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Phase-coherent control of photocurrent directionality in semiconductors

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We demonstrate that one can generate and control photocurrents in semiconductors, without bias voltage, through multiple-frequency phase-coherent laser excitation of donors.

The magnitude and direction of a photocurrent generated in a semiconductor are controlled by a bias voltage for a given concentration and spatial distribution of charge carriers.[1] The role of this voltage is to give *thermodynamic* preference to the flow of photoelectrons in one direction (the forward or backward direction in a *p-n* junction). In a *p*-type or *n*-type semiconductor the probability of carrier photoemission (from a single impurity) without an external voltage is anisotropic only inasmuch as the crystal possesses mass or dielectric constant anisotropies, but the probabilities of emission backward and forward along a given crystal axis are equal. Although photocurrents are commonly produced by laser illumination, the laser coherence does not affect the process.

In the present paper we suggest a scheme for generating and controlling photocurrents *without bias voltage*, relying instead on the coherence of the illuminating source. In the method one creates a superposition of two bound donor (or exciton) states which is then photoionized by two mutually phase-locked lasers at slightly different frequencies with the same polarization axis. The result is a current along the direction of polarization. This scheme constitutes an application, to solid-state devices, of a proposal by Brumer and Shapiro[2] for controlling gas phase reaction products. The realization of the scheme is discussed for shallow-level donors in semiconductors.

Consider a semiconductor doped with shallow-level donors. The **n**th bound-state wave function of such a donor is successfully described by the hydrogenic effective-mass theory[3] as follows:

$$\psi_{\mathbf{n}}(\mathbf{r})=\langle \mathbf{r}\,|\,\mathbf{n}\rangle = V^{-1/2}\int_{-\infty}^{\infty} B_{\mathbf{n},\mathbf{k}}u_{\mathbf{k}}(\mathbf{r})e^{i\mathbf{k}\cdot\mathbf{r}}d\mathbf{k}\,. \tag{1}$$

Here $u_{\mathbf{k}}(\mathbf{r})$ is the conduction-band Bloch state correlated to the asymptotic free-electron momentum $\hbar\mathbf{k}$, V is the normalization volume, $B_{\mathbf{n},\mathbf{k}}$ is the corresponding Fourier component of the hydrogenic wave function envelope $\chi_{\mathbf{n}}$. For semiconductors with effective-mass anisotropy, the $\chi_{\mathbf{n}}$ are evaluated variationally.[4-7] Although the theory described below holds for any superposition of bound donor states, a superposition of $|1s\rangle$ and $|2p_0\rangle$ states will be considered explicitly. For these cases the simple variational procedure introduced by Kohn and Luttinger,[7] whose results agree reasonably well with those of more refined procedures,[4,6] yields

$$\begin{aligned}\chi_{1s}&=\pi^{1/3}\exp\{-[(x^2+y^2)/a^2+z^2/b^2]^{1/2}\}\,,\\ \chi_{2p_0}&=\sqrt{2}\pi^{3/4}b^{-1}z\exp\{-[(x^2+y^2)/a^2+z^2/b^2]^{1/2}\}\,.\end{aligned} \tag{2}$$

Here the coordinates [normalized to the effective Bohr radius $a^*=\hbar^2/(m_\perp e^2)$] coincide with the main axes of the cubic crystal. Depending on the ratio $\gamma=m_\perp/m_\parallel$ (the parallel direction coinciding with z), the a and b parameters vary between $a=b=1$ for nearly isotropic materials with $\gamma=1$ (e.g., GaAs, GaSb, InAs) and $a\approx 4/3\pi$; $b\approx \frac{1}{3}(4/\pi)^{2/3}\gamma^{1/3}$ for highly anisotropic materials (e.g., Si or Ge) with $\gamma\ll 1$.

Let a superposition of the $|1s\rangle$ and $|2p_0\rangle$ states be prepared by some coherent process. As pointed out before,[2] this can be achieved by a short coherent laser pulse or various other means. It is possible to discriminate against the excitation of the $|2p_{\pm 1}\rangle$ states either by frequency tuning (e.g., the $2p_{\pm 1}$-$2p_0$ splitting is ~ 5 meV in Si), or by linearly polarizing the laser along the z axis. Consider now the simultaneous excitation of this superposition state to a kinetic energy level E_k in the conduction-band continuum by two z-polarized infrared or visible lasers with frequencies ω_{1s} and ω_{2p_0}; the former lifts the $|1s\rangle$ state to E_k and the latter lifts the $|2p_0\rangle$ state to E_k. In the Franck-Condon approximation[8] these excitations involve the energy conservation relation:

$$E_k=\frac{\hbar^2k_\perp^2}{2m_\perp}+\frac{\hbar^2k_z^2}{2m_\parallel}=\hbar\omega_{\mathbf{n}}-|E_{\mathbf{n}}|-\sum_p p\hbar\omega\,. \tag{3}$$

Here the **n**-state energy is measured from the conduction-band edge and the last term accounts for the emission ($p>0$) or absorption ($p<0$) of p phonons of frequency ω. For the sake of simplicity, we shall use the zero-phonon-frequency line;[8,9] hence

$$\hbar\omega_{1s}=E_k+|E_{1s}|\,,\quad \hbar\omega_{2p_0}=E_k+|E_{2p_0}|\,.$$

The consideration of a more realistic phonon spectral distribution poses no difficulties in principle, requiring only the averaging of the current calculated here over this distribution.

In what follows we consider only electric dipole induced optical transitions with the electric field along the z axis.

The electric dipole transition amplitudes from an impurity state $|\mathbf{n}\rangle$ to the asymptotic (far from impurity) plane wave $\langle\mathbf{r}|\mathbf{k}\rangle = V^{-1/2}e^{i\mathbf{k}\cdot\mathbf{r}}u_{\mathbf{k}}(\mathbf{r})$ is

$$\langle\mathbf{k}|\mu_z|\mathbf{n}\rangle = \frac{-ie\hbar}{m_{\parallel}(E_k+|E_{\mathbf{n}}|)}\langle\mathbf{k}|(-i\hbar\partial/\partial z)|\mathbf{n}\rangle . \tag{4}$$

The last factor is, using Eq. (1), simply given as

$$\langle\mathbf{k}|-i\hbar\partial/\partial z|\mathbf{n}\rangle = \hbar k_z\langle\mathbf{k}|\mathbf{n}\rangle = \hbar k_z B_{\mathbf{n},\mathbf{k}} . \tag{5}$$

Following Brumer and Shapiro[2] we now consider the photoionization of the superposition state,

$$|\psi\rangle = c_1|1\rangle + c_2|2\rangle , \tag{6}$$

where 1 denotes the $1s$ state and 2 the $2p_0$ state. We let a z-polarized two-color source, whose electric field is given as

$$\varepsilon_z(t) = \varepsilon_1\cos(\omega_1 t+\phi_1)+\varepsilon_2\cos(\omega_2 t+\phi_2) \tag{7}$$

act on this superposition state. The rate (probability per unit time and unit solid angle) of photoemission to a conduction state with momentum $\hbar\mathbf{k}$ resulting from this action is, by a simple extension of a first-order result for a single state,

$$P(\cos\theta) = (2\pi/\hbar)\rho(k)\Big|\sum_{n=1,2} e^{i\phi_n}\varepsilon_n c_n\langle\mathbf{k}|\mu_z|n\rangle\Big|^2 . \tag{8}$$

Here,

$$\begin{aligned} &\cos\theta = k_z/k;\ \sin\theta = k_{\perp}/\gamma^{1/2}k , \\ &k = (2m_{\parallel}E_k)^{1/2}/\hbar , \\ &\rho(k) = (m_{\perp}V/8\pi^3\hbar^2)k , \end{aligned} \tag{9}$$

and $\rho(k)$ is the density of final states. The Franck-Condon factor for the zero phonon line has been set here to unity.

Denoting $c_n = |c_n|\exp(i\alpha_n)$ and using Eqs. (4) and (5) in Eq. (8) gives the form

$$P(\cos\theta) = [A_1|B_{1s,\mathbf{k}}|^2 + A_2|B_{2p_0,\mathbf{k}}|^2 + A_{12}\cos(\alpha_1-\alpha_2-\phi_1+\phi_2+\alpha_{12})B_{1s,\mathbf{k}}B_{2p_0,\mathbf{k}}|]\cos^2\theta , \tag{10}$$

where

$$\begin{aligned} A_n &= \frac{2\pi e^2\hbar^3k^2\rho(k)|\varepsilon_n c_n|^2}{m_{\parallel}^2(E_k+E_n)^2} \quad (n=1,2) , \\ A_{12} &= \frac{4\pi e^2\hbar^3k^2\rho(k)|\varepsilon_1\varepsilon_2 c_1 c_2|}{m_{\parallel}^2(E_k+E_1)(E_k+E_2)} . \end{aligned} \tag{11}$$

Here α_{12} is defined by $B_{1s,\mathbf{k}}B^*_{2p_0,\mathbf{k}} = |B_{1s,\mathbf{k}}B_{2p_0,\mathbf{k}}| \times\exp(i\alpha_{12})$ and $E_1 = |E_{1s}|$, $E_2 = |E_{2p_0}|$.

The evaluation of $P(\cos\theta)$ requires the Fourier components $B_{\mathbf{n},\mathbf{k}}$. For the present choice of impurity states and z axis these components are obtained[10] from Eq. (2) as

$$\begin{aligned} B_{1s,\mathbf{k}} &= 8\pi^{4/3}a^2bV^{-1/2}/G^2 , \\ B_{2p_0,\mathbf{k}} &= -i\sqrt{2}(32)a^2b^2\pi^{7/4}V^{-1/2}a^*k_z/G^3 , \end{aligned} \tag{12}$$

with

$$G = G(\cos^2\theta) = [1+\gamma(a^*ak)^2 + (b^2-a^2\gamma)(a^*k)^2\cos^2\theta] .$$

It is clear from Eq. (12) that $\alpha_{12} = \pi/2$.

Given the above expression, the net current flowing in the z direction is given as an integral over all angles of Ω,

$$\begin{aligned} I_z^+ &= (eNV\hbar/m_{\parallel})\tau F\int_0^{2\pi}\int_0^{\pi} d\Omega\, P(\cos\theta)k\cos\theta \\ &= 256(eNV\hbar^4k^5/m_{\parallel}^3)\tau F a^4b^3\pi^{25/12}\frac{|\varepsilon_1\varepsilon_2 c_1 c_2|}{(E_k+E_1)(E_k+E_2)}\cos\left[\alpha_1-\alpha_2-\phi_1+\phi_2+\frac{\pi}{2}\right]\int_{-1}^{+1}dx\frac{x^4}{[G(x^2)]^5} , \end{aligned} \tag{13}$$

where τ is the free-electron collisional relaxation time, N is the donor concentration in cm^{-3}, and F is the x-y cross-sectional area of the sample.

We note that contributions from the diagonal A_1 and A_2 terms are odd in $\cos\theta$ and have vanished, whereas *the interference term induces a directional current flow.* The analytic form of the integral in Eq. (13) is readily obtainable[10] but adds little insight and hence is not given here. Considerable simplification results if $a^*k \ll 1$ in which case the integral reduces to $\frac{2}{5}$.

We have also extended this analysis[11] to a situation in which relaxation and inhomogeneous broadening of the bound superposition state play a role, by solving the corresponding semiclassical Bloch equations and treating the transition to the conduction band as a weak perturbation on the bound superposition, which is coherently mixed by an Rf source with Rabi frequency Ω_R. The form of Eq. (10) is recovered, as is the controlled current, with $|c_1c_2|$ of Eq. (11) being replaced by $D(0)r_0\Omega_R/\Gamma$ and $\alpha_1-\alpha_2$ by $\pi/2$. Here $D(0)$ is the center value of the inhomogeneously broadened profile, r_0 is the difference in populations of states 1 and 2, and Γ is the combined radiative and thermal inhomogeneous width of the 1-2 transition.[12,13]

Several additional remarks are in order: First, the phases ϕ_1 and ϕ_2 of Eq. (7) contain the spatial phase factors $\exp[i\mathbf{k}\cdot\mathbf{r}]$, where $\mathbf{k}$ is the light wave vector. The difference in the spatial phase can be exactly offset by the phase difference $\alpha_1-\alpha_2$ in the preparation step (e.g., in a Raman preparation of $|\psi\rangle$), or eliminated by phase matching. Second, there are substantial experimental simplications associated with applying the photodissociating lasers at the same time as initiating the preparation of

the superposition state. Third, two color light also causes excitation (via ω_{2p_0}) of the $|1s\rangle$ level to the state at $[E_k+|E_{2p_0}|-|E_{1s}|]$ and of the $|2p_0\rangle$ level (via ω_{1s}) to the state at $[E_k+|E_{1s}|-|E_{p0}|]$. These terms contribute to the A_1 and A_2 terms in Eq. (10) and hence do not contribute to degrade the controlled current I_z^+.

The magnitude and sign of the current are controllable for a given host material and superposition state parameters via (a) the optical phase difference $\phi_1-\phi_2$, (b) the donor number N, and/or (c) the ionizing field strengths ε_1 and ε_2 and their frequencies ω_1 and ω_2. To estimate a typical current, consider the I_z resulting from the following parameters: $\varepsilon_1=\varepsilon_2=0.1$ V/cm, $k=5\times10^7$ cm^{-1}, $|c_1c_2|=0.25$, and $\tau=10^{-14}$ to 10^{-13} sec. The latter corresponds to a mean-free-path ($\hbar k\tau/m$) of 100 to 1000 Å, a typical value for the ballistic electrons at the cited k value. Further $N(\mathrm{Si})V=(10^{18}\ \mathrm{cm}^{-3})V$ where V is the effective interaction volume. For a sample of $0.1\times10\times10$ μm, $V=10^{-11}$ cm^3. Utilizing Eq. (13), and these parameter values, we obtain a current $I_z=10$ to 100 mA. Thus, sizable currents may be readily produced, due to the high quantum efficiency of the silicon photoionization. Indeed, the sizable magnitude of this current indicates clearly that observable currents will arise even under far less favorable conditions, e.g., weaker maser mixing of the bound levels, substantially reduced dopant density, etc.

Equations (9)–(13) apply, evidently, to photoionization of other $|ns\rangle-|n'p_0\rangle$ superpositions, where $|n-n'|=1$, upon substituting the appropriate Fourier coefficients $B_{ns},\mathbf{k}$ and $B_{n'p_0},\mathbf{k}$. It may turn out to be more practical to use high-lying states, e.g., $|5s\rangle$ and $|5p_0\rangle$, for which $\hbar(\omega_2-\omega_1)$ is of the order 0.1 meV, two orders of magnitude smaller than the difference between $|1s\rangle$ and $|2p_0\rangle$ states in Si.

The proposed scheme, apart from its inherent novelty, merits attention also because of its potential applications. It may serve to measure the relative phase of two beams originating from a common laser, which is accumulated in their passage through different nonlinear optical elements. By modulating this phase, information can be transferred via corresponding modulations in the current through the crystal. Laser phase fluctuations, on a longer time scale than optical excitations, one can cause current reversals and thus be detectable using these schemes.

An interesting foreseeable application of these schemes is the monitoring of elastic (direction-changing) collisions which ballistic electrons undergo in semiconductors. Upon injecting the coherently generated electrons into a region of much lower potential energy than the donor-doped region, these electrons acquire high velocities, corresponding to ballistic propagation.[14] Direction-changing collisions in this region would reduce the fractional current in the direction of the laser polarization calculated above. Information on such collisions should be obtained by monitoring this current as a function of the electron propagation path, after subtracting the current of electrons undergoing inelastic (velocity-changing) collisions by means of a tunneling-hot-electron-transfer amplifier (THETA) device.[15] Thus, it should be possible to augment the performance of this device, which is capable presently of rendering accurate information only on velocity-changing collisions.

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