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Picosecond Response of Photoexcited GaAs in a Uniform Electric Field by Monte Carlo Dynamics

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Abstract

The transient electrical response of GaAs photoexcited by a sub-picosecond pulse, in the presence of a uniform biasing electric field, has been studied using a Monte Carlo calculation. Non-interacting electron transport is considered, using the three-valley model for the conduction band. Scattering from acoustic, optical, and intervalley phonons is included. The valence band dispersion relations and valence to conduction band momentum matrix elements needed to treat the optical absorption were obtained from a full zone $\mathbf{k}\cdot\mathbf{p}$ calculation. The optical absorption has been given a realistic treatment by including an effective energy linewidth resulting from the combination of the Fourier transform of the driving pulse, electron-phonon scattering, and the effect of the applied electric field. The average electron velocity is found to overshoot its steady state value only if the electric field is larger than a critical value which increases with the photon energy. For example, these calculations indicate that at 5.0 kV/cm, overshoot occurs for a photon energy of 1.5 eV but not for 1.7 eV. Velocity overshoot is seen to occur when the steady state average electron energy (for the given applied field) is larger than the average electron energy of the initial photoexcited distribution. The regime of applied field and photon energy necessary for overshoot is mapped out.

I. Introduction

Design of semiconductor devices with desired high-speed properties requires an understanding of how the microscopic *dynamics* of carrier transport results in a particular electrical response. The transient response of a semiconductor is a direct consequence of the relaxation of the carrier distribution towards its steady state. The relaxation depends on the interactions of the carriers with the lattice and with each other, as well as on the details of the band structure. An accurate microscopic model of the carriers' dynamics therefore can be a very useful predictive tool. The Monte Carlo (MC) method[1-5] can be employed in the accurate calculation of individual semiclassical electron (and hole) trajectories and thus, can ultimately give the macroscopic response. At the same time its appeal to physical intuition and direct interpretation make it ideally suited to these distribution relaxation problems.

Laser excitation provides a convenient method for suddenly changing the carrier distribution and then following the relaxation. Current experimental techniques allow for measurement of electrical transients in semiconductors on the sub-picosecond time scale. Experiments have been performed in which a colliding pulse, mode-locked (CPM) laser is used to produce a train of pulses that have durations of about 100 fs and energies of 2.0 eV. The beam of pulses is split into two beams, with a timing relationship that can be precisely varied. One beam is used to photogenerate electron-hole pairs in an electrically biased semiconductor sample, and the second beam is used in the temporal sampling of the electrical response.[6-8] The sampling can be performed either with a short response-time photoconductor or with an electro-optic polarizing material.

The CPM lasers that have been used for such experiments are tunable only over a very narrow spectral range near 2.0 eV. However, experimental studies for a range of photon energies are needed to fully characterize photoconductive responses, because the initial carrier distribution depends strongly on the photon energy. Experimental techniques to generate spectrally tunable subpicosecond pulses are becoming available.[9-10] To aid in the interpretation and understanding of these types of experiments, we present a MC calculation of the relaxation of the distribution for photoexcited GaAs in the presence of a uniform electric field. In particular,the dependence of the response on the photon energy will be determined.

The dynamics of carrier relaxation toward a steady state distribution depends on the initial distribution and on the steady state distribution. The initial distribution is determined by the characteristics of the laser pulse, principally the photon energy, and the steady state distribution is determined by the electric field. In this paper, we present a systematic series of Monte Carlo calculations of electron relaxation in GaAs. We consider the case in which the electrons are generated by a 100 fs optical pulse and the GaAs is biased by a uniform electric field. We investigate the dynamics of the electron relaxation as a function of the photon energy and the magnitude of the applied field.

Similar MC calculations[3] have been performed with a field, but using as initial condition the state with all carriers at rest at $\mathbf{k}=0$. It is impossible to create this situation experimentally. Photoexcitation results in the initial carrier distribution occupying two or three shells in $\mathbf{k}$-space, one from each of the highest valence bands. For the $\mathbf{k}=0$ initial condition, MC calculations predict that the transient average carrier velocity overshoots its steady state value, sometimes by a large

factor. The initial condition produced by photoexcitation, however, can lead to completely different results, such as a lack of any overshoot feature. In this paper, we determine the conditions on the photon energy and bias field for electron velocity overshoot to occur.

Our calculations indicate that an accurate determination of the photoexcited distribution (and therefore its average electron energy) is necessary for an accurate modeling of the electrical response. We find that velocity overshoot occurs only when the initial state average electron *energy* is less than the steady state value. The *initial* average energy is roughly proportional to the photon energy minus the band gap, and the *steady state* average energy is a nonlinear function of the applied field, so that the presence of velocity overshoot is determined by the relative size of the photon energy compared with the field.

We have calculated the photoexcited distribution by using valence bands obtained from a full zone $\mathbf{k}\cdot\mathbf{p}$ calculation, while using a nonparabolic effective mass fit to the Γ-valley of the conduction band. The fitting procedure for the conduction band (as opposed to the full zone $\mathbf{k}\cdot\mathbf{p}$ results) was used to be consistent with the description of the conduction band in terms of its Γ, L and X valleys in the MC calculation. In this way the scattering processes are easier to treat. Effects of an absorption linewidth have also been included, through use of the Maxwell-Bloch equations with a damping time T_2, applied to a light pulse with an electric field varying with time according to $\mathrm{sech}(t/\tau_0)$, where τ_0 is a width parameter. In these calculations, the laser intensity is assumed to have a full width at half maximum (FWHM) of 100 fs, and photon energies will be considered from 1.5 eV to 2.2 eV. The time T_2 is determined in a self-consistent manner by the total carrier scattering

rate and the applied electric field. Typically we will have $T_2 \ll \tau_0$ so the lineshape will be Lorentzian with energy width varying as $1/T_2$. Because of the energy dependence of the scattering rates, the linewidths will be functions of both the electric field and the photon energy. For moderately large fields and photon energies, the widths can become as large as $\approx$ 100 meV. We have found, however, that the net effect of this energy width for most cases of interest has been rather small.

To obtain the leading order response behavior, a number of simplifying assumptions have been used. First of all, for the low number density limit, carrier-carrier interactions are excluded. This approximation is valid at low laser intensity. At room temperature, the photogenerated carrier density should not exceed about 10^{17} cm^{-1}. The electric field is assumed to be uniform. This latter condition may actually be difficult to satisfy experimentally. Device response calculations based on a macroscopic continuum model indicate that the response of a high resistivity photoconductor to a moderate intensity sub-picosecond pulse involves a collapse of the electric field, due to the opposing directions of electron and hole drift.[11] Using low laser intensity, so that the photogenerated carrier density is small, minimizes the effects of the collapsing electric field.[11] We take into account only the transport of electrons, assuming that the lower drift velocity of the holes makes their contributions to the electrical response much smaller.[12] As mentioned above, the conduction band will be considered as composed of the valleys at Γ, L, and X. This means that the process of drifting from the Γ point over the energy peak and then into either the L or X valley is excluded. Typically the scattering rates increase strongly enough (moving up in the band) so that this process is improbable.

The band structure for GaAs as obtained from the full zone $\mathbf{k}\cdot\mathbf{p}$ calculation is

reviewed in Sec. II. Details of the model employed for the MC calculation, including the necessary scattering rate parameters and valley fitting constants, are discussed in Sec. III. Results of a steady state transport calculation for GaAs also are given. In Sec. IV, the details of how the photoexcited initial distribution was generated and how the linewidth was included are discussed. Results are presented in Sec. V for GaAs with fields up to 50 kV/cm and photon energies ranging from 1.5 to 2.2 eV. Our conclusions concerning the trends in the photo-responses are discussed in Sec. VI.

II. Band Structure for GaAs

It is straightforward to apply the **k**·**p** pseudopotential method to the GaAs band structure. The calculation was carried out by first using a basis of 113 plane wave states, corresponding to reciprocal lattice vectors of the zinc-blende structure with squared magnitudes of 0, 3, 4, 8, 11, 12, 16, and 20 (in units of $(2\pi/a)^2$). After diagonalization for **k**=0, only the 27 lowest energy states are retained. The spin-orbit interaction is added at this point, with splitting parameters $\Delta_0 = 0.34$ eV and $\Delta_1 = 0.22$ eV, and the 54 × 54 matrix is then diagonalized for a range of **k**. Input to the calculation is through the pseudopotential form factors, which have been taken from Ref. 13, for 300 K. We modified these form factors slightly to those given in the caption of Fig. 1, in order to enforce the band gap to be 1.44 eV, and the splittings from Γ to L and from Γ to X to be 0.33 eV and 0.52 eV respectively. The lattice spacing is taken to be $a = 5.6533$ Å.

The resulting band structure is shown in Fig. 1. An effective mass fit to the Γ-valley of the conduction band gives $m^*/m_0 \approx 0.077$, with a nonparabolicity pa-

rameter $\alpha \approx 0.8$ eV^{-1}. It is difficult to find a set of pseudopotential form factors that will produce an effective mass and nonparabolicity close to the accepted values of 0.063 and 0.69 eV^{-1} respectively.[4] Correspondingly, heavy hole and split-off hole masses are approximately 0.7 and 0.2, reasonably close to accepted values. Similarly we can estimate the resulting effective conduction band masses near the L and X points. Taking these as ellipsoidal valleys, approximate longitudinal and transverse masses turn out to be $m_l^L/m_0 \approx 1.5$, $m_t^L/m_0 \approx 0.12$, $m_l^X/m_0 \approx 1.5$, and $m_t^X/m_0 \approx 0.25$. These values were used in the fits to these valleys for the MC calculations. The major utility of this band structure calculation, however, was in employing its valence bands for the photoabsorption calculation, and also in using the valence to conduction band momentum matrix elements for correctly weighting the allowed optical transitions. The conduction band (Γ-valley), however, was fitted with an effective mass of 0.063 m_0 and nonparabolicity of 0.69 eV^{-1} for the photoabsorption calculation, in order to use the same fit in the MC calculation and therefore to conserve energy correctly.

III. Monte Carlo Model and Steady State Results

The MC method as applied to transport in semiconductors has been described in Ref. 1. In the semiclassical approximation, drift of a carrier in the applied electric field is treated classically as smooth motion in a band, interrupted by quantum mechanical scattering events that discontinuously change the carrier's wavevector $\mathbf{k}$. The conduction band can be described in terms of the valleys at Γ ($\mathbf{k}$=0), L ($\mathbf{k}=(\frac{1}{2},\frac{1}{2},\frac{1}{2})\frac{2\pi}{a}$ and symmetric points) and X ($\mathbf{k}=(1,0,0)\frac{2\pi}{a}$ and symmetric points), with intervalley phonon scattering making transitions between the valleys possible.

The L and X valleys are taken to be ellipsoidal and nonparabolic. In this way, in an applied field, the three X-valleys and four L-valleys become inequivalent for an arbitrary field direction, and effects of this **k**-space anisotropy are retained. There may be some dependence of the response on the field direction that cannot appear if all the valleys are taken to be spherical. This effect might be expected to be strongest for large electric fields. The ratios of transverse to longitudinal effective mass have been taken from the full zone **k**·**p** calculation, and the density of states masses $m_d = (m_l m_t^2)^{1/3}$ have been fixed to values given in Ref. 4, which also was the source for the valley nonparabolicity parameters. Thus the dispersion within each valley is given by a relation

$$\epsilon(k^*) = \epsilon_0 + \frac{1}{2\alpha}\left\{-1 + \left[1 + 4\alpha\frac{\hbar^2}{2m_0}k^{*2}\right]^{\frac{1}{2}}\right\}, \tag{1}$$

where

$$k^{*2} = \frac{m_0}{m_t}k_t^2 + \frac{m_0}{m_l}k_l^2. \tag{2}$$

Here ϵ_0 is the energy at the bottom of the valley, and k_l and k_t are the longitudinal and transverse components of **k** as measured in the valley, and α is the nonparabolicity parameter.

The electrons are assumed to scatter from acoustic, polar optic, nonpolar optic (only in the L valley), and intervalley phonons. In the low number density limit we exclude carrier-carrier interactions. A fictitious "self scattering" is also included, so that the time intervals between scattering events can be chosen from an exponential distribution. (See Ref. 1.) The interaction Hamiltonians for each type of scattering process and expressions for the scattering rates can be found in Refs. 1 and 2; coupling constants and phonon frequencies have been taken from Table 1 of Ref. 4.

The total scattering rate due to real physical processes (excluding self scattering) will depend on the valley as well as on the carrier energy. Measuring all energies relative to the energy at the Γ-point, the total scattering rates for GaAs at 300 K are shown in Fig. 2, including contributions of phonon emission and absorption processes.

For noninteracting electrons, a large number of single electron trajectories can be generated, and then averages of time-dependent quantities can be made by averaging over all the trajectories. For each trajectory, the final **k**-state after each scattering event and the times of the events are enough to reconstruct the electron's **k**-state at any desired time. For the scattering time intervals to belong to an exponential distribution, they are chosen with a random number $0 < r < 1$ by

$$\Delta t_s = \Gamma_0^{-1} \ln(r), \qquad \text{where} \qquad \Gamma_0 = \Gamma_{\text{phys}} + \Gamma_{ss}. \tag{3}$$

The constant Γ_0 is a scattering rate chosen to be greater than the sum of the rates of all the real physical processes, Γ_{phys}, and it is assumed that the electron energy never exceeds some chosen maximum value. The difference $\Gamma_0 - \Gamma_{\text{phys}}(\varepsilon)$ gives the self scattering rate Γ_{ss}, at the energy ε. When a self scattering event occurs, the electron's final state is set equal to its initial state, and it continues to drift in the field as if no scattering occurred. The term "self scattering" is a misnomer; a better name for it might be "non-scattering."

Between scattering events, the equation of motion is

$$\hbar \dot{\mathbf{k}} = q\mathbf{E}, \tag{4}$$

where q is the charge and $\mathbf{E}$ is the applied field. At each scattering time the type of scattering process is chosen randomly but according to the relative probabilities

of each of the allowed processes, using a random number generator. The random number generator is also used to choose an allowed final state, from those consistent with conservation of energy and momentum. For scattering from optical phonons, choosing from the allowed states is relatively easy because the phonon frequency is fixed. For acoustic phonons the choice is more difficult and a "rejection technique" for choosing final states of the desired distribution is used (as in Ref. 1).

The time-dependent response quantities of most interest are the average velocity (or current), the average carrier energy, and the relative populations of the valleys. The velocity is determined by

$$\mathbf{v} = \frac{1}{\hbar}\frac{\partial \varepsilon}{\partial \mathbf{k}}. \tag{5}$$

For the steady state case, time averages of these quantities can be easily calculated, and fewer electrons are needed to get small statistical errors than are needed for time-dependent problems. As an example, we have reproduced the calculation of the steady state conduction in GaAs at 300 K, using the trajectories of 400 electrons for 100 ps. The beginning 20 ps of each trajectory was discarded. Results for the average velocity and energy versus the applied field (in the [100] direction) are shown in Fig. 3. The initial condition consisted of all electrons starting at $\mathbf{k}$=0, with averages formed from the data after the initial transient passed. The carrier energy tends to saturate because the scattering rates, which increase with energy, compete against the applied field, tending to prevent the energy from increasing. The energy versus field curve will be relevant for estimating the velocity overshoot regime for the time dependent photoexcited transport problem.

IV. Generation of the Photoexcited Initial Distribution

We consider a laser pulse with vector potential varying as

$$\mathbf{A}(t) = A_0\,\hat{\epsilon}\,e^{i\omega_0 t}\mathrm{sech}(t/\tau_0), \tag{6}$$

with amplitude A_0, polarization unit vector $\hat{\epsilon}$, center frequency ω_0, and envelope width parameter τ_0. (For this envelope, the laser intensity FWHM = 1.76 τ_0.) If effects of the absorption linewidth due to damping are excluded, and also $\omega_0\tau_0 \gg 1$, then the rate for a transition from a valence band state $v_{\mathbf{k}}$ to a conduction band state $c_{\mathbf{k}}$ is given in the dipole approximation by Fermi's Golden Rule,

$$w = \frac{2\pi}{\hbar} \mid \langle c_{\mathbf{k}} | \mathcal{H}_{int} | v_{\mathbf{k}} \rangle \mid^2 \delta(\varepsilon_c - \varepsilon_v - \hbar\omega_0), \tag{7}$$

with interaction Hamiltonian

$$\mathcal{H}_{int} = \frac{e}{m_0 c}\mathbf{A}\cdot\mathbf{p}. \tag{8}$$

Here the envelope slowly modulates the transition rate. The general case with damping will be considered in more detail below. States in **k**-space are chosen satisfying the delta function, with the conduction band described by Eq. 1 and the valence band dispersion $\varepsilon_v(k)$ derived from the full zone **k**·**p** calculation. The delta function defines an equi-energy surface in **k**-space. After choosing a random direction in **k**-space, the magnitude of **k** for the transition from a chosen valence band is found numerically. When computing averages, the subsequent MC trajectory of the electron created at that **k** is then weighted by the momentum matrix element $\mid \langle c_{\mathbf{k}} | \hat{\epsilon}\cdot\mathbf{p} | v_{\mathbf{k}} \rangle \mid^2$. An initial distribution of electron states will be composed of either two or three peaks, due to the two or three allowed valence to conduction band transitions, and they will have small energy widths caused by the angular

dependence of the valence band dispersion relations. For transitions from a given hole band, numerical integration of the transition rate over the equi-energy surface then gives the relative contribution of transitions from that hole band to the absorption. Initial electron states are thus created from the two or three allowed transitions with the appropriate relative numbers (as determined by these integrals involving the optical matrix elements). The total absorption coefficient α_{abs} and the contributions from the three transitions are shown in Fig. 4.

Strictly speaking, Fermi's Golden Rule as given above should be used only in situations where the electron will not likely scatter during the laser pulse time interval. If the scattering rate is high enough, however, a linewidth is introduced due to the finite lifetime of the excited state. Similarly, an applied field also causes a finite lifetime and therefore a linewidth. The effects of both of these processes can be incorporated in terms of a damping time, T_2, as a parameter for the Maxwell-Bloch (MB) equation. If the electron-photon interaction Hamiltonian is written in the form

$$\mathcal{H}_{int} = \gamma(\mathbf{k})a(t), \tag{9}$$

with

$$\gamma(\mathbf{k}) = \frac{eA_0}{m_0 c}\langle c_{\mathbf{k}}|\hat{\epsilon}\cdot\mathbf{p}|v_{\mathbf{k}}\rangle, \qquad \text{and} \qquad a(t) = 2\cos(\omega_0 t)\mathrm{sech}(t/\tau_0), \tag{10}$$

then the transition rate $w(t)$ is given by

$$w(t) = \frac{1}{\hbar}a(t)F(t), \tag{11}$$

where $F(t)$ satisfies the MB equation in the form

$$\frac{d^2F}{dt^2} + \frac{2}{T_2}\frac{dF}{dt} + \left[(\frac{\Delta\epsilon}{\hbar})^2 + \frac{1}{T_2^2}\right]F = \frac{2\,|\,\gamma\,|^2}{\hbar}\left(\frac{da}{dt} + \frac{a}{T_2}\right). \tag{12}$$