Collective free-carrier scattering in semiconductors

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We present an analysis of an undoped semiconductor strongly pumped by a coherent electromagnetic radiation field. The free-carrier dynamics and the incident and scattered fields are described classically and self-consistently. The analysis predicts the existence of a collective instability which simultaneously generates a strong free-carrier grating along the propagation direction and a coherently backscattered radiation field. An example is given showing scattered intensities approaching 100% of the pump intensity. A realistic experiment to enable observation of this phenomenon is described.

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When electromagnetic radiation interacts with matter it is scattered, changing its momentum and/or energy. For sufficiently small particles, the scattering process can also result in significant changes in the particles' momentum and energy, e.g., Compton scattering.[1] When considering not just a single-particle interaction but an ensemble of particles, the particle dynamics and the radiation field form a coupled system. In order to describe this coupled system it is necessary to follow the evolution of all particle dynamics and the radiation field in a self-consistent way. This mutual interaction between the ensemble of particles and the radiation can give rise to a much richer class of phenomena than that resulting from only single-particle considerations. Examples of such collective radiation-matter interactions include superfluorescence,[2] the high-gain free-electron laser,[3] the collective atomic recoil laser,[4] and collective Rayleigh scattering.[5]

In this paper we predict the existence of a collective instability in an undoped semiconductor. It will be shown that this instability simultaneously generates a density grating of free electrons and holes and a radiation field coherently backscattered from an incident pump field. The formation of the free-carrier grating is induced by the periodic potential resulting from the interference of the scattered and incident fields, and so has a periodicity of half the radiation wavelength. As the free carriers are initially randomly distributed throughout the sample and we assume that the sample size is much greater than the radiation wavelength, the scattering of the pump field would initially be incoherent, i.e., the scattered radiation intensity would be proportional to the number of free carriers. However, we show that as a result of the induced free-carrier grating, it is possible for a *coherent* scattering process to occur. We shall also discuss several possible methods to observe this effect using, as an example, InSb.

To illustrate this phenomenon we consider the configuration as shown schematically in Fig. 1. It consists of a slab of semiconductor of thickness l_s with a constant equal number of free electrons and holes either intrinsic or photogenerated. A nonresonant pump field ($\mathbf{E}_2$) may be scattered by the free carriers to produce a scattered field ($\mathbf{E}_1$).

We begin by describing the evolution of the radiation fields in the presence of the free carriers by Maxwell's wave equation:

$$\left(\nabla^2-\frac{n^2}{c^2}\frac{\partial^2}{\partial t^2}\right)\mathbf{E}(z,t)=\mu_0\frac{\partial \mathbf{J}}{\partial t}, \tag{1}$$

where n is the background refractive index of the semiconductor. The radiation electric field resulting from the superposition of the incident and scattered fields is assumed to be a plane wave of the form

$$\mathbf{E}(z,t)=[\mathcal{E}_1(z,t)e^{i(kz-\omega t)}+\mathcal{E}_2(z,t)e^{-i(kz+\omega t)}+\text{c.c.}]\hat{\mathbf{x}}, \tag{2}$$

where $\omega=ck/n$, $\hat{\mathbf{x}}$ is a unit vector, and $\mathcal{E}_1(z,t)$ and $\mathcal{E}_2(z,t)$ are complex envelopes describing the scattered and incident fields, respectively. These envelopes are assumed to obey the slowly varying envelope approximation (SVEA) and dielectric reflections at the interfaces are neglected.

To model the evolution of the free carriers we consider the case where the frequency of both radiation pump and probe fields are sufficiently below the band-gap transition frequency to allow a classical description of the free-carrier dynamics. The evolution of each free carrier of charge q and effective mass m^* may be described by use of the Lorentz force equation, modified to include a damping term,

$$m^*\frac{d\mathbf{v}}{dt}=q(\mathbf{E}+\mathbf{v}\times\mathbf{B})-\gamma m^*\mathbf{v}, \tag{3}$$

where $\mathbf{v}$ is its velocity, $\mathbf{B}$ is the magnetic flux density of the radiation fields, and γ is a phenomenological damping rate due to intraband scattering processes. Defining the free-

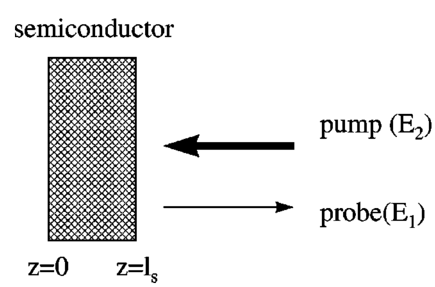

FIG. 1. Schematic diagram of a possible experimental configuration.

carrier type by the subscripts ''e'' (electron) and ''h'' (hole), the current density due to free carrier motion is given by

$$\mathbf{J}(\mathbf{r},t)=e\sum_{j=1}^{N}\left[\mathbf{v}_{h_j}\delta(\mathbf{r}-\mathbf{r}_{h_j}(t))-\mathbf{v}_{e_j}\delta(\mathbf{r}-\mathbf{r}_{e_j}(t))\right], \quad (4)$$

where e is the electronic charge magnitude and N is the total number of electrons and holes (assumed equal) within the semiconductor sample with position vectors $\mathbf{r}_e(t)$ and $\mathbf{r}_h(t)$, respectively. It can be seen from Eqs. (1), (3), and (4) that the free-carrier dynamics and the electromagnetic field are intimately coupled and will evolve self-consistently.

Substituting the radiation field Eq. (2) and its corresponding magnetic component into Eq. (3) we obtain an expression for the induced, oscillatory transverse velocity for each free carrier, i.e.,

$$\mathbf{v}_{\perp}=v_x\hat{\mathbf{x}}\approx\frac{q}{m^*}\left(\frac{\mathcal{E}_1 e^{i(kz-\omega t)}-\mathcal{E}_2 e^{-i(kz+\omega t)}}{\gamma-i\omega}+\text{c.c.}\right)\hat{\mathbf{x}}. \quad (5)$$

This constitutes a time-varying transverse current that may scatter incident electromagnetic radiation as described by Eq. (1). When the scatterers are free electrons, this process is commonly referred to as Thomson scattering.[1]

The scattering process described above involves only the transverse motion of the free carriers. It can be seen from Eq. (3), however, that there is also a longitudinal force acting on each free carrier,

$$m^*\frac{dv_z}{dt}=q(E_z+v_xB_y)-\gamma m^* v_z. \quad (6)$$

The second term in Eq. (6) arises from the cross product of the induced transverse free-carrier velocity and the magnetic field of the incident and scattered fields. This force, which can be significant, may be written as

$$qv_xB_y=-\frac{2q^2n}{m^{*2}\omega c}\frac{1}{1+\gamma^2/\omega^2}\left[(i\mathcal{E}_1\mathcal{E}_2^* e^{2ikz_j}+\text{c.c.})-\frac{\gamma}{\omega}(|\mathcal{E}_1|^2-|\mathcal{E}_2|^2)\right]. \quad (7)$$

The first term in Eq. (7) describes a position-dependent ponderomotive force whereas the second term describes a radiation-pressure-like force due to free-carrier absorption. Note that the force described in Eq. (7) is independent of charge, i.e., its direction is the same whether the carrier is an electron or a hole. The ponderomotive force is similar to that encountered in free-electron laser theory.[3] Its effect is to spatially bunch the free carriers to form a grating with a period of $\lambda/2$. This grating scatters the incident field coherently, amplifying the scattered field and increasing the ponderomotive forces on the free carriers. This positive feedback can give rise to an exponential instability in both the scattered field intensity and the free-carrier grating amplitude. This amplification is a collective process as it is not describable in terms of a superposition of the evolution of the individual carriers, but involves the mutual evolution of each free carrier in the presence of the fields emitted by the ensemble. Hence the effects that we predict in this paper may be described as a form of *collective Thomson scattering*.

In addition to the above ponderomotive and absorption forces there is an additional longitudinal force arising from the electric field E_z. This describes space-charge forces arising from the free-carrier charge density

$$\rho_{\text{sc}}=e\sum_{j=1}^{N}\left[\delta(\mathbf{r}-\mathbf{r}_{h_j}(t))-\delta(\mathbf{r}-\mathbf{r}_{e_j}(t))\right].$$

We assume that the space-charge field is purely longitudinal, consistent with our one-dimensional description of the interaction, and that it is approximately periodic in z with a period $\lambda/2$, consistent with the periodic ponderomotive potential, allowing it to be expanded in a Fourier series,

$$E_z(z,t)=i\frac{en_e}{2k\epsilon}\sum_m\left[\frac{(\langle e^{-i2mkz}\rangle_e-\langle e^{-i2mkz}\rangle_h)e^{i2mkz}}{m}-\text{c.c.}\right],\quad m>0. \quad (8)$$

In deriving Eq. (8), the space-charge field E_z has been averaged over an interval of half a radiation wavelength centered at position z consistent with the SVEA. $\langle\cdots\rangle$ is thus an average over the free carriers contained within the interval and so is itself a function of both z and t. As noted previously, the ponderomotive forces acting upon the electrons and holes tend to bunch them about the same periodic positions. This tends to reduce the effects of charge separation and any consequent space-charge forces. It should be noted therefore that the collective Thomson scattering instability described in this paper is distinct from the Raman instability in laser-plasma interactions,[6,7] which produces simultaneous amplification of an electromagnetic probe field and a *space-charge wave*.

We now introduce the scaled independent variables $\bar{t}=\omega\rho t$, $\bar{z}=\omega n\rho z/c$, the scaled complex field amplitudes

$$A_1=\frac{2en}{\rho\omega m_e^* c}i\mathcal{E}_1,\quad A_2=\frac{2en}{\rho\omega m_e^* c}\mathcal{E}_2,$$

and scaled scattering rates $\bar{\gamma}_{e,h}=\gamma_{e,h}/\omega$. The scaled position variable $\theta_j(t)=2kz_j(t)$ is a phase variable describing the position of the jth free carrier with respect to the ponderomotive potential.

Thus we obtain

$$\frac{d\theta_{ej}}{d\bar{t}}=p_{ej}, \quad (9)$$

$$\frac{dp_{ej}}{d\bar{\bar{t}}} = -\frac{1}{1+\bar{\gamma}_e^2}(A_1A_2^* e^{i\theta_{ej}} + \text{c.c.}) + \frac{\bar{\gamma}_e}{1+\bar{\gamma}_e^2}(|A_1|^2 - |A_2|^2) - i\frac{2}{\rho}\sum_m \left[\frac{(\langle e^{-im\theta}\rangle_e - \langle e^{-im\theta}\rangle_h)e^{im\theta}}{m} - \text{c.c.}\right] - \frac{\bar{\gamma}_e}{\rho}p_{ej}, \tag{10}$$

$$\frac{d\theta_{hj}}{d\bar{\bar{t}}} = p_{hj}, \tag{11}$$

$$\frac{dp_{hj}}{d\bar{\bar{t}}} = -\frac{\alpha^2}{1+\bar{\gamma}_h^2}(A_1A_2^* e^{i\theta_{hj}} + \text{c.c.}) + \frac{\alpha^2\bar{\gamma}_h}{1+\bar{\gamma}_h^2}(|A_1|^2 - |A_2|^2) + i\frac{2\alpha}{\rho}\sum_m \left[\frac{(\langle e^{-im\theta}\rangle_e - \langle e^{-im\theta}\rangle_h)e^{im\theta}}{m} - \text{c.c.}\right] - \frac{\bar{\gamma}_h}{\rho}p_{hj}, \tag{12}$$

$$\frac{\partial A_1}{\partial \bar{\bar{z}}} + \frac{\partial A_1}{\partial \bar{\bar{t}}} = \left(\frac{\langle e^{-i\theta}\rangle_e}{1+i\bar{\gamma}_e} + \alpha\frac{\langle e^{-i\theta}\rangle_h}{1+i\bar{\gamma}_h}\right)A_2 - i\left(\frac{1}{1+i\bar{\gamma}_e} + \frac{\alpha}{1+i\bar{\gamma}_h}\right)A_1, \tag{13}$$

$$-\frac{\partial A_2}{\partial \bar{\bar{z}}} + \frac{\partial A_2}{\partial \bar{\bar{t}}} = -\left(\frac{\langle e^{i\theta}\rangle_e}{1+i\bar{\gamma}_e} + \alpha\frac{\langle e^{i\theta}\rangle_h}{1+i\bar{\gamma}_h}\right)A_1 - i\left(\frac{1}{1+i\bar{\gamma}_e} + \frac{\alpha}{1+i\bar{\gamma}_h}\right)A_2, \tag{14}$$

where

$$\rho = \frac{e^2 n_e}{2\epsilon_0 n^2 \omega^2 m_e^*} = \frac{\omega_{pe}^2}{2\omega^2},$$

$\omega_{pe} = \sqrt{e^2 n_e/\epsilon m_e^*}$ is the electron plasma frequency, $\epsilon = \epsilon_0 n^2$ is the background permittivity, and $\alpha = m_e^*/m_h^*$, the ratio of the free-carrier effective masses. It can now be seen that there are only four free parameters: ρ, α, and $\bar{\gamma}_{e,h}$. In deriving Eqs. (9)–(14), the pump and scattered fields are assumed to be averaged fields over an interval of half a radiation wavelength centered at scaled position $\bar{z}$. This is consistent with the SVEA and corresponds to an interval of $\theta \in [0,2\pi)$.

This set of coupled partial-differential equations describes the scattering of light by free carriers in a semiconductor when interband processes can be neglected. The form chosen in Eq. (6) for the damping is appropriate when the semiconductor is at low temperature. This can be seen from Eqs. (10) and (12) in the absence of any interaction. In this case, the equilibrium value of the momentum variables $p_{e,h}$ is zero, indicating a cold system. This ''cold system limit'' is valid so long as the rate of decay of any free-carrier grating structure due to thermal motion is much slower than the rate of growth of the grating structure due to collective instability.[4] This is true for the examples shown in this paper. A more detailed discussion of this requires a linear analysis of Eqs. (9)–(14).

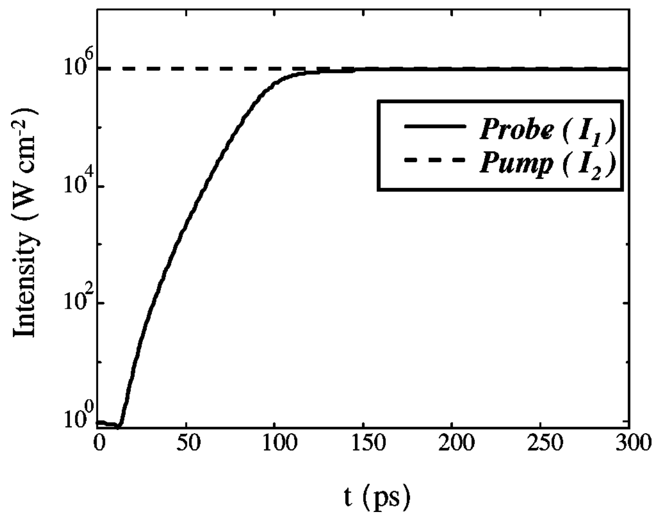

FIG. 2. Field intensities as functions of time at the right-hand edge of the sample ($z = l_s$).

We now use the above model to demonstrate collective scattering from free carriers in a sample of undoped InSb from the experimental schematic shown in Fig. 1. We assume a slab of InSb of thickness $l_s = 800$ μm irradiated by a pump beam with wavelength $\lambda = 10$ μm and intensity $I_2 = 1$ MW cm^{-2}, and a counterpropagating probe beam of the same wavelength but of much lower intensity $I_1 = 1$ W cm^{-2}. Both pump and probe are switched on at $t = 0$. In practice the experiment could also be performed with only a single (pump) beam, with the ''probe'' beam resulting from spontaneous emission due to inherent randomness in the electron/hole positions.

The scattering rates $\gamma_{e,h}$ encapsulate several processes. In particular they include carrier-carrier scattering, Auger scattering, and carrier-phonon scattering. As far as our study is concerned the main point is whether or not the bunching is robust enough to persist in the presence of such scattering. As such, we choose the scattering times for the electrons and holes to be 10 and 1 ps, respectively. At the sample temperature of ≈ 4 K this is a conservative estimate of the fastest electronic scattering time. The number density of free electrons and holes is assumed to be $n_e = n_h = 3.6 \times 10^{16}$ cm^{-3}. At 4 K this is much greater than the intrinsic carrier contribution so a photo-induced pumping of the population is envisaged. This adds the additional practical restriction that all collective processes occur on a time scale much smaller than the free-carrier recombination time of several nanoseconds. Taking the effective mass of the electrons and holes to be $m_e^* = 0.01m_0$ and $m_h^* = 0.18m_0$, respectively, these parameters correspond to scaled parameters $\bar{l}_s = 20$, $\alpha = 0.056$, $\rho = 0.01$, $\bar{\gamma}_e = 5 \times 10^{-4}$, $\bar{\gamma}_h = 5 \times 10^{-3}$, $|A_1| = 3.2 \times 10^{-4}$, and $|A_2| = 0.32$.

Figure 2 shows the results of a numerical solution of our

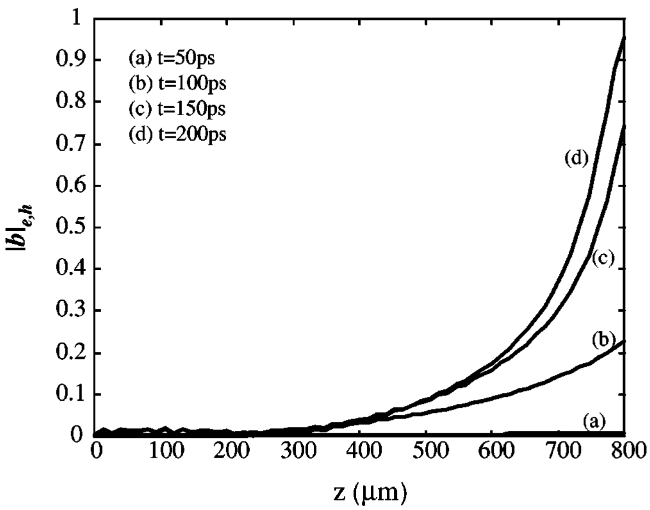

FIG. 3. Bunching parameter for electrons ($|b_e|$) and holes ($|b_h|$) as a function of z at different times: (a) $t=50$ ps, (b) $t=100$ ps, (c) $t=150$ ps, and (d) $t=200$ ps.

model. The pump and probe intensities are plotted at the right-hand face of the sample $[I_2(z=l_s,t),I_1(z=l_s,t)]$. The scattered probe intensity is seen to grow exponentially until $t\approx150$ ps where growth saturates and the intensity tends towards a steady-state value of $I_1\approx I_2$, indicating a reflectivity of almost 100%. The reason for this high reflectivity can be deduced from Fig. 3, which shows a plot of the modulus of the bunching parameter $b=\langle e^{-i\theta}\rangle$ for both electrons and holes as a function of position within the sample at four different times. The bunching parameter b is a measure of the spatial distribution of the free carriers on the wavelength scale. For uniformly distributed free carriers, $|b|=0$, and for perfectly bunched free carriers, where they have the same value of θ, then $|b|=1$. The behavior of $|b|$ shown in Fig. 3 for both holes and electrons indicates that the initially uniformly distributed electrons and holes become strongly and equally bunched on a scale of $\lambda/2$. The free carriers have therefore formed a strong grating that reflects the incident pump field. Furthermore, the strength of the grating, as measured by $|b|$, falls off quasiexponentially with distance from the front face of the sample. The fact that the bunching factors for the electrons and holes are equal is at first unexpected, given that the electrons have a significantly smaller effective mass than the holes ($\alpha\ll1$). This suggests that the electrons should bunch much faster than the holes. However, if $|b_e|\neq|b_h|$, the space-charge forces become nonzero and push the electrons and hole in opposite directions until $|b_e|=|b_h|$. The rate of growth of the electron bunching is therefore limited to the slower bunching of the holes via space charge.

We now discuss possible ways in which the phenomenon may be observed in a geometry similar to that shown in Fig. 1. An obvious method would be to measure the backscattered signal from the sample as a function of pump intensity. As the pump intensity increases the improved bunching of the free carriers is manifest as a superlinear increase in the backscattered signal. An alternative scheme would exploit diffraction from the free-carrier density grating of a second probe beam transverse to the pump-probe propagation direction. The z dependence of the grating strength could also be measured by scanning the diffracting probe along the length of the sample to provide a quantitative measure of the bunching similar to that of Fig. 3. If the pump were removed after strong bunching had been established, then both the reflectivity and the visibility of the diffraction pattern would be expected to decay. If the time scale of this decay is the same for both the reflectivity and the diffraction pattern, and is much longer than the decay time of any material nonlinear processes, then this would be evidence of the presence of the more persistent free-carrier density grating structure.

In conclusion, we have presented an analysis of an undoped semiconductor strongly pumped by a coherent electromagnetic radiation field using a classical, one-dimensional model in which free-carrier scattering was described phenomenologically using a relaxation time approximation. The free-carrier dynamics and the incident and scattered fields evolve self-consistently and produce a collective instability that simultaneously generates a strong free-carrier grating and a coherently backscattered radiation field. A possible low-temperature experiment using InSb was described which suggests that this phenomenon should be observable.

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