Schematic of quantum well energy levels and coupling laser resonance conditions. (i) Autler-Townes EIT, $\hbar\omega_c \sim E_{23}$; (ii) strongly driven two-level atom, $\hbar\omega_c \sim E_{12}$; (iii) phase-locked coherence, $\hbar\omega_c \sim E_{13}/2$. (b) Experimental coupling geometry.

10 nm $Al_{0.48}In_{0.52}As$ barriers, lattice matched to an undoped InP substrate. It came from the same wafer as the sample in Ref. [10] but had subband transition energies slightly shifted by wafer nonuniformity. Transition energies (matrix elements) were $E_{12} \sim 129$ meV (2.34 nm) and $E_{23} \sim 160$ meV (2.64 nm).

The sample was polished into a 45° wedge to allow one double-pass of the beams through the QW's (Fig. 1). Absolute transmission values T were obtained by ratioing p-polarized spectra, which couple to the subband system, with s-polarized spectra, which did not. The interaction region was shorter than the wavelengths, rendering dispersion and propagation effects negligible. Independently tunable ($\lambda \sim 6$–12 μm) synchronized coupling (ω_c) and probe (ω_p) laser pulses, with similar temporal profiles (FWHM ~100 psec), were generated in separate Er^{3+}:Cr^{3+}:YSGG-laser-pumped (where YSGG denotes yttrium scandium gallium garnet) optical parametric generators [11] based on $ZnGeP_2$ and CdSe nonlinear crystals. The pulses were sufficiently narrow linewidth and long compared with the subband dephasing and relaxation times for the measurement to be effectively continuous-wave and monochromatic.

Peak effective coupling field intensities, corrected for the experimental geometry and Fresnel losses, of up to $\approx 2.6\ \mathrm{MW\,cm^{-2}}$ were attained over the 300 μm pinhole which was attached to the sample facet for beam overlap optimization. The probe was $\sim 10^{-3}$ of the coupling beam intensity and the beams were separated by 10°. Linear absorption spectra [Fig. 2(a)] gave a Lorentzian $\gamma_{12} \sim 5$ meV spectrum for the unexcited $T = 30$ K sample. Under these conditions only the lowest state was populated so the E_{23} transition energies were measured using the induced absorption method of Ref. [9].

Tuning the coupling energy to the lower transition, ($\hbar\omega_c = E_{12}$) produced an absorption spectrum [Fig. 2(a)], characteristic of a strongly saturated two-level system transition. Interestingly, tuning ω_c to the E_{23} resonance (i.e., the Autler-Townes EIT condition) produced only small (except for the absorption reduction associated with partial populating of level $|2\rangle$) modifications to the absorption spectrum [Fig. 2(b)]. However, when the coupling beam frequency was tuned approximately halfway between E_{12} and E_{23} ($\hbar\omega_c \sim E_{13}/2$), a pronounced (67% reduction in absorption) narrow (FWHM ~3.6 meV) dip appears in the absorption spectrum [Fig. 2(c)]. Its spectral narrowness and the nonobvious relationship between its position (126 meV) and the coupling photon energy (144 meV) both suggest an origin in quantum interference. Moreover, the transparency feature disappeared when, under otherwise identical conditions, the coupling laser was detuned by an equal amount below E_{12} ($\hbar\omega_c = 116$ meV, Fig. 3).

We theoretically treat the three-level system within a density-matrix formalism whose time evolution, within a rotating frame and under the rotating-wave and relaxation-time approximations, is written as

$$
\begin{aligned}
\dot\rho_{11} &= \gamma_2\rho_{22} + i\Omega_{12}(\rho_{12} - \rho_{21}),\\
\dot\rho_{22} &= -\gamma_2\rho_{22} + \gamma_3\rho_{33} - i\Omega_{12}(\rho_{12} - \rho_{21}) + i\Omega_{23}(\rho_{23} - \rho_{32}),\\
\dot\rho_{33} &= -\gamma_3\rho_{33} - i\Omega_{23}(\rho_{23} - \rho_{32}),\\
\dot\rho_{12} &= \left(i\Delta_{21} - \frac{\gamma_{12}}{2}\right)\rho_{12} - i\Omega_{12}(\rho_{22} - \rho_{11}) + i\Omega_{23}\rho_{13},\\
\dot\rho_{13} &= \left(i\Delta_{31} - \frac{\gamma_{13}}{2}\right)\rho_{13} - i\Omega_{12}\rho_{23} + i\Omega_{23}\rho_{12},\\
\dot\rho_{23} &= \left(i\Delta_{32} - \frac{\gamma_{23}}{2}\right)\rho_{23} - i\Omega_{23}(\rho_{33} - \rho_{22}) - i\Omega_{12}\rho_{13},
\end{aligned}
$$

together with $\rho_{ij} = \rho_{ji}^*$ and the carrier conservation condition $\sum_{i=1}^{3}\rho_{ii} = 1$. Ω_{ij} are the Rabi frequencies (assumed real) of the coupling laser field and the $|i\rangle \leftrightarrow |j\rangle$ transition. We note that simultaneous coupling to both upper and lower dipole transitions is explicitly included, with significant consequences as shown below. Δ_{21} and Δ_{32} denote the single photon detunings and $\Delta_{31} = \Delta_{21} + \Delta_{32}$ the two-photon detuning of the coupling laser frequency from the electronic resonances. The γ_i denote the damping of the population states $|i\rangle$ and the γ_{ij} ($i \neq j$) are total coherence relaxation rates, given by $\gamma_{12} = (\gamma_2 + \gamma_{12}^{\mathrm{dph}})$, $\gamma_{23} = (\gamma_2 + \gamma_3 + \gamma_{23}^{\mathrm{dph}})$, $\gamma_{13} = (\gamma_3 + \gamma_{13}^{\mathrm{dph}})$, where $\gamma_{ij}^{\mathrm{dph}}$ is the dephasing decay rate of the quantum coherence of the $|i\rangle \leftrightarrow |j\rangle$ transition. In contrast to many atomic schemes the $\gamma_{ij}^{\mathrm{dph}}$, determined by electron-electron, interface roughness, and phonon scattering processes, are the dominant contributions to the γ_{ij} and the major obstacle to the observation of coherent effects such as EIT in QW systems. We solve these equations in the steady state limit numerically and then use linear response theory to calculate the absorption spectrum as a function of ω_p using the quantum regression theorem [12]. Absorption processes between both upper and lower pairs of states were coherently included.

The results of this analysis are also plotted in Fig. 2. In all cases we set $\hbar\Omega_{12} \approx \hbar\Omega_{23} \approx 5$ meV, corresponding

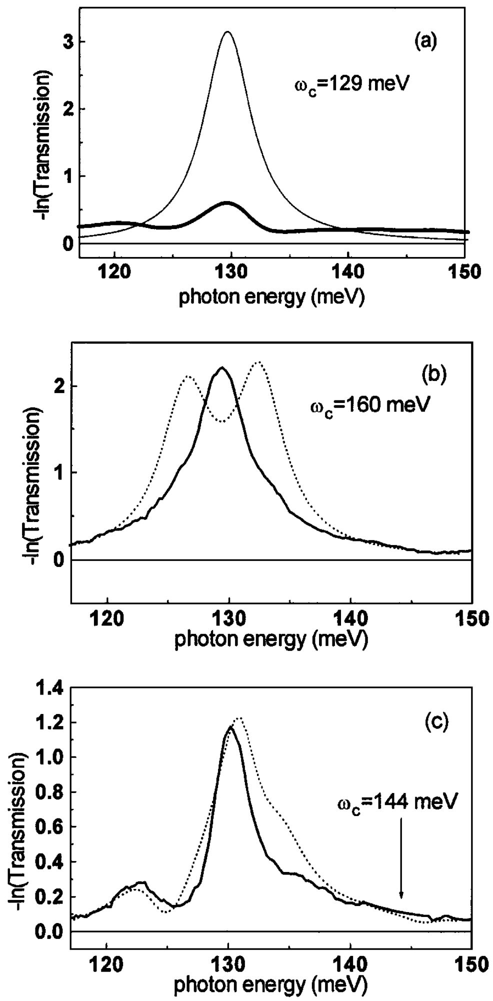

FIG. 2. Measured (heavy solid lines) and calculated (dotted lines) absorption across the 1-2 resonance for different coupling laser frequencies (a) $\hbar\omega_c \sim E_{12}$, (b) $\hbar\omega_c \sim E_{23}$, and (c) $\hbar\omega_c \sim E_{13}/2$. In (a) the light solid line denotes the linear absorption spectrum.

to the coupling laser intensity used, and $\gamma_2 \sim \gamma_3 = 1.3$ meV, equivalent to an intersubband relaxation time $\tau = 1$ psec [13] and $\gamma_{12} = \gamma_{23} = \gamma_{13} = 5$ meV. With these parameters, when $\omega_c = E_{23}$ the theory predicts Autler-Townes splitting but, significantly, no coherent EIT. The overall quantum coherence is not maintained sufficiently to reduce the absorption at line center below that of the two split Lorentzian ($|1\rangle \rightarrow |a\rangle$ and $|1\rangle \rightarrow |b\rangle$) lines. The model assumes a constant coupling intensity but, because the detectors are slow, the experimental signal is averaged over a temporal intensity profile, which is sufficient to obscure the expected incoherent splitting [Fig. 2(b)].

The strongest quantum interference effect is predicted with the coupling frequency tuned to the "two-photon resonance," $\omega_c = E_{13}/2$ [Fig. 2(c)]. When this occurs, the model finds dressed state energies of 132 and 116 meV for the superposition states $|a\rangle$ and $|b\rangle$. Following the principle of EIT, these two states are strongly coherently coupled by the two-photon coupling field. Absorption at ~125 meV, halfway between these two dressed states, is the result of two interfering pathways which contribute to the absorption dipole moment with opposite sign, giving strong absorption cancellation.

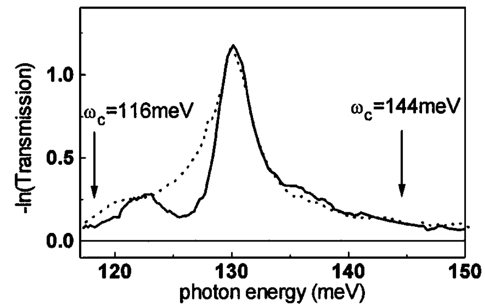

FIG. 3. Comparison between measured absorption spectra with $\hbar\omega_c \sim E_{13}/2$ (heavy line) and $\hbar\omega_c = 116$ meV (light line), i.e., at an equal but opposite detuning from the E_{12} resonance.

The reason that the EIT is more easily observed in the "two-photon resonant" case is that only then is the coupling energy greater than $\hbar\gamma_{13}$; i.e., the $|1\rangle \rightarrow |3\rangle$ coherence is being maintained by the phase-locked two-photon coupling. This is in contrast to the classic Autler-Townes condition, where the two-photon coupling is resonant when $\omega_c = \omega_{23}$ and $\omega_p = \omega_{12}$ but is much weaker because of the lower intensity of the probe.

In both model and experiment (Fig. 2), detuning $\hbar\omega_c$ away from $\sim E_{13}/2$ destroys the interference effect. The fact that the interference feature vanishes when $\hbar\omega_c$ is shifted to an equal but opposite detuning from E_{12} (Fig. 3) argues that it does not originate solely from single photon coupling to E_{12}. Also, at $\hbar\omega_c \sim E_{13}/2$, the critical role of two-photon coupling is evidenced by the strong intensity dependence of the interference feature seen in both experiment and theory (Fig. 4).

A more complete theoretical treatment, taking into account the modification to the transition energies by "many-body" interactions [14] (i.e., Coulomb, exchange, and depolarization energies) in the presence of the coupling fields is beyond the scope of this paper. We note, however, that the inhomogeneous broadening due to conduction band nonparabolicity, $\Delta_{ij}^{\mathrm{npb}} = \hbar^2 k_F^2/2M_{ij}$, where $M_{ij} = [(m_i^*)^{-1} - (m_j^*)^{-1}]^{-1}$, is sufficiently small [15] (~4.9 meV) compared with the unexcited transition linewidth that the "sole collective excitation" picture, as defined in Ref. [13] and assumed in our three-level treatment, should be valid under these measurement conditions. Also we note that "spectral holes," characteristic of inhomogeneous broadening contributions to the transition line shape, were not seen here. Although carrier redistribution within the subbands may alter the transition energies by up to the ~5 meV total many-body energy, this effect is too small to account for the ω_c

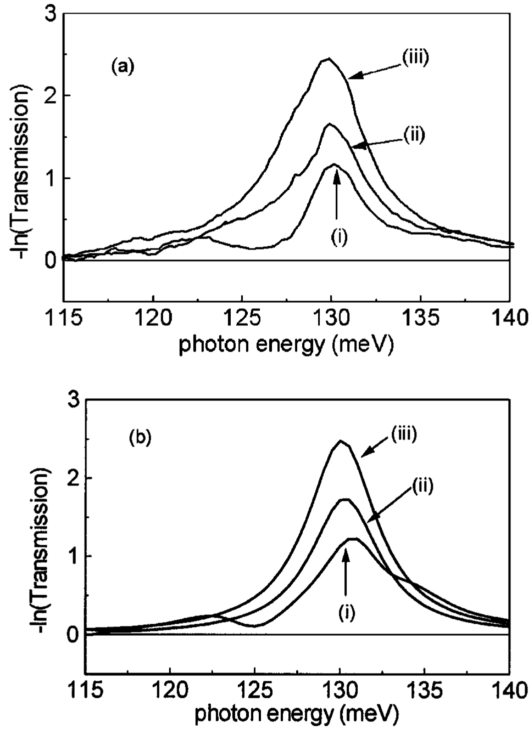

FIG. 4. Experimental (a) and theoretical (b) coupling intensity dependence of the EIT feature for the two-photon resonance case $\hbar\omega_c = E_{13}/2$: (i) $\hbar\Omega_{\mathrm{Rabi}} = 4.8$ meV, (ii) $\hbar\Omega_{\mathrm{Rabi}} = 1.6$ meV, and (iii) $\hbar\Omega_{\mathrm{Rabi}} = 0.48$ meV.

resonance behavior of the phase-locked coherence feature we observe.

In conclusion, we have observed a sharp interference feature associated with electromagnetically induced quantum interference in QW's, which is not related to saturation or spectral hole burning effects. We found that the induced transparency can be observed when the coupling field is two-photon resonant with the E_{13} subband transition, this behavior being fully consistent with a density matrix treatment which simultaneously includes coupling between all states in a three-level system.

We thank Peter Knight, John Marangos, and Matthew Foulkes for enlightening discussions and the UK Engineering and Physical Sciences Research Council for financial support.

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