Virtual Photoconductivity

E. Yablonovitch, J. P. Heritage, D. E. Aspnes, and Y. Yafet
Bell Communications Research, Navesink Research Center, Red Bank, New Jersey 07701-7040
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We show that a semiconductor photodiode, or photoconductor, will exhibit a strong reactive response to optical radiation tuned to the transparent region just below the band gap. This response is due to the excitation of a virtual electron-hole gas, readily polarized by the dc electric field, which can contribute a major change (>1) in the low-frequency dielectric constant. For ultrahigh-speed signals, this nondissipative photoresponse can become similar to conventional dissipative photoresponse by real carriers excited above the band gap. The influence of this reactive response on zero-point electromagnetic fluctuations may aid the laboratory detection of Unruh-Davies-Fulling-DeWitt radiation.

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Semiconductor photodiodes typically operate at optical frequencies ω_0 above the semiconductor band gap, $\omega_{cv} \equiv E_{cv}/\hbar$. If an incident beam is intensity modulated at a frequency ω, the output photocurrent will have an in-phase (dissipative) component $\mathrm{Re}\{J(\omega)\}$. If ω_0 is lowered below ω_{cv}, the probability of real (dissipative) transitions goes sharply to zero, vanishing exponentially in the Urbach absorption tail. Since the photocurrent tracks the absorption coefficient, $\mathrm{Re}\{J(\omega)\}$ also vanishes sharply, as is well understood in the conventional theory of semiconductor photoresponse.

In this paper we show that the corresponding out-of-phase (reactive) component, $\mathrm{Im}\{J(\omega)\}$, does *not* vanish as ω_0 is lowered below ω_{cv}. Indeed, for very-high-speed signals the nondissipative photoresponse of virtual carriers excited below the band edge can become similar to the conventional dissipative photoresponse by real carriers excited above the band edge. In effect, the virtual electron-hole pairs are readily polarized by the dc field of the photodetector, leading to large changes in the dielectric response of the material.

A closely related virtual excitation process was recently analyzed, by Chemla, Miller, and Schmidt-Rink[1] and by Yamanishi,[2] in the context of excitons in quantum wells electrically biased perpendicular to the layers. The excitation of a hole at one edge of a valence-band quantum well and an electron at the opposite edge of the corresponding conduction-band quantum well creates an electric dipole. Chemla, Miller, and Schmidt-Rink and Yamanishi studied the virtual excitation of such quantum confined excitons. As the quantum well is made wider and wider, the dipole obviously becomes larger and larger. The resulting polarization can be expressed as a nonlinear optical susceptibility $\chi^{(3)}(0,0,-\omega_0,\omega_0)$. We show that $\chi^{(3)}(0,0,-\omega_0,\omega_0)$ becomes largest when no potential barriers exist to block carrier motion. In other words, $\chi^{(3)}(0,0,-\omega_0,\omega_0)$ is largest for bulk semiconductor material. Quantum wells are not necessary, but if they are used their potential barriers should be parallel to the applied field so as not to impede the polarization of virtual carriers.

We consider the photoconductive response of a photodiode driven by light waves in the nonabsorbing, near-band-edge region. A particularly simple and physically appealing picture emerges, as illustrated in Fig. 1. Photons above the band edge produce free electrons and free holes. Photons below the band edge produce virtual electron-hole pairs. These are not quite free of one another, but behave as if they were bound by the detuning energy, $\Delta \equiv E_{cv} - \hbar\omega_0$. Since Δ can be made very small, the electron-hole pairs can appear weakly bound and therefore highly polarizable by the dc electric field of the photoconductor. Bulk excitons, which occur in semiconductor absorption spectra at low temperatures, are also highly polarizable and contribute to the virtual response. The strong photoconductive response is due to near-free virtual electrons and holes, and to virtual excitons if they are present.

In this spirit we write the low-frequency polarizability of virtual carriers using a simple heuristic model that

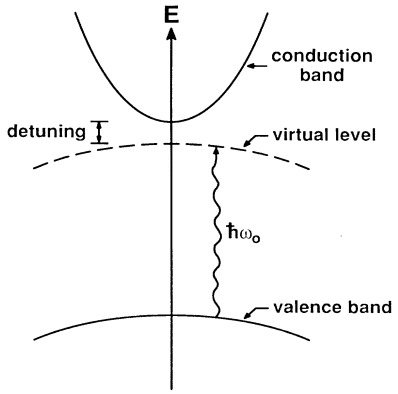

FIG. 1. The optical field creates a virtual level near the conduction-band edge. The virtual occupation probability is the modulus squared of the ratio of the dipole matrix element $|eX_{cv}E(\omega_0)|$ divided by the detuning energy $(E_{cv} - \hbar\omega_0)$. These virtual electron-hole pairs respond to electric bias as if they were bound together by the small detuning energy. The result is a large reactive component of photocurrent.

will be justified below. The standard expression for the polarizability of N virtual electron-hole pairs per unit volume with an effective binding energy Δ is

$$\chi(0)=\frac{e^2N}{m_{\rm eff}\{(\Delta/\hbar)^2-\omega^2\}}, \tag{1}$$

where e is the electronic charge and $m_{\rm eff}$ is the reduced mass. We assume $\omega\ll\Delta/\hbar$, and so drop ω from Eq. (1).

According to quantum perturbation theory, the virtual occupation probability of a level is the modulus squared of the ratio of the Rabi matrix element $|eX_{cv}E(\omega_0)|$ to the detuning energy $[E_{cv}(k)-\hbar\omega_0]$. We compute N by multiplying this modulus squared by the density of states in k space. The dc polarizability $\chi(0)$ becomes

$$\chi(0)\sim\frac{2}{(2\pi)^3}\int_{\rm BZ}d^3k\,\frac{e^2|eX_{cv}E(\omega_0)|^2\hbar^2}{m_{\rm eff}[E_{cv}(k)-i\Gamma-\hbar\omega_0]^4}, \tag{2}$$

where Γ is a phenomenological damping energy and $E_{cv}(k)$ is the valence-to-conduction band energy at the point k in the Brillouin zone (BZ). As long as the modulation frequency $\omega<\Delta/\hbar$, Eq. (2) will adequately describe the semiconductor response. In this paper we will attempt to justify Eqs. (1) and (2), which describe a simple heuristic model of virtual electron-hole pairs behaving as bound carriers.

There is an inverse process called electroreflectance (ER), which has been much studied[3] in the past. In ER, a dc electric field changes the *optical* dielectric susceptibility in the vicinity of the band gap, or at other critical points in the Brillouin zone. We now show that the expressions developed to describe ER will also describe virtual photoconductivity, with an appropriate permutation of variables. The change in optical polarization $P^{(3)}(\omega_0)$ due to a dc field can be derived from a free-energy function $F^{(3)}$:

$$P^{(3)}(\omega_0)=\frac{\partial F^{(3)}}{\partial E(\omega_0)}=\chi^{(3)}(-\omega_0,\omega_0,0,0)\frac{|E(0)|^2}{2}E(\omega_0), \tag{3}$$

where, following the usual nonlinear optical conventions, the total electric field is written

$$E(t)=E(0)+E(\omega_0)e^{-i\omega_0t}+E(-\omega_0)e^{i\omega_0t}.$$

The factor $\frac{1}{2}$ arises because the amplitude and root-mean-square values of the zero-frequency field components $E(0)$ are identical. The third-order dielectric free-energy function $F^{(3)}$ is therefore

$$F^{(3)}=\chi^{(3)}(-\omega_0,\omega_0,0,0)E(\omega_0)E(-\omega_0)\frac{|E(0)|^2}{2}, \tag{4}$$

from which the corresponding nonlinear dc polarization follows as

$$P^{(3)}(0)=\frac{\partial F^{(3)}}{\partial E(0)}=\chi^{(3)}(0,0,-\omega_0,\omega_0)E(-\omega_0)E(\omega_0)E(0). \tag{5}$$

The point is the two susceptibilities $\chi^{(3)}(0,0,-\omega_0,\omega_0)$ and $\chi^{(3)}(-\omega_0,\omega_0,0,0)$ in Eqs. (3) and (5) are the same since they are derived from the same free energy and differ by only a permutation of frequency arguments. We have the choice then of calculating the effect of a light wave on a dc dielectric property, or the effect of a dc field on the optical susceptibility. We choose the latter, invoking a previous derivation of low-field electroreflectance.[3] Using Eqs. (3) and (4), we then invert the calculation to obtain the virtual photoconductivity, Eq. (5).

The conventional linear optical susceptibility $\chi^{(1)}(\omega_0)$ of a semiconductor is given by

$$\chi^{(1)}(\omega_0)=\frac{2}{(2\pi)^3}\int_{\rm BZ}d^3k\,\frac{|eX_{cv}|^2}{E_{cv}(k)-i\Gamma-\hbar\omega_0}. \tag{6}$$

When ω_0 is tuned near ω_{cv} the denominator becomes near resonant and $\chi^{(1)}$ becomes quite sensitive to the exact values of $E_{cv}(k)$ and ω_0. It is well known in band theory[4] that a dc electric field produces a monotonic, linear change of wave vector with time t: $\mathbf{k}\rightarrow\mathbf{k}(t)=\mathbf{k}+e\mathbf{E}(0)t/\hbar$. Such a time-dependent k vector has a pronounced effect on the contribution of $E_{cv}(k(t))$ to the near-resonant denominator of Eq. (6). If $|E(0)|$ is so weak that the field-induced kinetic energy $e^2|E(0)|^2t^2/2m_{\rm eff}$ is less than the phenomenological damping energy Γ, then spectral line-shape theory gives the famous third-derivative line shape[3] of ER, i.e., a $\chi^{(3)}$ proportional to $\partial^3\chi^{(1)}/\partial\omega_0^3$. This may also be written[3]

$$\chi^{(3)}(-\omega_0,\omega_0,0,0)=\frac{1}{(2\pi)^3}\int_{\rm BZ}d^3k\,\frac{e^2|eX_{cv}|^2\hbar^2}{m_{\rm eff}[E_{cv}(k)-i\Gamma-\hbar\omega_0]^4}. \tag{7}$$

This is similar to Eq. (2), essentially justifying our original heuristic picture that virtual electron-hole pairs respond as if they were bound together by the detuning energy.

We now rewrite and simplify Eq. (7). Using Fermi's "golden rule," the matrix element and density of states $\rho(E)$ can be linked to the optical-absorption coefficient $\alpha(\omega_0)$ as follows:

$$2\pi\omega_0|eX_{cv}E(\omega_0)|^2\rho(E)=\alpha(E)cn|E(\omega_0)|^2/2\pi,$$

where E measures the kinetic energy from the bottom of the band and n is the optical refractive index. The formula for

$\chi^{(3)}(0,0,-\omega_0,\omega_0)$ becomes

$$\chi^{(3)}(0,0,-\omega_0,\omega_0)=\frac{1}{8\pi^2}\int_0^\infty \frac{e^2\hbar^3 cn}{E_{cv}m_{\rm eff}}\frac{\alpha(E)dE}{(E+\Delta)^4}. \quad (8)$$

By expressing $\chi^{(3)}$ in terms of the experimentally determinable absorption coefficient $\alpha(E)$, Coulomb effects on the matrix elements are mostly accounted for. Since $\alpha(E)$ is known, Eq. (8) can be integrated numerically. For direct-gap semiconductors such as GaAs, $\alpha(E)$ is zero below the band edge and approximately constant at $\alpha\approx 10^4$/cm above the band edge. Using these values Eq. (8) can be expressed in closed form:

$$\chi^{(3)}(0,0,-\omega_0,\omega_0)=\frac{1}{32\pi^2}\frac{e^2\alpha cn}{E_{cv}m_{\rm eff}}\left(\frac{\hbar}{\Delta}\right)^3 \approx \frac{1.1\times10^{-3}}{[\Delta\ (\text{meV})]^3}\ \text{esu}. \quad (9)$$

The ratio of optical Rabi matrix element[5] to detuning, $|eX_{cv}E(\omega_0)|/\Delta$, must be less than 1 in order to satisfy the perturbation limit for the optical field. As optical fields approach this limit and for a detuning energy $\Delta=12$ meV, the nonlinear zero-frequency susceptibility $\chi^{(3)}(0,0,-\omega_0,\omega_0)|E(\omega_0)|^2$ can become $\approx\frac{1}{4}$, which implies a change in low-frequency dielectric constant $\sim\pi$.

In Ref. 1, the quantum-confined-exciton (QCE) susceptibility $\chi^{(3)}(0,0,-\omega_0,\omega_0)$ was evaluated at a detuning energy of 42 meV. At the same detuning energy, the bulk $\chi^{(3)}(0,0,-\omega_0,\omega_0)$ due to virtual photoconductivity in Eq. (9) is 1.5×10^{-8} esu which is $\sim$15 times larger than the nonlinearity deduced[1] for the QCE case. This is due to the inherently limited electric polarization achievable for fields perpendicular to a narrow quantum well, as mentioned earlier. Photon absorption is *absent* in either instance, at least in the perturbative limit where the concept of a $\chi^{(3)}$ makes sense.

We now compare the sensitivity of virtual photoconductivity with that of conventional dissipative photoconductivity. A conventional photodiode responds to the zero-frequency dc bias field and to an amplitude-modulated optical wave, which may be regarded as the superposition of two closely spaced optical frequencies. Therefore, a conventional absorptive photodiode response can also be described in terms of a $\chi^{(3)}$. The difficulty is that photoconductive response will usually be described by material-dependent properties. Fortunately, in the limit of fast modulation frequency ω, many of these drop out. For example, at high ω we can neglect the momentum relaxation time, carrier recombination lifetime, and the carrier sweepout rate. This leads to the following simple $\chi^{(3)}$ for conventional absorptive photoconductivity:

$$\chi^{(3)}=\frac{1}{8\pi}\frac{e^2\alpha cn}{E_{cv}m_{\rm eff}}\left(\frac{i}{\omega}\right)^3. \quad (10)$$

A direct comparison of Eqs. (9) and (10) shows that the ratio of conventional dissipative photoconductivity to reactive virtual photoconductivity is simply $4\pi(\Delta/\hbar\omega)^3$. As the optical modulation sidebands approach the band edge, dissipation will not set in until $\hbar\omega\gtrsim\Delta$. For detuning $\sim\hbar\omega$, the two types of photoconductivity can be of similar magnitude, and one will merge smoothly into the other as the photon energy $\hbar\omega_0$ is tuned into the band gap E_{cv}. For extremely fast modulation frequencies (in the THz region), virtual photoconductivity would have a role to play. Furthermore, nondissipation of the light wave permits traveling-wave devices in which the response would build up with propagation path length.

At low temperatures[6] a sharp, distinct excitonic feature is present below the absorption edge in bulk material. The same feature appears right at[7] the absorption edge in room-temperature multiple-quantum-well structures (MQWS). It is well known in ER[3] that the exciton response is proportional to a first derivative of the linear spectrum rather than the third derivative. This is due to the simple field-induced Stark shift in exciton energy $E_{\rm ex}\rightarrow E_{\rm ex}-\kappa|E(0)|^2/2$, where κ is the exciton polarizability, as opposed to the more complex field-induced temporal variation contained in $E_{cv}\{\mathbf{k}\}\rightarrow E_{cv}\{\mathbf{k}+e\mathbf{E}(0)t/\hbar\}$. In the exciton case the susceptibility $\chi(\omega_0-E_{\rm ex}/\hbar)$ can then be expanded as

$$\chi^{(1)}+\{\partial\chi^{(1)}/\partial\omega_0\}\{\kappa|E(0)|^2/2\hbar\}.$$

Then similarly to Eqs. (6) and (7),

$$\chi^{(3)}(-\omega_0,\omega_0,0,0)=\kappa\int_{-\infty}^{\infty}\frac{|eX_{cv}|^2}{(E+\Delta)^2}|\phi(0)|^2 g(E)dE, \quad (11)$$

where $g(E)$ is a normalized exciton line-shape function, $|\phi(0)|^2$ is the probability density per unit volume of the exciton wave function at its origin, $|\phi(0)|^2 g(E)$ is a density of states per unit volume, E is the energy measured from $E_{\rm ex}$, and Δ is now the exciton detuning $(E_{\rm ex}-\hbar\omega_0)$. Implicit in Eq. (11) is the virtual-exciton phase-space[8] occupation probability $|eX_{cv}E(\omega_0)|^2/\Delta^2$ that was emphasized in Ref. 1. Indeed, Eq. (11) has the same physical meaning as Eq. (2): (occupation probability)$\times$polarizability. Once again Fermi's "golden rule" converts from matrix elements to absorption coefficients:

$$2\pi\omega_0|eX_{cv}\phi(0)|^2 g(E)=\alpha(E)cn/2\pi.$$

Equation (11) can be integrated numerically using the experimental excitonic absorption spectrum $\alpha(E)$ if known, or analytically if Δ is much greater than the exciton linewidth. Using the approximations[9] that $\kappa\sim e^2\hbar^2/m_{\rm eff}E_b^2$ and that the exciton oscillator strength is proportional to binding energy, $\int\alpha(E)dE\approx\alpha E_b$,

$$\chi^{(3)}(0,0,-\omega_0,\omega_0)\approx\frac{1}{4\pi^2}\frac{e^2\alpha cn}{E_{\rm ex}m_{\rm eff}}\left(\frac{\hbar^3}{\Delta^2 E_b}\right), \quad (12)$$

where E_b is the exciton binding energy. Equation (12) should describe excitons both in bulk material and in MQWS biased parallel to the layers. Perpendicularly biased MQWS excitons were treated in Refs. 1 and 2.

A comparison of Eqs. (9) and (12) shows that the virtual-excitonic response can be $\sim 8\Delta/E_b$ bigger than the virtual-free-carrier response. Numerical integration of Eq. (11) over the measured[7] MQWS excitonic absorption spectrum yields a response ratio $\sim\Delta/(3\text{ meV})$. In practice, the apparent superior response of virtual excitons is rather fragile. Thermally induced mixing of the exciton and continuum states reduces the excitonic oscillator strength at room temperature. Whether ER in room-temperature MQWS has first derivative (excitonic) *or* third derivative (near-free-carrier) behavior is still being checked[10,11] experimentally. In any case, strong optical matrix elements $|eX_{cv}E(\omega_0)| \gtrsim E_b$ (where E_b is now the difference energy between the exciton and the continuum threshold) will destroy the exciton, invalidating Eq. (12) and leaving only the virtual-free-carrier photoconductivity, Eq. (9).

In practice, changes in the low-frequency dielectric constant even larger than Eq. (9) would require stronger optical fields, but a dissipative nonvirtual response might occur. A sum to all orders in the optical matrix element squared, $|eX_{cv}E(\omega_0)|^2$, would be highly desirable to settle this question.

Possible applications of virtual photoconductivity include both ultrafast optical signal detection and the creation of large, sudden changes in the low-frequency dielectric constant of semiconductors. It was recently proposed[12] that zero-point electromagnetic field fluctuations would respond nonadiabatically if the change in dielectric constant were large and fast enough. The resulting microwave photons are called[12] Unruh-Davies-Fulling-DeWitt (UDFD) radiation. The optical excitation must be very clean and nondissipative, since dissipation would be linked to noise fluctuations which could overwhelm the UDFD radiation.

[1]D. S. Chemla, D. A. B. Miller, and S. Schmidt-Rink, Phys. Rev. Lett. **59**, 1018 (1987).

[2]M. Yamanishi, Phys. Rev. Lett. **59**, 1014 (1987).

[3]D. E. Aspnes, Surf. Sci. **37**, 418 (1973).

[4]C. Kittel, *Quantum Theory of Solids* (Wiley, New York, 1963).

[5]For $m_{\text{eff}}=0.1m$ in $\mathbf{k}\cdot\mathbf{P}$ band theory the matrix element $|X_{cv}| \approx 6.4$ Å.

[6]D. D. Sell, Phys. Rev. B **6**, 3750 (1972).

[7]D. S. Chemla, D. A. B. Miller, P. W. Smith, A. C. Gossard, and W. Wiegmann, IEEE J. Quantum. Electron. **20**, 265 (1989).

[8]The alternate matrix element $|eX_{cv}\phi(0)E(\omega_0)|^2$ merely has the effect of normalizing per unit volume rather than per phase-space cell. R. J. Elliot, Phys. Rev. **108**, 1384 (1957).

[9]Actually κ for the bulk exciton is $\frac{9}{8}$ larger. E. Merzbacher, *Quantum Mechanics* (Wiley, New York, 1966).

[10]B. V. Shanabrook, O. J. Glembocki, and W. T. Beard, Phys. Rev. B **35**, 2540 (1987).

[11]H. Shen, S. H. Pan, F. H. Pollak, and R. N. Sacks, Phys. Rev. B **37**, 10919 (1987).

[12]E. Yablonovitch, Phys. Rev. Lett. **62**, 1742 (1989).