

Carrier-Envelope Phase-Controlled Quantum Interference of Injected Photocurrents in Semiconductors

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We demonstrate quantum interference control of injected photocurrents in a semiconductor using the phase stabilized pulse train from a mode-locked Ti:sapphire laser. Measurement of the comb offset frequency via this technique results in a signal-to-noise ratio of 40 dB (10 Hz resolution bandwidth), enabling solid-state detection of carrier-envelope phase shifts of a Ti:sapphire oscillator.

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Phase control of mode-locked lasers has enabled revolutionary advances in the fields of both optical frequency metrology [1,2] and ultrafast technology [3,4]. In particular, these lasers make possible the generation of highly phase stable pulse trains [5] that allow for exploration of coherent control-type processes in atomic [6,7], molecular [8], and semiconductor systems [9]. More specifically, control of intense, ultrashort light pulses reveals the phase sensitivity of physical processes in the strong-field regime [10,11]. A recent illustration of this was presented by Baltuska *et al.* who demonstrated influence over the spectrum of high-harmonic generated light by controlling the phase of the driving field [11]. In the multiphoton regime, the photocurrent generated via interaction of ultrashort pulses and a gold cathode was manipulated by controlling the electric field of the pulse train [12].

Phase-stable mode-locked lasers also have strong consequences in the low-field regime given the potential for deterministic control, for example, over the reaction channels in chemical systems and population transfer between quantum states [13]. A convenient medium with which to explore the latter physical process is a semiconductor. In this Letter, we demonstrate directional control of injected photocurrents in low-temperature-grown GaAs (LT-GaAs) by stabilizing and manipulating the phase evolution of the pulses emitted by a Ti:sapphire (Ti:S) laser. This is a new solid-state approach capable of detecting the carrier-envelope comb offset frequency of a mode-locked laser [12,14].

A simple experiment for exploring quantum interference in a semiconductor relies on interfering one- and two-photon pathways between the continuum levels of the conduction and valence bands [15]. In the presence of a two-color light field with frequencies ν and 2ν , quantum interference between one- and two-photon transitions may occur. The carrier population excited to the conduction band states of a given crystal momentum $\hbar\mathbf{k}$ depends on the phase parameter

$$\Delta\phi = 2\phi_\nu - \phi_{2\nu}, \quad (1)$$

where ϕ_ν ($\phi_{2\nu}$) is the phase of the ν (2ν) field. In GaAs, when this phase parameter is such to produce constructive interference at \mathbf{k} it produces, to good approximation, destructive interference for $-\mathbf{k}$ [see Fig. 1(a)].

This asymmetry in population can be understood by considering the \mathbf{k} dependence of the transition amplitudes. The one-photon transition amplitude is proportional to the interband momentum matrix element, which is independent of \mathbf{k} close to the band edge. The two-photon transition amplitude is dominated by two band terms, in which the intermediate band is the same as the initial or final band. The two band terms are proportional to the product of the interband momentum matrix and an intraband momentum matrix element, which is proportional to \mathbf{k} . Consequently, the interference term is odd in \mathbf{k} . The net result of this population imbalance in momentum space is a current density (J) that depends on $\Delta\phi$ through [16]

$$\frac{dJ(\Delta\phi)}{dt} = C(E_\nu)^2 E_{2\nu} \sin(\Delta\phi) - J/\tau, \quad (2)$$

where E_ν and $E_{2\nu}$ are the electric fields oscillating at the optical frequencies ν and 2ν , respectively, τ is a phenomenological current decay time due to electron momentum relaxation (~ 200 fs), and C is a constant that includes the semiconductor nonlinear susceptibility. The first term on the right side of Eq. (2) accounts for the injected photocurrent, while the second term accounts for current relaxation. For a rigorous derivation and explanation of photocurrent injection in semiconductors, see Ref. [16].

Directional control via quantum interference of injected photocurrents in a semiconductor was first demonstrated by Haché *et al.* in bulk GaAs using the fundamental light from an optical parametric amplifier and its second harmonic [9]. The phase parameter between the one- and two-photon transitions, Eq. (1), was adjusted

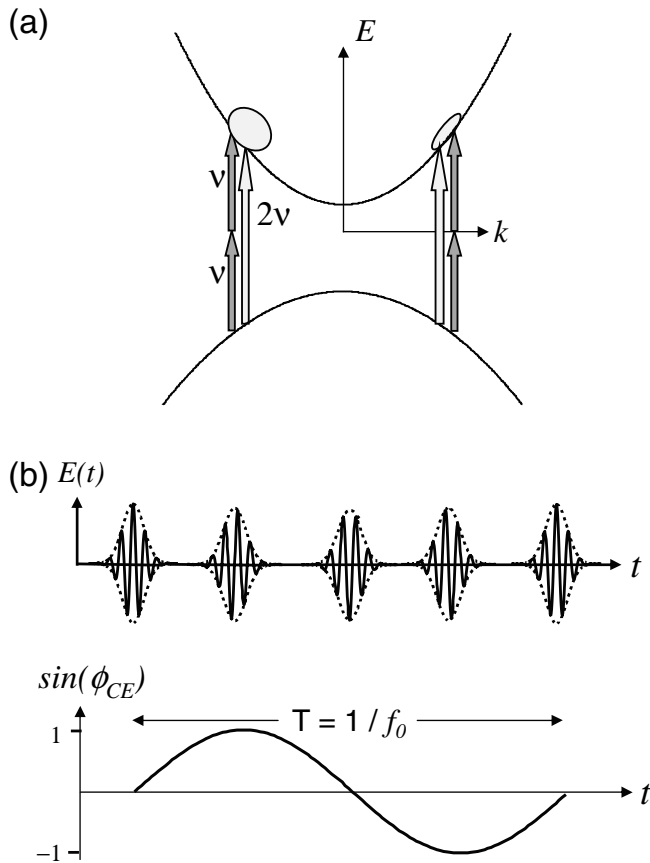


FIG. 1. (a) Schematic depicting the quantum interference of one- and two-photon pathways between the valence and conduction bands in LT-GaAs. Coherent control of the injected photocurrent is obtained using an octave spanning spectrum with a controlled carrier-envelope phase slip. (b) The carrier-envelope phase change between pulses evolves at a defined rate, $f_0 = 1/T$.

using a two-color interferometer and changing the relative phase between the fundamental and second harmonic light using a delay stage. In contrast, for the experiments presented here, quantum interference control (QIC) in LT-GaAs is obtained by adjusting the overall phase of light pulses emitted by a Ti:S laser. This scheme allows for single-pulse control of the injected photocurrent in a semiconductor.

Neglecting cavity dispersion, the optical field of a mode-locked laser is a series of identical pulses separated in time by one cavity round trip, t_{rt} . The formation of pulses results from the coherent addition of a series of extremely narrow cw wavelengths that satisfy the resonant condition of the cavity. Because of this condition, the optical spectrum is a series of equally spaced frequency elements that are integer multiples of the laser repetition rate, f_{rep} , the fundamental resonant mode of the cavity. The inclusion of dispersion, however, complicates the cavity resonance condition, such that $\nu_n \neq n f_{rep}$. Intracavity dispersion causes a difference between the group and phase velocities of the light pulses. This results in an intracavity time rate of change of the carrier-

wave phase with respect to the modulation envelope, $d\phi_{CE}/dt$ [see Fig. 1(b)]. Kerr-lens mode locking of the longitudinal cavity modes transfers the phase information of the laser pulse train to the electric field of each comb element comprising the optical spectrum. That is, $E(\nu_n) = E_{\nu_n} \exp\{-i[2\pi n f_{rep} t + \phi_{CE}(t)]\}$. The common carrier-envelope (CE) phase, ϕ_{CE} , imparted to each mode results in a rigid shift of the optical spectrum [17]. As a result, each optical comb element is shifted such that $\nu_n = f_0 + n f_{rep}$, where

$$f_0 = \frac{1}{2\pi} \frac{d\phi_{CE}}{dt}. \quad (3)$$

Given Eq. (3), coherence in the CE phase of the laser pulse train is obtained via stabilization of f_0 , which fixes the laser cavity dispersion. As shown in Fig. 2, the offset frequency of the laser is measured using one- and two-photon optical interference via harmonic comparison between the comb lines ν_n and ν_{2n} [3,17]. Stabilizing the Ti:S laser in this manner can yield carrier-envelope phase coherence times that extend over tens of minutes [18]. For details about the phase coherence of the laser and the laser itself please refer to Ref. [18].

Control of the injected photocurrent generated in LT-GaAs requires coherently related light with frequencies ν and 2ν . Because the Ti:S spectrum does not span the necessary bandwidth, the laser light is externally broadened in microstructure (MS) fiber [19]. The broadening mechanism coherently increases the existing laser spectrum, thus transferring the CE phase information of the pulse train to the field of the newly generated frequency components. Injected photocurrent in LT-GaAs is obtained by focusing the broadened light between two gold electrodes separated by $10 \mu\text{m}$ (see Ref. [20] for sample details and related work). The generated photocurrent results in a charge separation that is measured as a voltage at the electrodes. LT-GaAs is used rather than GaAs because the trapping of carriers at defects in the LT-grown material removes free charge, thereby inhibiting discharge of the effective capacitor.

To facilitate signal detection and avoid low-frequency noise, we modulate the carrier-envelope phase of the laser pulse train by stabilizing the laser offset frequency to 2.38 kHz. Notwithstanding that ϕ_{CE} is common to light at both ν and 2ν , it results in a phase difference in the transition pathway, $\Delta\phi = \phi_{CE}(t) = 2\pi f_0 t + \phi_0$ [see Eq. (1)]. As a consequence, we observe a QIC signal that varies sinusoidally at the modulation frequency f_0 , with a phase offset ϕ_0 .

The creation of extra above gap carriers, not contributing to the interference signal, can in some instances reduce the detected photocurrent [21]. The loss in sensitivity that results, however, is dependent on several parameters that are difficult to determine exactly (e.g., pulse duration, pulse chirp, and peak intensity). The production of incoherent carriers is of particular concern

when using an extremely broadband source such as that generated from broadening in MS fiber. To ascertain the net affect these carriers have on the photocurrent signal we use incident light that is both filtered and unfiltered. Figure 3(a) shows the spectrum of the photocurrent measured for the two different incident optical spectra [see Fig. 3(b)]. As observed in Fig. 3, the use of above gap light (>874 nm) does not significantly influence the observed signal-to-noise ratio (SNR), which exceeds 40 dB (10 Hz resolution bandwidth). The detected signal, measured using a lock-in amplifier, yields a maximum signal strength slightly greater than $20 \mu\text{V}$ for a pulse repetition rate of 93 MHz, with an average power (spot size) at ν and 2ν of 11.9 mW ($10.2 \pm 0.7 \mu\text{m}$) and 1.53 mW ($11.5 \pm 0.7 \mu\text{m}$), respectively. Currently, the SNR is dominated by electronic pickup from the sample, although distinct sidebands appear on the carrier due to light noise, which we believe to result from inadequate phase noise suppression in the stabilization feedback loop.

Next we use lock-in detection to demonstrate the sensitivity of the QIC signal to small static shifts in the carrier-envelope phase. To do this, we insert a $176 \mu\text{m}$ zinc borosilicate glass plate between the MS fiber and the interferometer used to lock the laser offset frequency. Rotation of the glass plate results in a shift in ϕ_{CE} (due

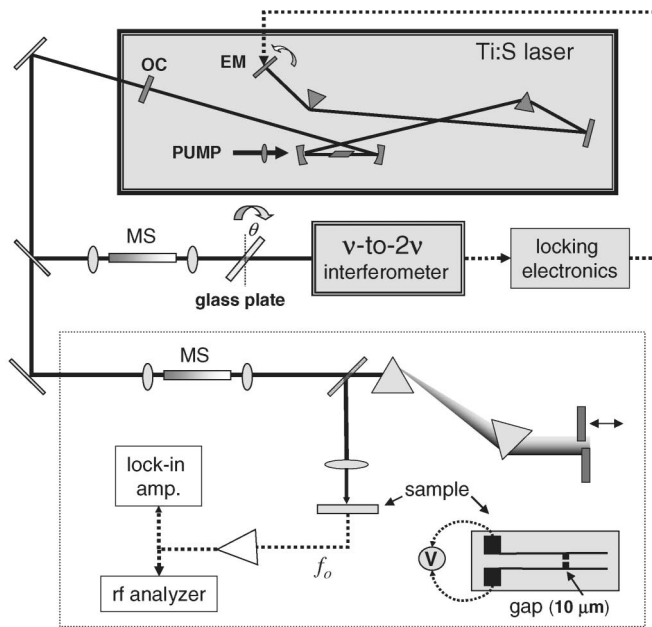


FIG. 2. Experimental setup used for quantum interference control of injected photocurrents in LT-GaAs. The laser offset frequency is measured using a ν -to- 2ν interferometer and stabilized via negative feedback to the laser. Microstructure fiber is used for external broadening of the Ti:S laser spectrum for stabilization and QIC characterization. Because of fiber dispersion, a prism sequence for time delay compensation and a split mirror after the second prism are used for spectral filtering. Light at ν and 2ν are p polarized such that the electric field oscillates transversely along the axis between the two gold electrodes on the LT-GaAs sample (inset).

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to the glass dispersion) that is measured by the ν -to- 2ν interferometer (see Fig. 2). The stabilization loop compensates for the ensuing phase error by adjusting ϕ_0 of the laser output, which produces a corresponding shift in the phase of the measured lock-in signal [Fig. 4(a)]. The measured phase shifts in Fig. 4(a) versus plate rotation are compared with those calculated using the dispersion and thickness of the glass plate [see Fig. 4(b)]. The measured and calculated changes correspond well up until a plate rotation angle of 30° , where the observed discrepancy may be the result of beam misalignment into the ν -to- 2ν interferometer.

In summary, we have demonstrated quantum coherent control using the carrier-envelope phase of a stabilized pulse train from a Ti:S laser. This is implemented in LT-GaAs where it results in directional control of injected photocurrents. Additionally, we have shown that QIC in LT-GaAs is a convenient medium for solid-state detection of changes in the pulse train carrier-envelope phase. Quantum interference in the semiconductor system is

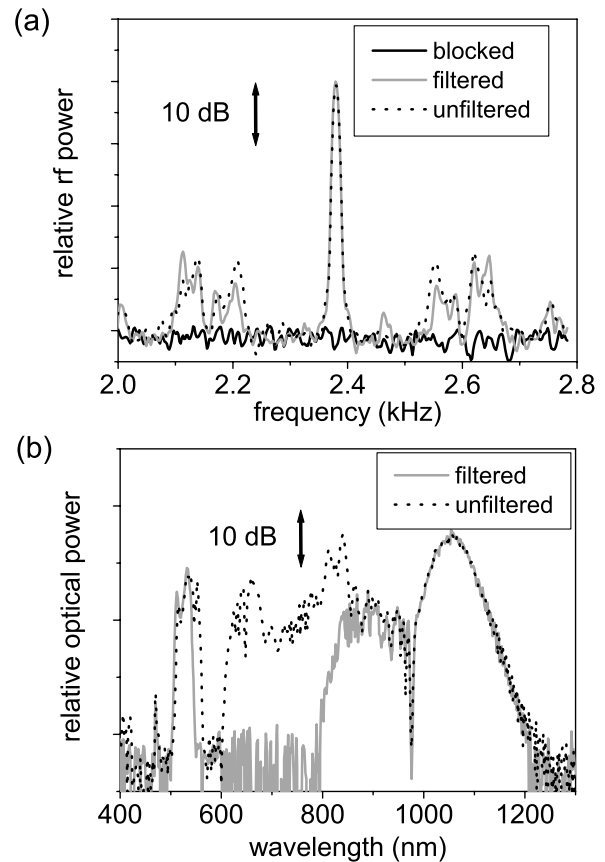


FIG. 3. (a) Spectrum of the measured QIC signal (10 Hz bandwidth, 100 averages). The linewidth (resolution limited) of the measured QIC signal indicates the stability of the laser offset frequency. The dotted and solid gray lines show the QIC signals corresponding to the optical spectra in (b). The solid black line gives the case when the light is blocked. (b) The dip at ~ 500 nm corresponds to unavoidable optical filtering due to the small gap between the split mirrors in Fig. 2.

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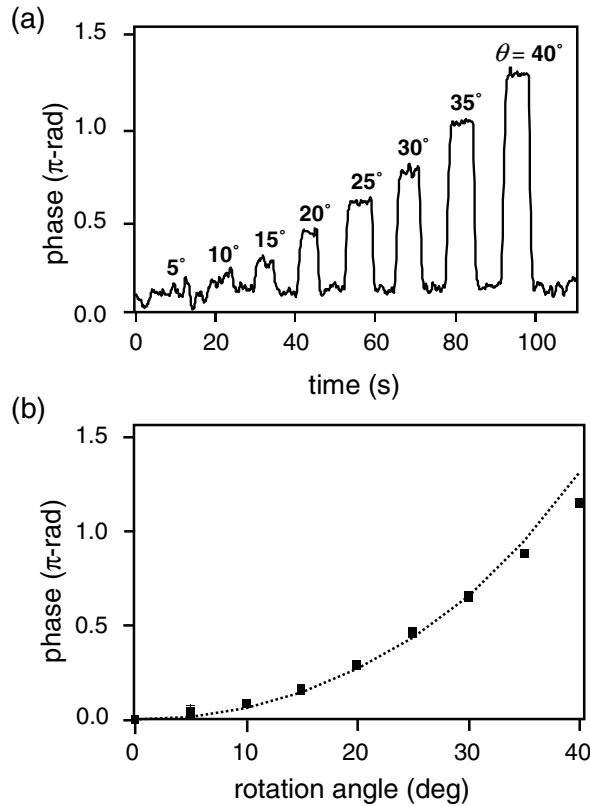


FIG. 4. (a) Phase of the QIC signal as measured relative to the reference used for laser stabilization. The time record of the QIC phase (100 ms time constant) shows the phase jumps associated with rotations of the glass plate from 0° to θ and back again for eight different rotation angles. (b) Comparison between the phase change measured in (a) (squares) and the calculated carrier-envelope phase change (dashed line).

advantageous over more common phase-sensitive schemes in that detection suffers only minimal phase offsets due to dispersion [22]. With increased SNR of the QIC signal, injected photocurrents in semiconductors could be utilized for stabilization of the laser offset frequency. Enhancement of the SNR should be possible through improved engineering of the electrode geometry and noise suppression.

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