Observation of Coherently Controlled Photocurrent in Unbiased, Bulk GaAs

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We demonstrate room temperature coherent generation and control of a directional photocurrent in bulk GaAs via simultaneous one- and two-photon interband absorption processes using phase-related 1 ps or 175 fs pulses at 0.775 and 1.55 μm. Electrical currents generated in low-temperature-grown (LT) and normal bulk GaAs are collected via gold electrodes. Current densities as high as 3 nA/μm^2 in LT-GaAs are measured for injected carrier densities as low as 10^{14} cm^{-3} and for peak irradiances of 18 MW cm^{-2} (1.550 μm) and 3 kW cm^{-2} (0.775 μm). [S0031-9007(96)02103-5]

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The idea of controlling optical, physical, and chemical processes in matter using the coherence properties of light has long captured the interest of many scientists [1–10]. In 1967 Manykin and Alfanasev [1] proposed absorption control of an atomic medium by phase control of two or more incident beams. In essence, interference between different quantum mechanical pathways connecting the same initial and final states controls the transfer of electrons. Although ideas related to coherent control lay virtually dormant for many years, recently they have been revived and demonstrated in a variety of atomic and molecular systems [2,5]. For a discrete initial state and a final state in the continuum, it has been suggested [3] that directional ionization can occur, and indeed this has been observed in atomic Hg [5] using 554 and 185 nm beams and in *n*-doped AlGaAs/GaAs quantum well superlattices at 82 K using 10.6 and 5.3 μm pulses [6]; directional ionization of defects has also been proposed to lead to space-charge fields and enhanced second-harmonic generation in optical fibers [7]. Although the results of a photoemission experiment have been interpreted in terms of coherent control [4], the possibility of such coherent effects in solids or polyatomic molecules where the final states belong to a continuum manifold has been considered remote because of assumed rapid dephasing phenomena. For photochemistry involving polyatomic molecules, there may also be difficulties in achieving state selectivity for a particular reaction pathway, e.g., breaking a predefined bond. However, for generation of directional photocurrents in a semiconductor via band-to-band transitions, state selectivity is not a consideration since all optically coupled states can contribute to current flow; control can therefore be achieved via a spectrum of wavelengths. Furthermore the use of bulk material permits other degrees of freedom, such as optical polarization to control currents generated along a particular direction rather than, e.g., a quantum well growth direction. Therefore, a demonstration of coherent control of photocurrent magnitude and directionality in bulk, unbiased semiconductors via interband transitions is appealing both from a technological and a scientific point of view. Earlier [8,9] we showed in principle that control of current should be observable both in centrosymmetric and noncentrosymmetric semiconductors even though carrier dephasing times may be $\sim$100 fs; such effects have also been considered by Khurgin [10] in the context of terahertz generation. Here we report the observation of this phenomenon at room temperature in low-temperature-grown and normal GaAs using femtosecond and picosecond laser pulses. Generation and control of electrical currents by such short pulses opens up new applications in optoelectronics and terahertz wave generation.

When a bulk, intrinsic semiconductor with direct band gap E_g is illuminated with two monochromatic beams of frequency ω and 2ω, the two- and one-photon interband transitions lead to an electron (e) and hole (h) current density which satisfies the equation [9]

$$\dot{\mathbf{J}}_{e,h} = \hat{\eta}_{e,h} : \mathbf{E}^{\omega}\mathbf{E}^{\omega}\mathbf{E}^{-2\omega} + \text{c.c.} - \mathbf{J}_{e,h}/\tau_{e,h}\,, \quad (1)$$

where $\hat{\eta}_{e,h}$ are fourth rank tensors with purely imaginary elements, $\mathbf{E}^{\omega}$ and $\mathbf{E}^{-2\omega}$ are the (complex) field amplitudes associated with the two beams, and $\tau_{e,h}$ are phenomenological current relaxation times. The generation of a macroscopic current can be understood simply as a breakdown in time-reversal symmetry in the presence of a third-power field. Alternatively it can be seen as resulting from an anisotropic, polar distribution of carriers in k space because of interference of single and two-photon absorption processes coupling the same semiconductor Bloch states. The $\tau_{e,h}$, which for free electrons and holes have values of $\sim$100 fs, require that pulsed, high intensity radiation be used to observe macroscopic currents. For GaAs, η_{xxxx} and $\eta_{xyxy} = \eta_{xxyy}$ are the largest tensor elements for both electrons and holes (with the electron terms dominating), whereas η_{xyyx} is much smaller for $\lambda > 775$ nm; cubic symmetry permits exchange of x, y, and z to define other tensor elements. In our experiments we take advantage of the η_{xxxx} tensor element, so that the total current density generation rate can be written as

$$\dot{J} = 2|\eta_{xxxx}|\,(E_x^{\omega})^2 E_x^{2\omega} \sin(2\phi_{\omega} - \phi_{2\omega}) - \frac{J}{\tau}\,, \quad (2)$$

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where ϕ_ω and $\phi_{2\omega}$ are the phases of the x components of the field amplitudes and τ is an effective relaxation time which reflects carrier dephasing and radiation damping; the appearance of the phase term indicates that both the current amplitude and directionality (along the x axis in this case) can be controlled through the phase difference of the two beams.

The experimental arrangement is illustrated in Fig. 1. Two different optical sources, permitting a wide range of optical intensities and pulse repetition rate, were used. The first is an optical parametric generator (OPG) (Coherent model 9400) which provides 175 fs pulses at 250 kHz, with average power up to 30 mW and tunable between 1.1 and 2.9 μm. This source was used near 1.55 μm for which the pulse-time-bandwidth product is 0.65, whereas 0.44 is expected for a Gaussian temporal profile. The other source is a KTP-based optical parametric oscillator (OPO) similar to a sub-100 fs version [11]. It produces 1 ps pulses (time-bandwidth product of 0.48) tunable near 1.55 μm with average power up to 150 mW at 82 MHz. Approximately 20 μW of second-harmonic average power could be generated from either source in a 1 mm thick BBO (β-barium borate) crystal using type I phase matching; the 1.55 and 0.775 μm beams will be referred to as the ω and 2ω beams. A 1 mm thick BK7 glass window with $\lambda/5$ surface flatness can be rotated through an angle θ ($= 0°$ for normal incidence) to vary the relative phase of the beams. The window was used in double pass configuration to minimize beam lateral shift. A 0.75 mm thick quartz half-wave plate at ω (full-wave plate at 2ω) converts the initially orthogonal polarized beams to the same linear polarization state. Thereafter the pulses are focused at normal incidence to a 40–75 μm diameter spot size onto the GaAs sample using a $f = 2.5$ cm gold, spherical mirror. The angle between incident and reflected beams from focusing mirrors are minimized to produce minimal beam astigmatism. The total time delay due to group velocity mismatch between the two pulses before the sample is estimated to be <110 fs.

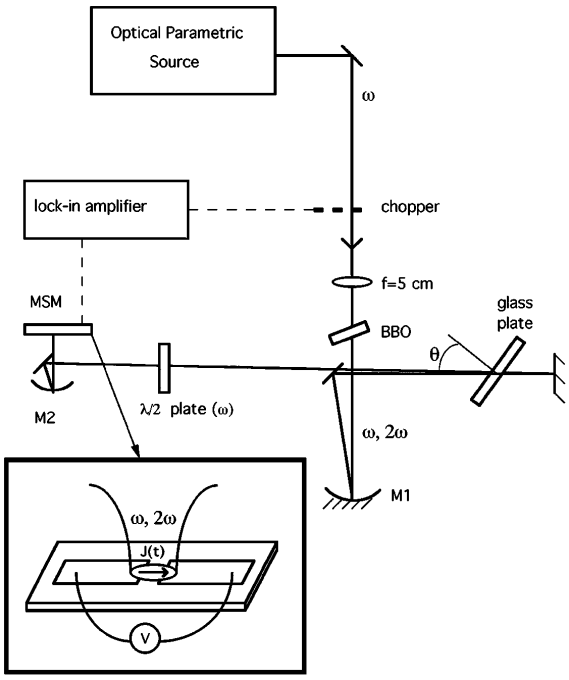

FIG. 1. Schematic diagram of the experimental setup; optical parametric source is an OPO or OPG as described in text; BBO: β-barium borate frequency doubling crystal; M1 and M2 are curved gold mirrors with focal lengths of 10 and 2.5 cm, respectively.

For current generation and detection we use an electrically unbiased, planar, metal-semiconductor-metal (MSM) structure. One of the semiconductors is a 1 μm epilayer of undoped, annealed (10 min at 600 °C) GaAs grown at low temperature (200 °C) on a GaAs substrate with (001) orientation. The epilayer, hereafter referred to as LT-GaAs (low-temperature-grown GaAs), has a 2% excess arsenic concentration which produces a high resistivity [12] ($>10^6$ Ω cm) and an electron, hole trapping time of $\sim$1 ps [13]. The fast trapping time prevents charge accumulation effects, allowing a low carrier density regime to be accessed with the high repetition rate OPO. The other substrate is normally grown, undoped, 350 μm thick, high mobility (100) GaAs with resistivity of 10^8 Ω cm; the electron-hole recombination time in such material is typically >10 ns for carrier densities below 10^{18} cm^{-3}. Several pairs of 200 μm $\times$ 250 μm gold electrodes of thickness 170 nm and with gaps from 5 to 50 μm have been deposited on the samples using photolithography. The electrodes are placed so as to have the gap direction parallel to the (100) crystallographic axis in accordance with Eq. (2); no particular care was taken to produce Ohmic contacts in these initial experiments.

With the OPG the peak irradiance of the ω and 2ω beams is 700 and 14 MW cm^{-2}, respectively. Such pulses independently create surface peak carrier densities of $\sim 10^{15}$ and $\sim 10^{17}$ cm^{-3}, and steady-state densities $<10^{14}$ cm^{-3} in normal GaAs. The OPO produces peak irradiances of 18 MW cm^{-2} (ω) and 3 kW cm^{-2} (2ω) and peak carrier densities of $\sim 10^{13}$ and $\sim 10^{14}$ cm^{-3}. For these estimates, we have assumed that the linear and two-photon absorption coefficients of normal GaAs are 2×10^4 cm^{-1} [14] and 5 cm/GW [15]; the optical properties of LT-GaAs do not differ significantly from those of GaAs for the wavelengths considered here [16]. The MSM position is adjusted to align the middle of the gap with the beam's focal point. Lock-in amplification and detection were used to determine the accumulated charge on the electrodes through measurement of the steady-state voltage (V) and a knowledge of the circuit parameters.

Figure 2(a) shows the signal from an unbiased, 25 μm gap LT-GaAs MSM as a function of θ, when each beam is independently incident on the sample and when both beams are present. In all cases the beam polarization is across the gap [along the (100) direction]. When the beams are separately present a small induced voltage is

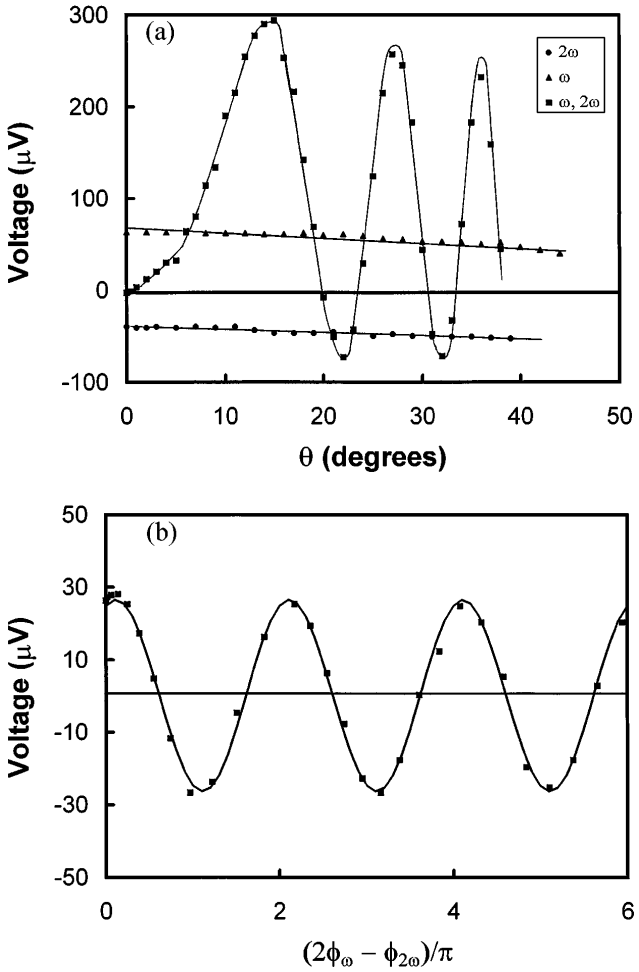

FIG. 2. (a) Induced voltage on a 25 μm gap LT-GaAs MSM detector in the presence of the ω beam (triangles), the 2ω beam (circles), and both beams (squares) from the OPO as a function of glass plate rotation angle θ; the voltage is adjusted to read zero volts when the two beams are simultaneously present on the sample with $\theta = 0$. (b) Induced coherently controlled current signature as a function of $\Delta\phi$ for a 5 μm gap MSM; the solid curve is the best fit for a sine function.

detected which is almost independent of θ. The origin of this offset signal is not clear; Dupont *et al.* [6], in their multiquantum well experiments, observed a similar effect. Schottky barrier phenomena associated with non-Ohmic contacts could create internal fields which can displace photogenerated carriers. In our case the offset voltage level depends on the focal spot location on the MSM, and can be minimized by positioning the spot between the electrodes. From an experimental standpoint, therefore, the only consequence is that the offset voltage requires the beams to be positionally stable when the phase difference between two beams is adjusted.

When both beams are incident on the sample, a modulation in the voltage is observed with variation of θ. This modulated signal is observed for all MSM gaps between 5 and 50 μm. As Fig. 2(a) shows, the modulation is not sinusoidal since $\Delta\phi = 2\phi_\omega - \phi_{2\omega}$ varies nearly quadratically with θ. Figure 2(b) shows the modulated component of the signal (background subtracted) for the 5 μm gap MSM as a function of $\Delta\phi$, which has been determined from θ and the window refractive indices of 1.5013 and 1.5118 for ω and 2ω, respectively [17]. As expected, no modulation is observed when the beams are orthogonally polarized, since current generation is based on the weak η_{xyyx} element. Nor is signal observed when both beams are polarized perpendicular to the electrode-gap direction (current generation parallel to the electrodes). The solid curve shown in Fig. 2(b) represents the best fit to the data for a function of the form $A\sin(\Delta\phi + \phi_0)$ where A and ϕ_0 are fitting parameters. The phase offset ϕ_0, which depends on the dispersion of air, lenses, sample, etc., can be determined using cascaded frequency doubling [18]. Nonetheless, the data clearly have the expected functional dependence on $\Delta\phi$.

Phase-related current control could not be achieved in GaAs MSM samples with the OPO, possibly since the recombination time for the low density carriers is much longer than the pulse period. In such case the steady-state carrier density is larger than the injected carrier density per pulse. The fact that we observe a large ($>$10 mV) θ-independent signal when the two beams are used is consistent with this interpretation. On the other hand, use of the OPG source yields measurable coherently controlled current in both samples, with the LT-GaAs producing a slightly smaller response for the same irradiance. Figure 3 shows the modulated component of the current signal for a 10 μm gap MSM on normal GaAs; the solid curve is a sine function based on BK7 refractive indices and the window thickness. The data are noisier than those achieved with the OPO. This may reflect aspects of the behavior of the non-Ohmic contacts at higher carrier densities or the poorer beam quality (particularly phase uniformity) of the OPG source; this currently is under investigation. From a knowledge of the circuit elements we estimate the peak current density (averaged over the optical absorption depth of the 2ω pulse) during OPG excitation to be 500 nA/μm^2, which is over 150 times larger than the corresponding value (3 nA/μm^2) attained in LT-GaAs using the OPO, although $(E^\omega)^2E^{-2\omega}$ in this case is $\gg$150 times larger.

A preliminary indication of the dependence of the generated current in LT-GaAs on the fundamental beam irradiance was made by varying the average power P^ω before the doubling crystal. From Eq. (1) and the second-harmonic conversion process, $\dot{J} \propto |E^\omega|^4 \propto (P^\omega)^2$. For $35 < P^\omega < 150$ mW we obtained a modulation amplitude $V \propto (P^\omega)^{1.7\pm0.2}$. The deviation from quadratic behavior is not unexpected since measuring the steady-state voltage across the MSM is not equivalent to measuring $\dot{J}$ directly. Indeed, $\dot{J}$ can only be obtained after accounting for the time dependence of the current evolution which is influenced not only by the circuit parameters but also

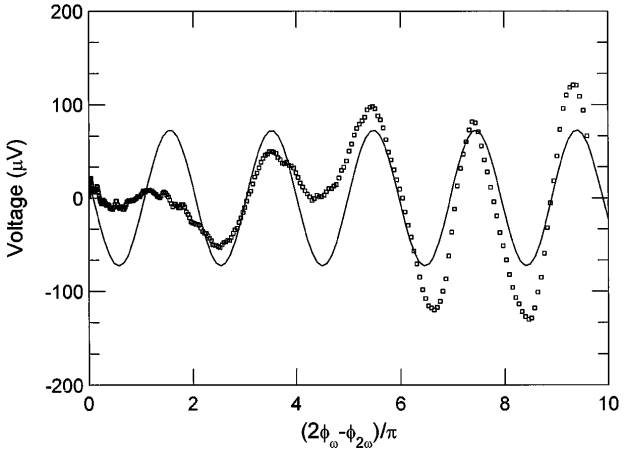

FIG. 3. Induced coherently controlled current signature as a function of $\Delta\phi$ on a 10 μm gap MSM on normal GaAs using the OPG. The solid curve is the best fit for a constant amplitude sine function.

by effects such as carrier dynamics and charge collection efficiency. Nonetheless, the preliminary results, along with the observed polarization effects rule out higher order nonlinear processes which could possibly lead to current generation, such as simultaneous first order, optically induced carrier generation and third order rectification (producing a dc electric field). The quantitative scaling behavior of the current generation rate with optical power over a much larger power range is under investigation.

The peak value of J obtained with the OPO, 3 nA/μm^2, is approximately an order of magnitude lower than the value predicted by theory [9] for the peak irradiances quoted above and assuming $\tau_{e,h} = 100$ fs. The value of $\tau_{e,h}$ may, in fact, be smaller than this in LT-GaAs because of the presence of arsenic defects. Carrier-carrier scattering does not dominate carrier dephasing at the low densities produced by the OPO [19]; it may, however, play a role at higher carrier densities as generated by the shorter pulse OPG, and carrier-carrier scattering and terahertz generation may play a role in determining the magnitude of the peak current density.

Although the density of photoexcited carriers in these experiments can be quite low, the injection speeds can be high. For example, the effective speed of the electron distribution is $\dot{J}/e\dot{n}$ where $\dot{n}$ is the total carrier generation rate; from our data we calculate a peak speed of 10^3 ms^{-1} in LT-GaAs. Note that this value is attained as a *drift* speed in a thermal distribution of carriers for an electron mobility [20] of $\sim 10^{-1}$ m^2 V^{-1} s^{-1} and a dc electrical bias of 10^4 V m^{-1}. Hence, although the carrier densities quoted here are much lower than those typically encountered, the much higher effective speed of the electrons (and, by analogy, holes) make up for that. Note that the maximum speed occurs when the rates of single and two-photon absorption are equal. Our experiments were not conducted under such optimum conditions.

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