Phase-Controlled Currents in Semiconductors

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The direction of emission of photoexcited electrons in semiconductors is controlled by adjusting the relative phase difference between a midinfrared radiation and its second harmonic. This is achieved by using quantum interference of electrons produced with one- and two-photon bound-to-free intersubband transitions in AlGaAs/GaAs quantum well superlattices.

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Controlling the magnitude and the polarity of the current flowing through a semiconductor is essential to modern electronic technology. Normally one adjusts the internal electric field and, in this way, changes the velocity of the carriers or their concentration in the active layers. One can optically control the magnitude of the current via optical pumping [1], for instance, but in most applications, the direction of the electron photoemission cannot be selected optically since, in symmetric structures, positive and negative propagating electronic waves are equally excited by the electromagnetic field. In this Letter, we experimentally show that, by introducing and controlling a polar asymmetry in the optical field, the magnitude and the *direction* of the photocurrent can be determined. The principle of our experiments is to have different optical routes leading to the same final state in the continuum of a semiconductor structure. Then, provided the phase is preserved in each quantum pathway, strong interference effects in the final state wave function are expected just as in the case of Fano resonances [2]. The analogy of a Michelson interferometer is useful. In a Michelson interferometer, the phase difference between two beams determines the direction of the recombined light. In our work, we optically populate the arms of an electron interferometer and the phase difference determines the direction of emission of the photocarriers.

In recent years, phase control (coherent control) has been the subject of considerable attention in many areas of physics. Until now, most laser coherent-control experiments have been carried out in atomic systems [3–6]. For instance, it was shown how five-photon ionization of xenon can be suppressed due to a destructive interference between a three-photon absorption process and a one-photon absorption process, the latter being induced by the third harmonic polarization created in the Xe gas [3,4]. This passive phenomenon was made active when Chen, Yin, and Elliot [5] demonstrated how the total photoionization of mercury due to the simultaneous irradiation of two laser fields can be modulated by controlling the phase difference between the two fields. Unlike mixing between one- and three-photon processes, which leads to modulation of the total cross section of the transition, it was shown on rubidium atoms that the interference between the one- and two-photon transition moments changes the angular distributions of the photoelectrons [6]. A similar experiment studied the control of photoemission from Sb-Cs photocathodes [7]. Since coherent-control effects are based on quantum interference, it should be possible to observe them in other systems. For instance, in physical chemistry, control over the branching ratio between chemically distinct photodissociation products of molecules is widely discussed [8] and the process may be responsible for laser-induced second harmonic emission in glass optical fibers [9] and in semiconductor-microcrystallite-doped glasses [10]. To our knowledge, the idea to test these effects in semiconductors through the photocurrent has been proposed only once [11]. In Ref. [11], the authors suggest preparing a coherent superposition of two donor $|s\rangle$ and $|p\rangle$ states and, from there, to excite a free state using two laser beams tuned at the energy separation between the levels. They predicted that the direction of the photocurrent depends directly on the relative phase between the two beams.

Although coherent control should be an extremely general process, to perform an experiment in semiconductors, it seems advisable to stay close to the atomic physics analogy. N-doped quantum wells (QWs) are very well suited for such an experiment. The highly symmetric ground level of these materials can be used as the initial state for the multiple pathways required for the coherent-control experiments. Because of the large dipole moments between electronic states in the same band, high second [12], and third order [13] intersubband nonlinear susceptibilities have been measured and, recently, the $\chi^{(3)}$ coefficient associated with the two-photon intersubband absorption has been reported in standard QW infrared photoconductors [14]. Here we report that phase-controlled directionality of photocurrents can be achieved on such semiconductor structures by mixing one- and two-photon excitations, just as in the rubidium experiment [6].

The semiconductor structure we studied was grown by molecular beam epitaxy on a (001) semi-insulating GaAs substrate. It consists of 25 $GaAs/Ga_{0.74}Al_{0.26}As$ Si-δ doped ($n_s = 5 \times 10^{11}$ cm^{-2}) QWs with a width $d = 55$ Å and separated by 325 Å. This structure is clad between n^+ doped (1.5×10^{18} cm^{-3}) GaAs layers. Using standard photolithographic techniques 120×120 μm^2 mesas were etched and NiGeAu alloyed contacts were processed onto the n^+ layers. One of the quantum pathways used for the coherent-control experiments is two-photon absorption at 10.6 μm. To preserve the phase during this excitation it is useful to avoid populating any intermediate resonant state. This is why the QW parameters are set so that the energy between the first two subbands (152 meV) is far detuned from the photon energy at 10.6 μm (117 meV). To phase control the direction of the photocurrent without leads, photovoltaic behaviors should also be avoided. Therefore, it is important to minimize any asymmetry in the structure. It is known that the dopant segregation during the growth contributes to the asymmetry of the photoresponse. By placing the Si dopants away from the center of the QWs by 22 Å, the I-V characteristic of our device is almost symmetric and the photovoltaic response is minimized [15]. Finally, a 45° edge facet was processed to couple the optical fields with the intersubband dipoles.

We excite electrons from the ground level of the QWs to a highly energetic free state $|E\rangle$ using two quantum pathways: a two-photon intersubband absorption at 10.6 μm and a single-photon process at 5.3 μm, the latter wavelength having a constant phase relation with the former. Then the resulting photocurrent is recorded versus the phase difference between the two fields.

One way to understand the experiments is to interpret these phenomena in terms of *wave-function interferences*. If one describes the degenerate states in the continuum as even and odd states, 10.6 μm excitation leads to the symmetric state $|E,S\rangle$ and 5.3 μm to the antisymmetric state $|E,A\rangle$ (Fig. 1). These phase-locked excited states interfere and, depending on the phase of each beam ($\varphi_{10.6}$, $\varphi_{5.3}$), the electron is partially or completely described by a positive or negative progressive wave. Then, assuming perfect contacts, the classical photoconductive model leads to the following expression for the current density j:

$$j \approx q \frac{n_s}{L} \frac{2\pi}{\hbar} \mu^{(2)} \mu^{(1)} E_\omega^2 E_{2\omega} D_{1D}^*(E) \tau \times \frac{\hbar k' \sin(2\varphi_{10.6} - \varphi_{5.3})}{m' \sqrt{1 + [(m'k/mk' - mk'/m'k)/2]^2 \sin^2(kd)}}, \quad (1)$$

where $\mu^{(1)}$ and $\mu^{(2)}$ stand for the dipole moments of the linear and quadratic absorption, E_ω and $E_{2\omega}$ are the amplitudes of the fundamental and second harmonic fields projected on the [001] active axis, m and m' are the effective masses in the well and barrier, k and k' represent the longitudinal free electron momentum in the well and the barrier, L is the superperiod of the structure (380 Å), τ is the collision relaxation time of the ballistic electrons, and D_{1D}^* is the one-dimensional density of state. In deriving Eq. (1), we assume that the coherence length of the hot electrons is greater than the size of the QW. Thus, the ballistic electrons can escape from the attractive potential of their original well to give rise to a photocurrent. The coherence lifetime τ limited by the phonon-LO scattering [16] is about 100 fs which gives a 600 Å coherence length. The sin function indicates that, by adjusting the phase difference $\Delta\varphi = 2\varphi_{10.6} - \varphi_{5.3}$, the electrons can be directed to the right or to the left of the quantum well. The expression within the square root describes the energy dependence of the dipole moment $\langle E,S|\partial/\partial z|E,A\rangle$, and, therefore, the strength of the interference. When $k' \sim k$, as it is for the excitation high in the continuum, this expression approaches unity.

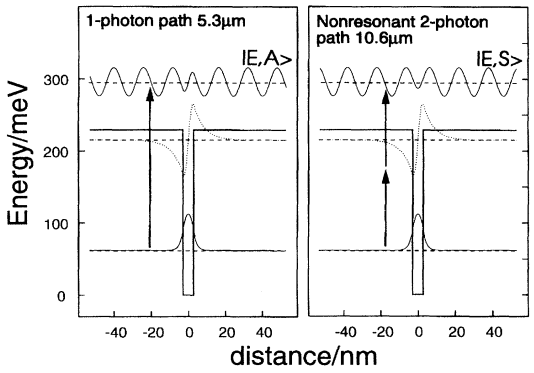

FIG. 1. Energy band diagram of a 55 Å $GaAs/Ga_{0.74}Al_{0.26}As$ QW and wave functions of the states implied in a 5.3 μm single-photon pathway and a 10.6 μm two-photon process. Neither dephasing nor reflections of the electronic waves on the neighbor QWs are considered in this simplified figure.

At this stage, one can see the close analogy with charge oscillations previously reported in coupled QWs [17,18]. However, unlike the experiments of Roskos *et al.* [17] the excited symmetric and antisymmetric states in our experiment are degenerate. This means that to change the interference condition (in a sense to simulate quantum beats) the experimentalist must externally change the phase relation between the symmetric and antisymmetric states.

Although our experiment is easily interpreted as interference of simultaneously excited electronic wave functions of opposite symmetries, another point of view based on *interference of dipole moments* also gives a simple interpretation of the same phenomenon. This arises immediately if one chooses to describe the continuum states as positive $|+k\rangle$ or negative $|-k\rangle$ progressive waves. In that case, the current in the coherent-control experiments depends directly on the rate of transitions $W_{|0\rangle \to |\pm k\rangle}$. From perturbation theory, the probabilities of transitions are related to the interference between the one- and two-photon dipole moments, i.e., $W \propto |\mu^{(1)} E_{2\omega} + \mu^{(2)} E_\omega^2|^2$. They os-

cillate with $\Delta\varphi$ and they are 180° out of phase with each other. Thus, the experiment consists in *controlling the relative cross sections* associated with the $|0\rangle \rightarrow |+k\rangle$ and $|0\rangle \rightarrow |-k\rangle$ transitions.

The experimental configuration to demonstrate coherent control of photocurrents in the multiquantum well structure is shown in Fig. 2. A single-mode hybrid TEA CO_2 laser produces 100 ns pulses at a repetition rate of 2 Hz. These pulses are gently focused on a type I phase matched $AgGaSe_2$ doubling crystal. This crystal is used with an energy conversion efficiency of about 2×10^{-4}. The relative phase between the colinear fundamental (10.6 μm) and second harmonic beams is changed by a dispersive NaCl crystal mounted on a rotating stage. Then the relative amplitude of the fundamental and second harmonic fields is adjusted by a set of filters: CaF_2 windows to attenuate the 10.6 μm beam in discrete steps and a 1 atm SF_6-He cell for a variable 10.6 μm attenuation depending on the SF_6 partial pressure. After this adjustment, the strengths of the two pathways in this "electron interferometer" are approximately equal. In practice, in our experiments, the incident pulse energy at 10.6 μm is ~10 μJ and the 5.3 μm pulse energy is ~50 nJ.

An optical arrangement that includes a nonsaturated nonlinear crystal and handles the fundamental and harmonic radiation together with no dispersive curved optical elements is ideally suited for coherent-control experiments. The phase relationship is maintained and even the intensity profile of the fundamental and harmonic is optimum. The beams are focused onto the detector with a $f/8$ concave mirror. The measured spot size at 10.6 μm is ~50 μm.

Because of the type I phase matching of $AgGaSe_2$, the polarization of the two beams was not optimum to couple to the QW intersubband transitions. We used the polarization sensitivity of the QW detector to select the appropriate polarization component. Usually the resulting coupling efficiency in our configuration was 25% and 2% for the one- and two-photon processes, respectively. The signals from the QW structure and a reference detector (photon drag) are then amplified by 50 Ω input electronics and recorded with a digital scope or with gated integrators for different relative phase delays.

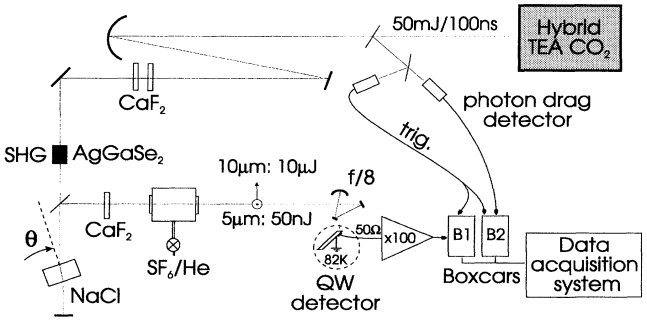

FIG. 2. Experimental setup of the coherent-control experiments. The signal from the QWs is recorded versus the angle of incidence θ on the dispersive NaCl crystal. No external voltage is applied on the QW structure.

Figure 3 shows the QW pulse response under different conditions. The first two oscilloscope traces from the top represent the signal recorded with only one incident wavelength and with the detector at zero bias. Because of some residual photovoltaic behavior, a small background 5.3 μm signal (top trace) is still detected. This response follows the polarization selection rule on intersubband transitions and it can be canceled out with an external electric field of 0.2 kV/cm. At 10.6 μm an opposite polarity pulse is observed. It is composed of two contributions: (1) a positive two-photon QW contribution which is comparable to the 5 μm signal and strongly dependent on the polarization [14] and (2) a negative component which is attributed to single-photon or multiphoton emission from deep levels in the AlGaAs barrier.

The three lower oscilloscope traces in Fig. 3 were taken with both the 10.6 and 5.3 μm beams simultaneously illuminating the detector. They show strong modulation of the photocurrent with the angle of incidence θ of the mid-IR light on the NaCl plate. By rotating the salt crystal by a few tenths of degrees, we are able to invert the direction of emission of the photocarriers. The pulses at $\theta = 12.8°$ and 13.7° are very much like the square of the 10.6 μm pulse shape and they effectively vary quadratically with the 10.6 μm incident (before the doubling crystal) energy. Figure 4 shows the integrated QW response plotted as a function of the relative phase difference between the two beams. From the period of the fringes, the deduced dispersion $n_{2\omega} - n_\omega$ of our NaCl window is 2.54×10^{-2}, which is 7% lower than the data book value. The amplitude of the oscillations varies linearly with the 10.6 μm pulse energy (after the doubling

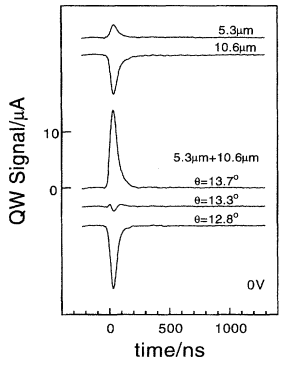

FIG. 3. Oscilloscope traces of the QW pulse response. The residual response with only one beam (the first two curves) at 5.3 and 10.6 μm are background signals for the coherent-control experiment. The three lower traces represent the QW response when the 10.6 and 5.3 μm pulses are both illuminating the sample. From $\theta = 12.8°$ to 13.7° a phase shift of π is imposed.

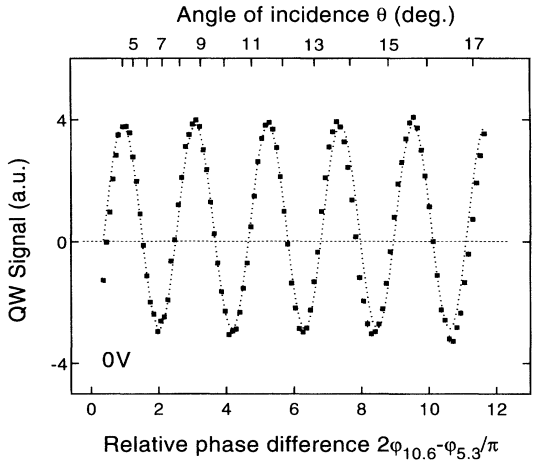

FIG. 4. Integrated QW response versus the angle of incidence. Dashed line: sinusoidal fit.

crystal) and, if the sample is oriented so that 5.3 μm polarize along the plane of the QWs, one observes a very strong reduction of the modulation. However, the 10 μA amplitude of the oscillations is 1 order of magnitude lower than the estimated value given by Eq. (1). A model of ballistic transport in a superlattice environment which includes the influence of the rectifying contacts is required to extract information on the scattering processes from these coherent-control experiments.

Although we have emphasized results for symmetric QWs, on a similar sample (58 Å $GaAs/Ga_{0.7}Al_{0.3}As$) with a *center* Si-δ doping, an equally periodic response was also observed. At zero bias, because of a stronger photovoltaic behavior (internal electric field in the barrier of 4 kV/cm), we modulated the photocurrent by 25%. With an external voltage we significantly increased the contrast of the fringes. However, in these asymmetric samples, we could not invert the direction of the current due to a non-negligible 10.6 μm background signal from deep levels.

In conclusion, our experiments show that coherent control based on quantum interferences can be successfully applied in semiconductor physics. More specifically, by interfering degenerate states in the conduction band of a quantum semiconductor structure, we show that the magnitude and even the direction of the photocurrent is governed by the phase of electromagnetic waves. In general, phase will be just as important a variable as the more traditional optical parameters of intensity or frequency in any process involving at least two phase preserving routes to a same final state. With bulk materials [19], the technique presented here might find many applications in optoelectronics technology.

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