Coherent Control of Photocurrent Generation in Bulk Semiconductors

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(Received 2 October 1995)

We show theoretically that interband transitions in a bulk semiconductor via coherent one- and two-photon absorption leads to the formation of an electrical current whose direction is controlled by the relative phase of the beams. The phenomenon can occur in centrosymmetric and noncentrosymmetric materials; easily measurable currents are predicted for GaAs under realistic experimental conditions.

PACS numbers: 72.40.+w, 42.50.Ar, 42.65.–k

The understanding and control of electrical current in a semiconductor is of obvious fundamental and technological importance. In this Letter we argue that it should be possible to inject current in a bulk, undoped semiconductor and control its direction by simply adjusting the relative phase of two beams that are optically generating carriers across the gap.

Phenomena responsible for the optical injection of current have been studied for many years. Photovoltaic effects rely on a lack of inversion symmetry to allow injected carriers to form a current [1]. More recently, work in atomic physics [2,3] has shown that current injection is possible even for materials with a center of inversion symmetry, without the aid of asymmetric scattering and interaction effects. The simplest example is the ionization of an atom by coherent optical beams at frequency ω (leading to two-photon ionization) and 2ω (leading to one-photon ionization) [2]. Since the one- and two-photon processes connect the initial state to final states that are degenerate but of different parity, adjusting the relative phase of the two beams alters the combination of such final states selected; in general the selected state will not be of definite parity, and a current can appear. In a Fermi's golden rule calculation, this results from an interference of the probability amplitudes for one- and two-photon ionization [4].

In solids, phenomenological arguments clearly show that such an injected current is also allowed [5,6]; a two-beam photoionization experiment has been interpreted in terms of such injected currents [7]; it has been suggested that second-harmonic generation in optical fibers is due to such currents injected from defects [8]; the injection of currents from midgap impurities in semiconductors has been calculated [9]; and "atomiclike" coherent current generation from quantum wells has been observed [10]. But it does not seem to be appreciated that substantial, coherently controlled current injection should be possible in a bulk, undoped semiconductor—even one with a center of inversion symmetry—by exciting it across the band gap. In this Letter we consider subjecting a semiconductor to two coherent beams with frequencies ω and 2ω satisfying $E_g/2 < \hbar\omega < E_g$, where E_g is the fundamental band gap; we calculate the size of the injected current for GaAs, and find that it should be observable under reasonable experimental conditions [11]. While a complete calculation would treat both the injection and subsequent transport at a fully quantum mechanical level, in a preliminary treatment here we calculate the injection rate using Fermi's golden rule and model the subsequent transport with a hydrodynamic model of the electron-hole plasma.

To calculate the injection, in an independent particle approximation the important states are the ground (initial) state $|0\rangle$ and states of the form $|cv,\mathbf{k}\rangle = a^\dagger_{c\mathbf{k}} b^\dagger_{v\mathbf{k}}|0\rangle$, where $a^\dagger_{c\mathbf{k}}$ ($b^\dagger_{v\mathbf{k}}$) creates an electron (hole) at wave vector $\mathbf{k}$ in conduction (valence) band c (v). In the presence of a classical electromagnetic field, we look for a ket of the form $|\Psi(t)\rangle = c_0(t)|0\rangle + c_{cv,\mathbf{k}}(t)|cv,\mathbf{k}\rangle$, where a summation over c, v, and $\mathbf{k}$ is implied. The coefficients $c_0(t)$ and $c_{cv,\mathbf{k}}(t)$ are determined from perturbation theory; we use the usual minimal coupling Hamiltonian in the long wavelength limit. For intrinsic semiconductors the injection rates for electron and hole *densities*, $\dot{n}_e(t)$ and $\dot{n}_h(t)$, respectively, are equal. Considering the interaction to be on for a time Δt,

$$\dot{n}_h = \dot{n}_e = \dot{\nu}^I = \frac{1}{V\Delta t}\langle\Psi(\Delta t)|\sum_{c,\mathbf{k}} a^\dagger_{c\mathbf{k}} a_{c\mathbf{k}}|\Psi(\Delta t)\rangle, \quad (1)$$

where V is the normalization volume of the sample. The usual Fermi's golden rule approximations then yield a $\dot{\nu}^I$ that can be written as the sum of one- and two-photon terms, $\dot{\nu}^I = \dot{n}_i^{(1)} + \dot{n}_i^{(2)}$, where i refers to either electrons or holes. We find

$$\dot{n}_i^{(1)} = \hat{\xi}_1(2\omega) : \mathbf{E}(-2\omega)\mathbf{E}(2\omega), \quad (2)$$

$$\dot{n}_i^{(2)} = \hat{\xi}_2(\omega) : \mathbf{E}(-\omega)\mathbf{E}(-\omega)\mathbf{E}(\omega)\mathbf{E}(\omega), \quad (3)$$

where $\mathbf{E}(\omega)$ and $\mathbf{E}(2\omega)$ are the electric field amplitudes at the indicated frequencies; we have neglected a set of correction terms that arise in the absence of a center of inversion. The tensors $\hat{\xi}_1$ and $\hat{\xi}_2$ are given by

$$\hat{\xi}_1(2\omega) = \frac{2\pi e^2}{\hbar^2}\sum_{c,v}\int \frac{d\mathbf{k}}{4\pi^3}\,\delta(\omega_{cv}(\mathbf{k}) - 2\omega) \times \frac{\mathbf{v}_{vc}(\mathbf{k})\mathbf{v}_{cv}(\mathbf{k})}{\omega_{cv}(\mathbf{k})}, \quad (4)$$

$$\hat{\xi}_2(\omega) = \frac{32\pi e^4}{\hbar^4} \sum_{c,v,\alpha,\alpha'} \int \frac{d\mathbf{k}}{4\pi^3}\, \delta(\omega_{cv}(\mathbf{k}) - 2\omega) \times \frac{\{\mathbf{v}_{\alpha v}(\mathbf{k})\, \mathbf{v}_{c\alpha}(\mathbf{k})\}\{\mathbf{v}_{\alpha' v}(\mathbf{k})\mathbf{v}_{c\alpha'}(\mathbf{k})\}}{\omega_{cv}^4(\mathbf{k})[\overline{\omega}_{cv}(\mathbf{k}) - \omega_\alpha(\mathbf{k})][\overline{\omega}_{cv}(\mathbf{k}) - \omega_{\alpha'}(\mathbf{k})]}, \quad (5)$$

where $\mathbf{v}_{\alpha\alpha'}(\mathbf{k})$ denote the matrix elements of the velocity operator between the indicated bands at wave vector $\mathbf{k}$, and the curly brackets $\{\}$ denote a symmetrized form with respect to Cartesian components; Greek indices such as α and α' range over both conduction and valence bands [dispersion relations $\omega_c(\mathbf{k})$ and $\omega_v(\mathbf{k})$], $\omega_{cv}(\mathbf{k}) \equiv \omega_c(\mathbf{k}) - \omega_v(\mathbf{k})$ and $\bar{\omega}_{cv}(\mathbf{k}) \equiv [\omega_c(\mathbf{k}) + \omega_v(\mathbf{k})]/2$. The tensor $\hat{\xi}_1(2\omega)$ is related to the linear absorption coefficient at 2ω; $\hat{\xi}_1(2\omega) = 2\epsilon_0 \mathrm{Im}[\hat{\epsilon}(2\omega)]/\hbar$, where $\hat{\epsilon}(2\omega)$ is the relative dielectric tensor at frequency 2ω. A similar relation holds between $\hat{\xi}_2(\omega)$ and the imaginary part of the nonlinear susceptibility $\hat{\chi}^{(3)}$ describing two-photon absorption.

Figures 1 and 2 show the calculated tensor components for bulk GaAs. We employ a parabolic band approximation (PBA), considering the momentum matrix elements to be $\mathbf{k}$ independent [12]. We also use a band structure from a self-consistent calculation using the full-potential linear augmented plane wave method within the local density approximation (LDA) [13]; self-energy corrections are included at the level of the "scissors" approximation, which corrects for the LDA band gap and necessitates a corresponding modification of the velocity matrix elements [14], and spin-orbit effects are included. Since LDA calculations give inaccurate effective masses near the gap, the PBA calculation based on experimentally determined effective masses should be a better estimate there; further from the gap, on the other hand, the full band structure calculation should be more reliable. To aid in the comparison of these two we also present in Fig. 1 the results of a PBA calculation *using the LDA effective masses;* comparing with the LDA calculation yields a sense of where the PBA assumptions break down. The PBA curves in Fig. 1 exhibit the familiar dependence on the approximate density of states $D(\omega) \sim \sqrt{\hbar\omega - E_g}$; for the zinc-blende structure $\hat{\xi}_1$ has only a diagonal component. In Fig. 2 we present results for the independent components of $\hat{\xi}_2$. The parabolic band approximation gives $\xi_2^{xyyx} = \xi_2^{xxxx}/2$, proportional to $D^3(\omega)$, and $\xi_2^{xxyy} = 0$; note that this holds approximately in the LDA calculation. Nevertheless, we see from Fig. 2 that there is a significant discrepancy between the results of the PBA and the LDA. This is due primarily to the incorrect PBA assumption of $\mathbf{k}$-independent matrix elements $\mathbf{v}_{cv}$.

The distribution of injected carriers in $\mathbf{k}$ space is asymmetric, with $|c_{cv,\mathbf{k}}|^2 \neq |c_{cv,-\mathbf{k}}|^2$. This follows from the form of $|c_{cv,\mathbf{k}}|^2$, which is the sum of terms $\hat{A} : \mathbf{E}(2\omega)\mathbf{E}(-2\omega)$, $\hat{B} : \mathbf{E}(\omega)\mathbf{E}(\omega)\mathbf{E}(-\omega)\mathbf{E}(-\omega)$, and $\hat{C}\mathbf{v}_{cc}(\mathbf{k}) : \mathbf{E}(-2\omega)\mathbf{E}(\omega)\mathbf{E}(\omega)$, where tensors $\hat{A}, \hat{B}$, and $\hat{C}$ are even in $\mathbf{k}$. It is the last of these terms that is odd in $\mathbf{k}$ and leads to the asymmetry of $|c_{cv,\mathbf{k}}|^2$, resulting in a net current density injection rate $\dot{\mathbf{J}}$. To calculate this rate we need only use Eq. (1) with the electron density operator replaced by the current density operator; however, we calculate the injected electron current density ($\dot{\mathbf{J}}_e^I$) and the hole current density ($\dot{\mathbf{J}}_h^I$) separately. The total current density injection rate is then $\dot{\mathbf{J}}^I = \dot{\mathbf{J}}_e^I + \dot{\mathbf{J}}_h^I$, where $\dot{\mathbf{J}}_{e(h)}^I = \hat{\eta}_{e(h)}(2\omega) : \mathbf{E}(-\omega)\mathbf{E}(-\omega)\mathbf{E}(2\omega) + \text{c.c.}$,

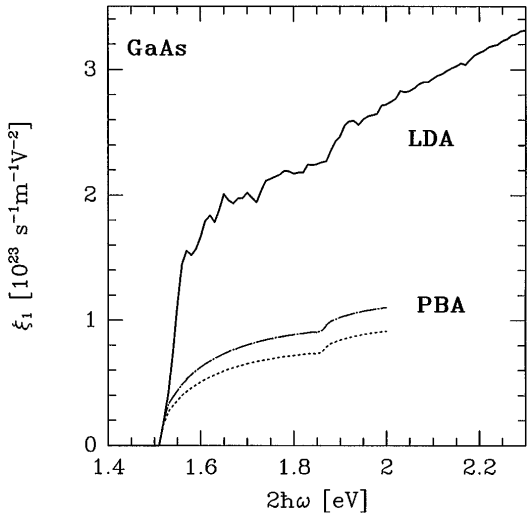

FIG. 1. Plot of the one-photon injection coefficient $\hat{\xi}_1$ as a function of $(2\hbar\omega)$. The solid line represents the results from the LDA and the other two curves refer to those of the PBA: dotted line—experimental effective masses, and dash-dotted line—LDA effective masses. The jaggedness in the LDA results is in part due to numerical artifacts; a bin size of 0.1 eV was used in all LDA calculations in this Letter.

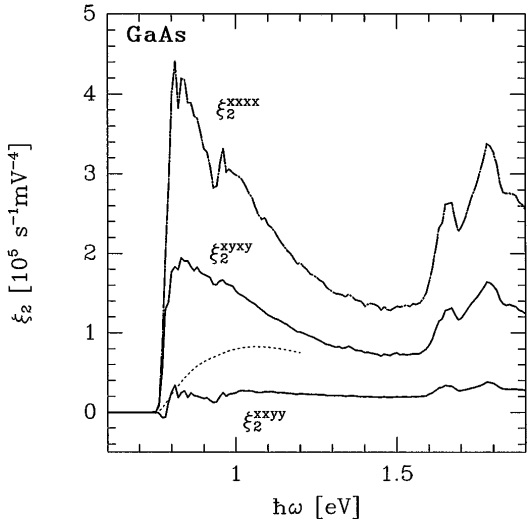

FIG. 2. Tensor components of the two-photon injection coefficient $\hat{\xi}_2$ as a function of the fundamental beam energy $(\hbar\omega)$. For comparison, PBA results for $\hat{\xi}_2^{xxxx}$ using experimental effective masses are represented by the dotted line.

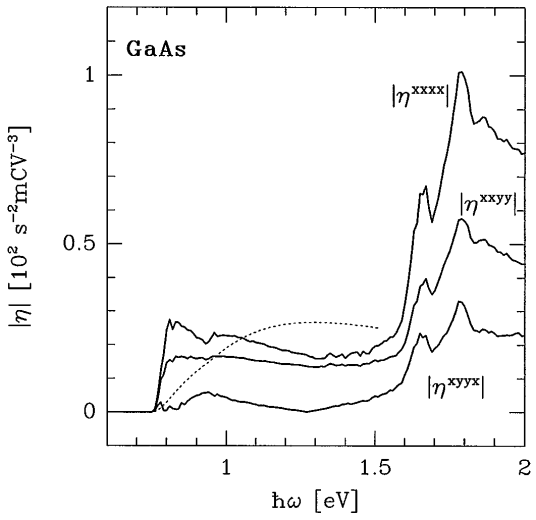

FIG. 3. Tensor components of the total current generation tensor $\hat{\eta}$ as a function of the fundamental beam energy ($\hbar\omega$). PBA results for $\hat{\eta}^{xxxx}$ using experimental effective masses are given by the dotted line.

and the tensors $\hat{\eta}_{e(h)}$ are given by

$$\hat{\eta}_{e(h)}(2\omega) = +(-)i\frac{8\pi e^4}{\hbar^3}\sum_{c,v,\alpha}\int\frac{d\mathbf{k}}{4\pi^3}\,\delta(\omega_{cv}(\mathbf{k})-2\omega) \times\frac{\mathbf{v}_{cc(vv)}(\mathbf{k})\{\mathbf{v}_{v\alpha}(\mathbf{k})\mathbf{v}_{\alpha c}(\mathbf{k})\}\mathbf{v}_{cv}(\mathbf{k})}{\omega_{cv}^3(\mathbf{k})[\omega_\alpha(\mathbf{k})-\overline{\omega}_{cv}(\mathbf{k})]}, \tag{6}$$

where we have again neglected correction terms that can arise in the absence of a center of inversion. Since $\mathbf{E}(-\omega)=\mathbf{E}^*(\omega)$ and $\mathbf{E}(2\omega)$ are complex amplitudes, the resulting $\dot{\mathbf{J}}^I_{e(h)}$ are clearly sensitive to the relative phase of the two beams. Numerical results for the total $\hat{\eta}=\hat{\eta}_e+\hat{\eta}_h$ are plotted in Fig. 3. Parabolic band calculations give an approximated energy dependence similar to those of $\hat{\xi}_2$: $\eta^{xxyy}=\eta^{xxxx}/2$, proportional to $D^3(\omega)$, and $\eta^{xyyx}=0$.

To describe the formation of a directional current in the presence of the inevitable scattering and recombination processes, we now use the calculated $\dot{\nu}^I$ and $\dot{\mathbf{J}}^I_{e,h}$ as source terms in the hydrodynamic equations governing the evolution of the electron (n) and hole (p) densities, the electron ($\mathbf{J}_e$) and hole ($\mathbf{J}_h$) current densities, and the electric field $\mathbf{E}$ inside the sample. Denoting by n_0, p_0, and $\mathbf{E}_0$ the indicated dark values, we introduce dark electron and hole conductivities σ_e and σ_h and obtain linearized equations for the deviations of the fields (denoted by "tildes") from their dark values,

$$\frac{\partial}{\partial t}\tilde{n}-\frac{1}{e}\frac{\partial}{\partial z}\tilde{J}_e+\frac{1}{\tau}\tilde{n}=\dot{\nu}^I, \tag{7}$$

$$\frac{\partial}{\partial t}\tilde{J}_e-\frac{e\mu_e}{\tau_e}E_0\tilde{n}+\frac{1}{\tau_e}\tilde{J}_e-\frac{eD_e}{\tau_e}\frac{\partial}{\partial z}\tilde{n}=\frac{\sigma_e}{\tau_e}\tilde{E}+\dot{J}^I_e,$$

with similar equations for $\tilde{p}$ and $\tilde{J}_h$, together with the Poisson equation. Here we have assumed that the current injection is in the $\hat{z}$ direction, perpendicular to capacitor plates which bound the material; $\tau_{e(h)}$ and $D_{e(h)}$ denote the electron (hole) scattering time and diffusion coefficient. The solution of these equations depends on the initial and boundary conditions, which reflect the experimental situation. For simplicity we assume zero bias ($\mathbf{E}_0=0$) so that any current injection is due to the coherent interaction, which we consider to be homogeneously distributed along the active region. Under these conditions we consider a capacitor filled with bulk GaAs and illuminated laterally by laser pulses of frequencies ω and 2ω and duration 2 ns. As boundary conditions we consider the time derivative of the surface charge on the plates to be equal to the current density accumulated there. In this geometry we obtain a homogeneous current and electric field inside the capacitor, and a voltage accumulating across the capacitor. In Fig. 4 we plot the calculated current within a capacitor assuming typical experimental parameters; we have taken $\hbar\omega=1$ eV and assume the GaAs sample is sufficiently thin that the relative phase of the two beams can be considered uniform. Although the calculation is admittedly simplistic, the results indicate an effect that should be easily observable; other experimental geometries also lead to the prediction of observable signals. For pulses in the fs regime, where higher beam intensities can be achieved, it is necessary to consider loss of plasma energy due to electromagnetic radiation.

Although we have used GaAs for our sample calculations, we stress that the effect does not vanish if a center of inversion is present in the crystal; indeed, there are yet further terms that arise in the absence of a center of

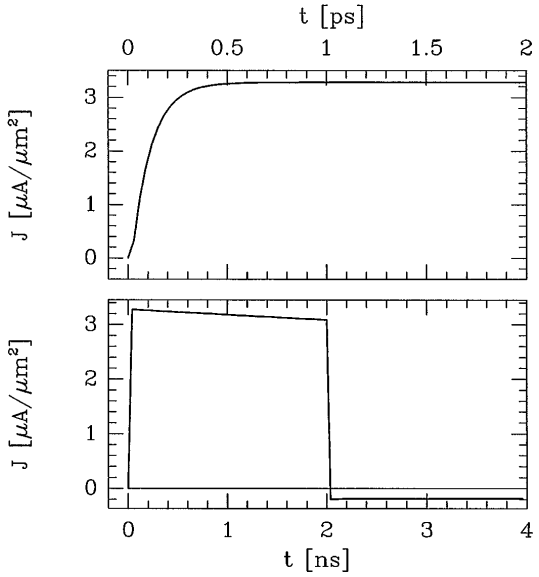

FIG. 4. Plot of the output current density $\mathbf{J}$ induced in a metal capacitor by a 2 ns laser pulse. Very early time behavior is resolved in the upper curve; the lower curve shows the current variation on the time scale of the laser pulse. We adopt the following values: $\tau_e=100$ fs, $\tau_h=50$ fs, $\tau=1$ ns, laser beam intensities $\mathbf{I}(\omega)=10^7$ W/cm^2, and $\mathbf{I}(2\omega)=10^6$ W/cm^2.

inversion symmetry, alluded to above, to which we plan to return in a future communication. We note that silicon may not be a viable candidate for observing such coherently controlled current injection, for if $2\hbar\omega$ is above the direct band gap in silicon then $\hbar\omega$ is above the indirect gap, and one-photon indirect absorption may flood the sample with carriers. But Ge does not suffer from this problem, and LDA band structure calculations indicate that, at the appropriate frequencies, the effect should be observable there as well. Thus we can reasonably expect that it should be possible to coherently control the current optically injected in a variety of bulk semiconductors; this is of interest from both fundamental and technological points of view.

We thank Dr. Claudio Aversa, of the Quantum Institute of the University of California at Santa Barbara, for helpful discussions; we are indebted to Professor Henry Krakauer of the College of William and Mary for providing us with the FLAPW program. This work was supported by the Natural Sciences and Engineering Research Council of Canada, and the Ontario Laser and Lightwave Research Center.

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