Modeling of Ultrafast Propagation and Quantum Kinetics for Laser-Generated Electron-Hole Plasmas in Nanowires

Jeremy R. Gulley
Department of Physics, Kennesaw State University, 370 Paulding Avenue, Kennesaw, GA 30101,USA
jgulley@kennesaw.edu

Danhong Huang
Space Vehicles Directorate, Air Force Research Lab, 3725 Aberdeen Avenue, Kirtland AFB, NM 87117, USA

Abstract: Simulations solve a quantum-kinetic model for ultrafast carrier dynamics in nanowires coupled to resonant scattering of laser pulses. Both transverse and resulting longitudinal electric fields play significant roles in the nanowire dynamics.

1. Introduction

We show the results of simulations solving a self-consistent quantum-kinetic model for the ultrafast laser-generated plasma dynamics in direct-gap semiconductor quantum wires [1]. These simulations strongly couple the plasma dynamics of the nanowire to resonant scattering of ultrashort light pulses propagating through the wire. The electron and hole distributions in momentum space are driven both by the transverse field of the propagating laser pulse and by the longitudinal field resulting from the spatial distribution of laser-generated carriers. The calculations include many-body scattering and dephasing due to carrier-carrier collisions, carrier-phonon collisions, and carrier transport effects. The goal is to study the interaction between the localized electronic response of quantum wires and the spatial-temporal features and phases of the incident light pulses. Future studies will characterize correlations between both pump laser fields and applied DC bias voltages with the localized longitudinal electromagnetic field due to induced plasma oscillations in nanowires.

2. Theory

2.1. Pulse Propagation

A strongly chirped near-IR laser pulse, with electric field $\mathbf{E}(\mathbf{r},t)$ that is polarized in the x-direction, is propagating in the y-direction along a GaAs nanowire [1]. We simulate the propagation by solving the Maxwell equations for the electric and magnetic fields in a non-magnetic medium using a Psuedo-Spectral Time Domain method. This method casts the Maxwell equations in the Fourier transformed wave-vector ($\mathbf{q}$) space [2] and allows for simple separation of fields into transverse and longitudinal parts for 1D, 2D, and 3D propagation simulations.

2.2. Laser-Semiconductor Plasma Interaction

The laser-excited electron-hole plasma in the quantum wire is spin-degenerate, and the quantum-kinetic semiconductor Bloch equations (SBEs) describing the electron-hole dynamics are given by [1,3,4]

$$\frac{dn_k^{\rm e}(t)}{dt} = \frac{2}{\hbar}\sum_{k'} {\rm Im}\left\{\mathbf{p}_{k,k'}(t)\cdot\mathbf{Q}_{k',k}(t)\right\} + \left.\frac{\partial n_k^{\rm e}(t)}{\partial t}\right|_{\rm rel}, \tag{1}$$

$$\frac{dn_{k'}^{\rm h}(t)}{dt} = \frac{2}{\hbar}\sum_{k} {\rm Im}\left\{\mathbf{p}_{k,k'}(t)\cdot\mathbf{Q}_{k',k}(t)\right\} + \left.\frac{\partial n_{k'}^{\rm h}(t)}{\partial t}\right|_{\rm rel}, \tag{2}$$

$$\begin{aligned} i\hbar\frac{d\mathbf{p}_{k,k'}(t)}{dt} = &\left[\varepsilon_k^{\rm e}+\varepsilon_{k'}^{\rm h}+\varepsilon_{\rm G}+\Delta\varepsilon_k^{\rm e}+\Delta\varepsilon_{k'}^{\rm h}-i\Delta_{k,k'}^{\rm eh}(t)\right]\mathbf{p}_{k,k'}(t)-\left[1-n_k^{\rm e}(t)-n_{k'}^{\rm h}(t)\right]\mathbf{Q}_{k,k'}(t) \\ &+i\sum_{q\neq 0}\Lambda_{k,q}^{\rm e}(t)\,\mathbf{p}_{k+q,k'}(t)+i\sum_{q'\neq 0}\Lambda_{k',q'}^{\rm h}(t)\,\mathbf{p}_{k,k'+q'}(t)\,, \end{aligned} \tag{3}$$

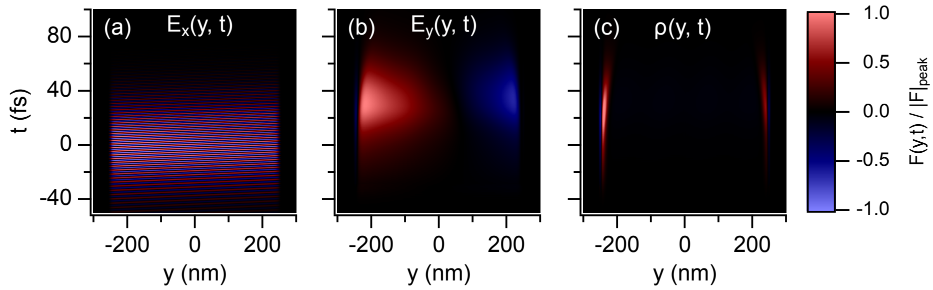

Fig. 1. The (a) transverse $E_x(y,t)$ and (b) longitudinal $E_y(y,t)$ electric fields along the axis of the 500 nm long nanowire, as well as (c) the corresponding 1D electron-hole plasma density $\rho(y,t)$. All are shown as functions of wire position y, where $y=0$ is the wire's center, and time t relative to the peak of the pump pulse at $t=0$. All plots are scaled relative to their individual peak value.

where $\mathbf{p}_{k,k'}(t) = \sum_{\sigma=x,y} p^{\sigma}_{k,k'}(t)\,\hat{\mathbf{e}}^{\sigma}_{\mathbf{d}}$ is the microscopic polarization, $\mathbf{Q}_{k,k'}(t) = \sum_{\sigma=x,y} Q^{\sigma}_{k,k'}(t)\,\hat{\mathbf{e}}^{\sigma}_{\mathbf{d}}$ is the renormalized Rabi frequency, $n^{\mathrm{e}}_{k}(t)$ and $n^{\mathrm{h}}_{k'}(t)$ are the electron (e) and hole (h) occupation numbers at momenta k, and k', respectively. In Eq. (3) ε_{G} is the wire bandgap, $\varepsilon^{\mathrm{e}}_{k}$ and $\varepsilon^{\mathrm{h}}_{k'}$ are the kinetic energies of electrons and holes, $\Delta\varepsilon^{\mathrm{e}}_{k}$ and $\Delta\varepsilon^{\mathrm{h}}_{k'}$ are the Coulomb renormalization [5] of the kinetic energies of electrons and holes, $\Delta^{\mathrm{eh}}_{k,k'}(t) = \Delta^{\mathrm{e}}_{k}(t) + \Delta^{\mathrm{h}}_{k'}(t)$ is the diagonal dephasing rate (quasi-particle lifetime), while $\Lambda^{\mathrm{e}}_{k,q}(t)$ and $\Lambda^{\mathrm{h}}_{k',q'}(t)$ are the off-diagonal dephasing rates [1] (pair-scattering) for electrons and holes. The Boltzmann relaxation terms in Eqs. (1) and (2) contain carrier-carrier Coulomb scattering, carrier-phonon scattering, and carrier transport. From these solutions we calculate the 1D wire polarization [1] for use in the Maxwell Equations:

$$\tilde{\mathbf{P}}_{\mathrm{qw}}(q,t) = \sum_{\sigma=x,y} \tilde{P}^{\sigma}_{\mathrm{qw}}(q,t)\,\hat{\mathbf{e}}^{\sigma}_{\mathbf{d}} \tag{4a}$$

$$\tilde{P}^{\sigma}_{\mathrm{qw}}(q,t) = \frac{d_{\mathrm{cv}}\alpha}{2\delta_0\mathscr{L}} \sum_{k} p^{\sigma}_{k+q,k}(t) + \mathrm{H.C.}\,, \tag{4b}$$

where $\mathbf{p}_{k+q,k}(t)$ is determined by Eq. (3), δ_0 is the thickness of each quantum wire, d_{cv} is the dipole moment between the valence and conduction bands, and H.C. stands for the Hermitian conjugate term. The 1D free-charge density distribution in the wire is given for the Maxwell equations by $\tilde{\rho}(q,t) = \tilde{\rho}^{\mathrm{h}}(q,t) + \tilde{\rho}^{\mathrm{e}}(q,t)$, where $\tilde{\rho}^{\mathrm{h}}(q,t)$ and $\tilde{\rho}^{\mathrm{e}}(q,t)$ are the charge-density distributions of holes and electrons in the nanowire. We calculate these charge densities from $\mathbf{p}_{k,k'}$, n^{e}_{k}, and n^{h}_{k} within the random-phase approximation at each time step [1,6]. Using the total charge density, we calculate the longitudinal displacement field by Gauss's Law: $\tilde{\mathbf{D}}^{\parallel}(q,t) = -i\mathbf{q}\tilde{\rho}(q,t)/q^2$. This, in combination with Eq. 4a, allow us to obtain the longitudinal field everywhere in the wire: $\varepsilon_0\varepsilon_b\tilde{\mathbf{E}}^{\parallel}(q,t) = \tilde{\mathbf{D}}^{\parallel}(q,t) - \tilde{\mathbf{P}}^{\parallel}_{\mathrm{qw}}(q,t)$, where ε_b is the relative dielectric permittivity of the AlAs host material surrounding the nanowire.

3. Results

We performed simulations solving Equations (1)-(4), coupled to the Maxwell equations and the constitutive relations, for a strongly chirped, 800 nm wavelength, 40 fs laser pulse propagating through a 500 nm length GaAs quantum wire. Figure 1 plots the resulting transverse $E_x(y,t)$ and longitudinal $E_y(y,t)$ electric fields along the axis of the quantum wire, as well as the 1D electron-hole charge density. It shows each as a function of the position in the wire and of time. The pulse propagates in the positive y direction and the peak of the pulse occurs at $t=0$ fs. Here, $E_x(y,t)$ acts as a pump pulse, exciting the electron-hole plasma and creating the space-time charge density $\rho(y,t)$. The charge density results in a longitudinal electric field $E_y(y,t)$ present throughout the wire. The peak of the longitudinal field is an order of magnitude lower than the pump field peak (3×10^6 V/cm), but with a lifetime twice as long as the pump pulsewidth, it continues to drive the electronic response long after the pulse is gone.

We use these simulations to look for correlations between both resonant laser fields and applied DC electron-hole currents with longitudinal electron-hole plasma response. Fig. 2 shows the incident, transmitted, and reflected field spectra of the pump pulse in the simulations for Fig. 1. Since the incident pulse was strongly chirped, it has many frequencies on both sides of the wire band gap, $\varepsilon_G = 0.97\omega_0$, where $\omega_0 = 2.35\times10^{15}$ rad/s is the incident pulse central frequency. Note that for a quantum wire, being a 1D system, the density of states at the band edge approaches infinity, and therefore has strong absorption there. The transmitted pulse in Fig. 2(a) shows clear

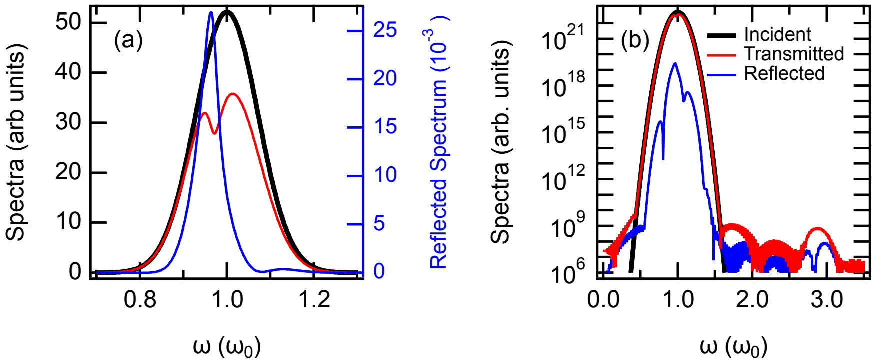

Fig. 2. The incident, transmitted, and reflected field spectra of the pump pulse shown on a linear scale (a) and logarithmic scale (b). The incident pulse is shown in black, the transmitted pulse in red, and the reflected pulse in blue. In (a) the left vertical axis measures the incident and transmitted pulses while the right vertical axis measures the reflected pulse and is scaled down by 10^{-3}. The horizontal axis is measured in units of the fundamental pump field frequency ω_0.

evidence of absorption just below ω_0, while the reflected pulse exists mostly at frequencies below this value. The shape of the reflected pulse spectrum is altered by resonant excitation of the electron-hole plasma, and is correlated to the plasma dynamics in the wire. Meanwhile, the transmitted field in Fig. 2(b) shows evidence of nonlinear optical response with a notable signal at $3\omega_0$, and is correlated to the transverse nonlinear optical response of the wire.

4. Conclusions

We use a recently developed model [1] to examine the localized electron-hole plasma response of nanowires to resonant interaction with chirped ultrashort laser pulses, as well as the spatial-temporal features of those scattered light pulses. Future studies will characterize correlations between both pump laser fields and applied DC bias voltages with the localized longitudinal electric field due to induced long-lasting plasma oscillations in semiconductor nanowires.

References

1. J. R. Gulley and D. Huang,"Self-consistent quantum-kinetic theory for interplay between pulsed-laser excitation and nonlinear carrier transport in a quantum-wire array," Opt. Express **27**(12), 17,154–17,185 (2019).
2. A. Taflove and S. C. Hagness, *Computational Electrodynamics: The Finite-Difference Time-Domain Method* (Artech House, Inc., 2000), 3rd ed.
3. J. R. Kuklinski and S. Mukamel, "Generalized semiconductor bloch equations: Local fields and transient gratings," Phys. Rev. B **44**, 11,253–11,259 (1991).
4. R. Buschlingern, M. Lorke, and U. Peschel, "Light-matter interaction and lasing in semiconductor nanowires: A combined finite-difference time-domain and semiconductor bloch equation approach," Phys. Rev. B **91**, 045,203 (2015).
5. M. Lindberg and S. W. Koch, "Effective bloch equations for semiconductors," Phys. Rev. B **38**, 3342 (1988).
6. G. Gumbs and D. H. Huang, *Properties of Interacting Low-Dimensional Systems* (John Wiley & Sons, 2011).