Two-level approach to saturation properties in semiconductor materials

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Saturation properties of absorption, dispersion, and mixing susceptibilities in semiconductors have been described using the density matrix approach. This model, which takes into account both intraband thermalization and interband relaxation including Auger recombination, allows for numerical computation of the susceptibilities whatever the temperature and laser intensity are. As a simplification, an equivalent two-level system is derived which represents very well absorption and mixing susceptibilities for any laser wavelength, dispersion being correctly described by this model only when the pump frequency is close to or smaller than the band-gap energy. Such an equivalence is very easy to handle and allows for great simplification when the susceptibilities are introduced into the propagation equations.

I. INTRODUCTION

Due to their large nonlinearities semiconductor materials are very attractive for applications such as bistability or phase conjugation. As a consequence, nonlinear processes due to interband absorption in semiconductors have received great attention, since lasers can be used to saturate the electron-hole pair creation. Absorption was first phenomenologically modeled by using a two-level system.[1] More recently, the band structure was introduced by Wherret and Higgins[2] in the "direct saturation" model in which the semiconductor is described by a superposition of independent two-level systems of different resonance frequencies. However, this description, which does not take intraband relaxation into account, could not reproduce the intensity dependence of the third-order susceptibility of a semiconductor-doped glass.[3] Another scheme, named the "Burnstein-Moss" model, also used by Wherret and Higgins[2] takes intraband relaxation into account. Using this theory very simple expressions can be obtained for the semiconductor susceptibilities, but only in the limit of very small temperatures. Another model was used by Miller *et al.*[4] which describes the nonlinear refractive index of semiconductors at room temperature. Nevertheless, as it involves Boltzmann statistics in order to describe the free-carrier quasiequilibrium, the simple expressions derived from this model cannot be used at high medium saturation. In the case of semiconductor lasers a scheme has been developed which correctly models matter-radiation interaction in the semiconductor whatever the temperature is.[5] Unfortunately, this method, which needs some numerical integrations, uses a series development for the induced polarization. This approximation, which is perfectly correct in the case of semiconductor lasers, cannot be used for strong saturation of absorption in a pure semiconductor.

In this paper, we use a density matrix approach derived from Ref. 5 in order to model strong light-matter interaction in semiconductors. As the susceptibilities derived from this theory depend implicitly on the pump intensity, the paper presents a model using an equivalent two-level system for the description of the intensity saturation of the interband absorption. Indeed, the quantitative description of absorptive, dispersive, and mixing properties is much easier to obtain when using this equivalence. Section II describes the general formalism of our model, which takes intraband thermalization and interband relaxation, including Auger recombination, into account. This section also details the case of the saturation of absorption and dispersion by a monochromatic wave and that of degenerate or nearly degenerate four-wave mixing processes. Section III extends results for the two-level susceptibilities[6] in the case in which Auger recombination is taken into account. In Sec. IV the equivalence between the semiconductor and the two-level system is defined and a comparison is made between susceptibilities calculated by the "exact" and "two-level" models. This section also includes a discussion which demonstrates that absorption and mixing susceptibilities are always well described by using the two-level approach, whereas the equivalence is valid for dispersion only when the pump frequency is near or below that of the semiconductor band gap. An instruction guide to the equivalence is also presented which shows that this technique is very simple to handle. Finally, the conclusion summarizes the paper and discusses possible applications of the model.

II. SEMICONDUCTOR MODEL

A. General formalism

The semiconductor is described in reciprocal space by two parabolic valence and conduction bands. For the wave vector k in the first Brillouin zone, the energies in the conduction and valence bands are

$$E_c(k) = \frac{\hbar^2 k^2}{2m_c} + E_g , \tag{1a}$$

$$E_v(k) = -\frac{\hbar^2 k^2}{2m_v} , \tag{1b}$$

where m_c and m_v are the effective masses and E_g the

gap energy.

Matter-radiation interaction inside the semiconductor is described by using the density matrix formalism for electrons[5]

$$i\hbar\frac{\partial\rho}{\partial t} = \left[\frac{\partial\rho}{\partial t}\right]_{\text{stim}} + i\hbar\left[\frac{\partial\rho}{\partial t}\right]_{\text{int}} + i\hbar\left[\frac{\partial\rho}{\partial t}\right]_{\text{spon}} + i\hbar\left[\frac{\partial\rho}{\partial t}\right]_{\text{Auger}}$$

$$= (H_0 - d\mathscr{E}, \rho) - \frac{i\hbar}{2}[(\rho - \tilde{\rho})\Gamma + \Gamma(\rho - \tilde{\rho})] - \frac{i\hbar}{2}[(\rho - \rho_0)\gamma + \gamma(\rho - \rho_0)] - \frac{i\hbar}{2}(\rho\mathscr{A} + \mathscr{A}\rho) . \tag{2}$$

In this expression H_0 is the unperturbed Hamiltonian of the semiconductor, the corresponding energies being given by Eqs. (1), d is the dipole moment of an electron-hole pair,[7] and $\mathscr{E}$ is the electromagnetic field. $\tilde{\rho}$ is the quasiequilibrium distribution function reached through the intraband relaxation when the electromagnetic field is present. As the intraband relaxation is much faster than any other relaxation process,[8] the quasiequilibrium is described by Fermi-Dirac statistics:

$$\tilde{\rho}_c(k,\phi_c) = \frac{1}{1 + \exp\{[E_c(k) - \phi_c]/k_B T\}} , \tag{3a}$$

$$\tilde{\rho}_v(k,\phi_v) = \frac{1}{1 + \exp\{[E_v(k) - \phi_v]/k_B T\}} , \tag{3b}$$

where k_B is the Boltzmann constant, T the temperature, and ϕ_c and ϕ_v the quasi-Fermi-levels in the conduction and valence bands, respectively. As the Fermi-Dirac statistics are used rather than the Maxwell-Boltzmann statistics used in Ref. 4, the present calculations are valid whatever the field intensity is. ρ_0 is the distribution function at thermal equilibrium without any perturbing electromagnetic field. For the large-gap semiconductors considered in this paper all the electrons are supposed to be in the valence band and, therefore, $\rho_{0c}(k) = 0$ and $\rho_{0v}(k) = 1$. Γ, γ, and $\mathscr{A}$ are the operators describing the intraband relaxation, the spontaneous emission, and the Auger recombination, respectively. Under the adiabatic approximation the operators Γ, γ, and $\mathscr{A}$ have only diagonal elements. Assuming that the relaxation processes are k independent allows us to define the intraband relaxation time $\tau = \Gamma_{kk}^{-1}$, the spontaneous emission lifetime $T_1 = \gamma_{kk}^{-1}$, and the Auger relaxation time $A^{-1}N^{-2}$ (Ref. 9) where A is the Auger constant and

$$N = \frac{1}{\pi^2} \int_0^\infty k^2 dk\, \tilde{\rho}_c(k,\phi_c)$$

$$= \frac{1}{\pi^2} \int_0^\infty k^2 dk[1 - \tilde{\rho}_v(k,\phi_v)]$$

is the free-carrier density. The transverse relaxation time of the electron-hole dipole is

$$T_2 = \left[\frac{1}{\tau} + \frac{1}{T_1} + AN^2\right]^{-1} .$$

Using such a formalism the polarization is

$$P(t) = \text{Tr}(\rho d) . \tag{4}$$

If the electromagnetic field can be written

$$\mathscr{E}(t) = E(t)\exp(-i\omega t) + \text{c.c.} , \tag{5}$$

where $E(t)$ is slowly varying in time, it is possible to use the rotating-wave approximation[10] in the resolution of Eqs. (2). Moreover, as the thermalization to quasiequilibrium induced by intraband relaxation is reached at a time which is much faster than any other time constant involved in this problem, the intraband relaxation terms are eliminated by replacing ρ by its quasiequilibrium value $\tilde{\rho}$ in Eqs. (2). For the same reason the coherence follows the slowly varying field envelope $E(t)$ and the adiabatic approximation can be used.[11]

As a consequence, integration of Eq. (2) and the use of the normalized variable $\Delta = \{\omega - [E_c(k) - E_v(k)]/\hbar\}T_2$ then gives, for the polarization,

$$P(t) = \frac{cn\sigma}{8\pi^3\omega}\left[\frac{2m}{\hbar T_2}\right]^{3/2}$$

$$\times \int_{-\infty}^{\Delta_0} (-\Delta + i)[\tilde{\rho}_v(\Delta,\phi_v) - \tilde{\rho}_c(\Delta,\phi_c)]g(\Delta)$$

$$\times (\Delta_0 - \Delta)^{1/2} d\Delta\, E(t)\exp(-i\omega t) + \text{c.c.} \tag{6}$$

In this expression $m = (m_c^{-1} + m_v^{-1})^{-1}$ is the reduced mass, $\sigma = 4\pi\omega d_{vc}^2 T_2/\hbar cn$ is the absorption cross section of the electron-hole pair, n is the refractive index, d_{vc} is the electron-hole pair dipole moment which is assumed to be k independent, and $\Delta_0 = (\omega - E_g/\hbar)T_2$ is the normalized laser-band-gap detuning. $g(\Delta) = 1/(1 + \Delta^2)$ is the Lorentzian absorption profile of the electron-hole pair of wave vector k.

The quasi-Fermi-levels $\phi_c(t)$ and $\phi_v(t)$ fixing the value of the polarization are determined through the population density matrix equation

$$T_1 \frac{dN}{dt} = \frac{1}{4\pi^2} \left[\frac{2m}{\hbar T_2} \right]^{3/2} \left| \frac{E(t)}{E_S} \right|^2$$

$$\times \int_{-\infty}^{\Delta_0} [\tilde{\rho}_v(\Delta,\phi_v) - \tilde{\rho}_c(\Delta,\phi_c)]g(\Delta)$$

$$\times (\Delta_0 - \Delta)^{1/2} d\Delta - (1 + AT_1 N^2)N \qquad (7)$$

and the charge conservation condition

$$\int_{-\infty}^{\Delta_0} [\tilde{\rho}_c(\Delta,\phi_c) + \tilde{\rho}_v(\Delta,\phi_v) - 1](\Delta_0 - \Delta)^{1/2} d\Delta = 0 . \qquad (8)$$

In Eq. (7) $|E_s|^2 = \hbar^2/(4d_{vc}^2 T_1 T_2)$ is the saturation intensity of the electron-hole pair.

The exponential decay of the population relaxation assumed in our model gives a Lorentzian spectral weight function. Using such a spectral profile leads to an unbounded phase for the polarization. This drawback can be circumvented if we take the Gaussian decay in the wings of the line into account. Indeed, the use of the screened potential in the statistical theory of line broadening by electron-electron scattering results in a Gaussian decay having a linewidth 10–40 times larger than the Lorentzian profile.[12,13] Therefore, the line profile used is $g(\Delta) = [\exp(-\Delta^2/\Delta_1^2)]/(1+\Delta^2)$ rather than $1/(1+\Delta^2)$.

B. Absorption and dispersion saturation of a monochromatic wave

Under continuous or long pulse duration illumination of the semiconductor by a monochromatic wave $E(t) = E(\omega)$, Eq. (7) is integrated in the steady-state approximation. The free-carrier density is, therefore, defined by

$$N_0(1 + AT_1 N^2) = \frac{1}{4\pi^2} \left[\frac{2m}{\hbar T_2} \right]^{3/2} \left| \frac{E(\omega)}{E_s} \right|^2$$

$$\times \int_{-\infty}^{\Delta_0} [\tilde{\rho}_0(\Delta,\phi_v) - \tilde{\rho}_c(\Delta,\phi_c)]$$

$$\times g(\Delta)(\Delta_0 - \Delta)^{1/2} d\Delta . \qquad (9)$$

The quasiequilibrium Fermi levels $\phi_c(|E/E_s|^2)$ and $\phi_v(|E/E_s|^2)$ can, then, easily be determined by numerically solving Eqs. (8) and (9). Figure 1 shows the intensity dependence of the normalized quasi-Fermi-level positions $\Delta_v = T_2\phi_v(|E/E_s|^2)/\hbar$ and $\Delta_c = [\omega - \phi_c(|E/E_s|^2)/\hbar]T_2$ for $\Delta_0 = 1$. The solid and dashed curves correspond to no Auger recombination and relatively strong Auger recombination $(AT_1 = 1.5 \times 10^{-35}$ cm$^6)$, respectively. Both curves have the same limits Δ_v^∞ and Δ_c^∞ when the laser intensity is increased towards infinity. As expected, the saturation of the semiconductor absorption is reached at higher intensities when Auger recombination is taken into account. Let us underline that this ω frequency saturation of the semiconductor absorption corresponds, in fact, to a band-filling-induced equilibrium between gain for frequencies lower than ω and losses for frequencies higher than ω.

The knowledge of $\phi_{v,c}(|E/E_s|^2)$ allows us to calculate the susceptibility at frequency ω;
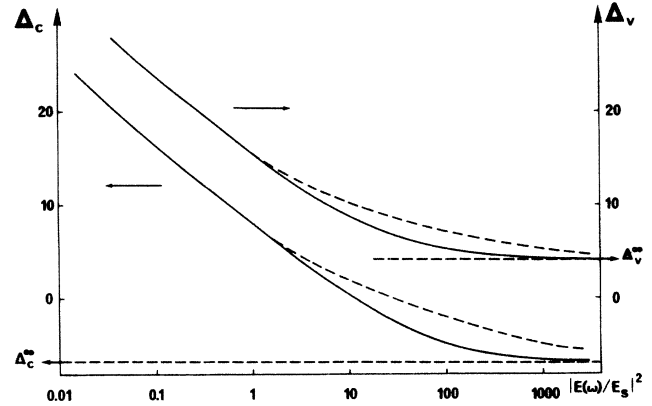

FIG. 1. Intensity dependence of the normalized quasi-Fermi-level position $(\Delta_0 = 1)$: solid lines, without Auger recombination; dashed lines, with Auger recombination $(AT_1 = 1.5 \times 10^{-35}$ cm$^6)$.

$$\chi(\omega) = \frac{cn}{8\pi^3\omega} \left[\frac{2m}{\hbar T_2} \right]^{3/2}$$

$$\times \int_{-\infty}^{\Delta_0} (-\Delta + i)[\tilde{\rho}_v(\Delta,\phi_v) - \tilde{\rho}_c(\Delta,\phi_c)]$$

$$\times g(\Delta)(\Delta_0 - \Delta)^{1/2} d\Delta . \qquad (10)$$

In this expression the intensity is a hidden parameter which enters through ϕ_v and ϕ_c.

C. Degenerate or nearly degenerate four-wave mixing

In this case the complex field amplitude is $E(t) = E(\omega) + E^-(\omega - \delta\omega)\exp(i\delta\omega t) + E^+(\omega + \delta\omega)\exp(-i\delta\omega t)$, where the pump-probe detuning $\delta\omega$ equals zero in the case of degenerate four-wave mixing. In the case of nearly degenerate four-wave mixing the field modulation period $2\pi/\delta\omega$ is assumed to be much larger than the intraband relaxation time τ $(\delta\omega\tau \ll 1)$. In the parametric approximation, the probe and conjugate amplitudes $E^-(\omega - \delta\omega)$ and $E^+(\omega + \delta\omega)$ are much smaller than the pump field $E(\omega)$ so that one can consider that the free-carrier density at quasiequilibrium is slightly modulated at the low frequency $\delta\omega$. The carrier densities in the conduction and valence bands N_c and N_v, which are governed by the energy of the corresponding quasi-Fermi-levels, are given by the following relations:

$$N_c = N_0 + \left[\frac{\partial N_c}{\partial \phi_c} \right]_{N_c = N_0} \Delta\phi_c \exp(i\delta\omega t) + \text{c.c.} , \qquad (11a)$$

$$N_v = N_0 + \left[\frac{\partial N_0}{\partial \phi_v} \right]_{N_v = N_0} \Delta\phi_v \exp(i\delta\omega t) + \text{c.c.} , \qquad (11b)$$

where N_0 is the carrier density induced by $E(\omega)$ alone

TABLE I. Expressions of the $f_J(\Delta)$ functions.

$$f_1(\Delta)=[\bar{\rho}_v(\Delta,\phi_v)-\bar{\rho}_c(\Delta,\phi_c)]g(\Delta)$$
$$f_2(\Delta)=\bar{\rho}_v(\Delta,\phi_v)[1-\bar{\rho}_v(\Delta,\phi_N)]$$
$$f_3(\Delta)=\bar{\rho}_c(\Delta,\phi_c)[1-\bar{\rho}_c(\Delta,\phi_c)]$$
$$f_4(\Delta)=(-\Delta+i)g(\Delta)f_2(\Delta)$$
$$f_5(\Delta)=(-\Delta+i)g(\Delta)f_3(\Delta)$$
$$f_6(\Delta)=f_2(\Delta)g(\Delta)$$
$$f_7(\Delta)=f_3(\Delta)g(\Delta)$$

and $\Delta\phi_c$ and $\Delta\phi_v$ are the change in Fermi-level positions induced by the field modulation. In order to ensure the electric neutrality in the semiconductor, the carrier density variations must be the same in both bands:

$$\left[\frac{\partial N_c}{\partial\phi_c}\right]_{N_c=N_0}\Delta\phi_c=\left[\frac{\partial N_v}{\partial\phi_v}\right]_{N_v=N_0}\Delta\phi_v . \tag{12}$$

Solving Eqs. (7) to first order in $E^{\pm}(\omega\pm\delta\omega)$ and using the neutrality condition[12] allow us to calculate the polarization induced at the frequencies $\omega\pm\delta\omega$ by using relation (6), also written to first order in $E^{\pm}(\omega\pm\delta\omega)$:

$$P(\omega+\delta\omega)=[\mathcal{X}(\omega)+\mathcal{X}']E^+(\omega+\delta\omega)+\mathcal{X}''E^{-*}(\omega-\delta\omega) , \tag{13a}$$

$$P(\omega-\delta\omega)=[\mathcal{X}(\omega)+\mathcal{X}']E^-(\omega-\delta\omega)+\mathcal{X}''E^{+*}(\omega+\delta\omega) . \tag{13b}$$

The nonlinear susceptibilities $\mathcal{X}'$ and $\mathcal{X}''$ entering Eqs. (13) are

$$\mathcal{X}'=\frac{cn\sigma}{8\pi^3\omega}\left[\frac{2m}{\hbar T_2}\right]^{3/2}\left[-\frac{i}{2a}\left[\frac{I_1I_4}{I_2}+\frac{I_1I_5}{I_3}\right]\left|\frac{E}{E_S}\right|^2\right] , \tag{14a}$$

$$\mathcal{X}''=\frac{cn\sigma}{8\pi^3\omega}\left[\frac{2m}{\hbar T_2}\right]^{3/2}\left[-\frac{i}{2a}\left[\frac{I_1I_4}{I_2}+\frac{I_1I_5}{I_3}\right]\frac{E^2}{|E_s|^2}\right] , \tag{14b}$$

where

$$a=\delta_1-\frac{i}{2}\left|\frac{E}{E_s}\right|^2\left[\frac{I_6}{I_2}+\frac{I_7}{I_3}\right]-i(1+3AT_1N_0^2) . \tag{15}$$

In these expressions $\delta_1=\delta\omega T_1$ and I_j $(j=1-7)$ are integrals defined by the following relation:

$$I_j=\int_{-\infty}^{\Delta_0}f_j(\Delta)(\Delta_0-\Delta)^{1/2}d\Delta , \tag{16}$$

the functions $f_j(\Delta)$ being given in Table I.

III. TWO-LEVEL SYSTEM

The two-level system including Auger recombination is schematized in Fig. 2. The pump frequency ω is detuned by $\delta/T_{2,2N}$ from the two-level resonance ω_{12}, $T_{2,2N}$ being the transverse relaxation time of the two-level system and $\delta=(\omega-\omega_{12})T_{2,2N}$ the normalized detuning parameter. Auger recombination is taken into account by adding a bath of mean energy $2\hbar\omega_{12}$. Due to
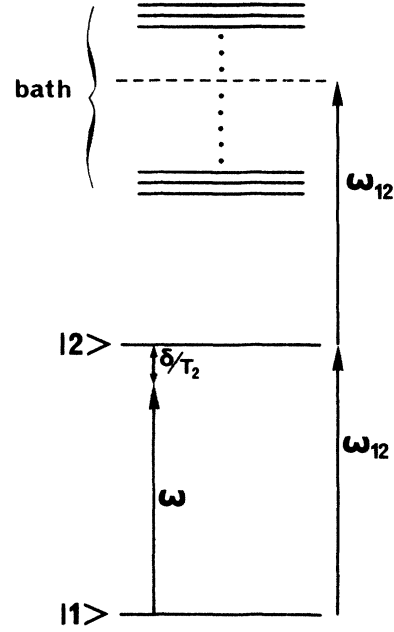

FIG. 2. Energy diagram of the two-level system.

the large bandwidth of the bath its lifetime is very short, so that total population can be repartitioned only between levels $|1\rangle$ and $|2\rangle$. Using the density matrix formalism, the Fourier components of the polarization can be easily calculated as

$$P(\omega)=\mathcal{X}_{2N}E(\omega) , \tag{17a}$$

$$P(\omega+\delta\omega)=(\mathcal{X}_{2N}+\mathcal{X}_{2N}')E^+(\omega+\delta\omega)+\mathcal{X}_{2N}''E^{-*}(\omega-\delta\omega) , \tag{17b}$$

$$P(\omega-\delta\omega)=(\mathcal{X}_{2N}+\mathcal{X}_{2N}')E^-(\omega+\delta\omega)+\mathcal{X}_{2N}''E^{+*}(\omega+\delta\omega) . \tag{17c}$$

The susceptibilities used in Eqs. (17) are

$$\mathcal{X}_{2N}=\frac{cn\sigma_{2N}}{4\pi\omega}\frac{-\delta+i}{1+\delta^2}N_{2N}(1-2\rho_2) , \tag{18a}$$

$$\mathcal{X}_{2N}'=\frac{cn\sigma_{2N}}{4\pi\omega}\frac{-\delta+i}{1+\delta^2}\frac{N_{2N}(1-2\rho_2)}{D}\frac{|E(\omega)|^2}{|E_{S,2N}|^2(1+\delta^2)} , \tag{18b}$$

$$\mathcal{X}_{2N}''=\frac{cn\sigma_{2N}}{4\pi\omega}\frac{-\delta+i}{1+\delta^2}\frac{N_{2N}(1-2\rho_2)}{D}\frac{E^2(\omega)}{|E_{S,2N}|^2(1+\delta^2)} , \tag{18c}$$

with

$$D=\delta_{1,2N}+i\left[1+3A_0\rho_2^2+\frac{|E(\omega)|^2}{|E_{S,2N}|^2(1+\delta^2)}\right] . \tag{19}$$

In these expressions in which $\delta_{1,2N}=\delta\omega T_{1,2N}$ and $A_0=A_{2N}N_{2N}^2T_{1,2N}$, σ_{2N} and $|E_{S,2N}|^2$ are the absorption cross section and saturation intensity of each two-level system; for the N_{2N} two-level systems the popula-

tion relaxation properties are described by the longitudinal relaxation time $T_{1,2N}$ and the Auger constant A_{2N}. Finally, the occupation probability ρ_2 in level $|2\rangle$ is given by

$$A_0\rho_2^3 + \left[\frac{|E(\omega)|^2}{|E_{S,2N}|^2(1+\delta^2)}\right]\rho_2$$

$$-\frac{1}{2}\frac{|E(\omega)|^2}{|E_{S,2N}|^2(1+\delta^2)} = 0 \ . \qquad (20)$$

Equation (20) always has only one real solution so that the susceptibilities are entirely determined when this equation is solved. Let us underline that when Auger recombination is negligible $(A_0=0)$ the susceptibilities given by Eqs. (18) reduce to the expressions given in Ref. 6.

IV. SEMICONDUCTOR TWO-LEVEL SYSTEM EQUIVALENCE

A. Principle of equivalence

In order to set up an equivalence for the description of the saturation properties of light-semiconductor interaction, it is necessary to relate the two-level parameters to that of the semiconductor itself. This equivalence is summarized in Table II. First of all, the relaxation properties which do not involve any interaction with the electromagnetic field must be the same for the equivalent two-level system and the real semiconductor $(T_{1,2N}=T_1$ and $A_{2N}=A)$. Then, at complete saturation $[\ |E(\omega)|^2\rightarrow\infty]$ the number of excited states is the same for the two descriptions, so that $N_{2N}=2N_\infty$, where $N_\infty=(1/\pi^2)\int_0^\infty k^2dk\ \bar{\rho}_c(k,\phi_c^\infty)$ is the free-carrier density for a complete saturation of the absorption at frequency ω. Finally, at low intensity $[\ |E(\omega)|^2\rightarrow0]$ the susceptibility at frequency ω can be written

$$\chi(\ |E(\omega)|^2\rightarrow0)=\frac{\sigma cn}{4\pi\omega}N_0\left[-\frac{N_0'}{N_0}+i\right] , \qquad (21)$$

where

$$N_0=\frac{1}{2\pi^2}\left[\frac{2m}{\hbar T_2}\right]^{3/2}\int_{-\infty}^{\Delta_0}g(\Delta)(\Delta_0-\Delta)^{1/2}d\Delta$$

and

$$N_0'=\frac{1}{2\pi^2}\left[\frac{2m}{\hbar T_2}\right]^{3/2}\int_{-\infty}^{\Delta_0}\Delta g(\Delta)(\Delta_0-\Delta)^{1/2}d\Delta$$

represent the density of states inside the absorptive and dispersive profiles, respectively. The normalized detuning giving the same low-intensity susceptibility for the equivalent two-level system is therefore $\delta=N_0'/N_0$. From these three relations one can easily derive the equivalent two-level absorption cross section $\sigma_{2N}=(1+\delta^2)\sigma N_0/(2N_\infty)$ and saturation intensity $I_{S,2N}=2I_SN_\infty/[N_0(1+\delta^2)]$.

B. Comparison of "exact" and "two-level" susceptibilities

The semiconductor parameters used in these calculations are summarized in Table III. The "exact" susceptibilities χ, χ', and χ'' were numerically calculated through Eqs. (10) and (14) by using the values of the quasi-Fermi-levels first determined by numerically solving the integral Eqs. (8) and (9) with respect to the normalized pump intensity $|E(\omega)/E_s|^2$ ranging from 0.01 to 1000. The "two-level" susceptibilities were simply evaluated through Eqs. (18)–(20) and use of Table II. The calculations were made for four different laser-band-gap normalized detunings $(\Delta_0=-1, 0, 1,$ and 10) and without $(A_0=0)$ or with $(A_0=200)$ Auger recombination. In all the subsequent figures the exact and two-level susceptibilities normalized to the imaginary part of the low-intensity susceptibility are drawn in dotted and solid lines, respectively.

Figure 3 shows the relative variation $\Delta\alpha/\alpha$ of the imaginary part of χ versus $|E(\omega)/E_s|^2$ for $\Delta_0=-1, 0,$ 1, and 10. This figure demonstrates that the saturation of absorption is well represented by the equivalent two-level system. The agreement between the two models is good again in the case of the imaginary part of χ' (or χ''), as demonstrated by the plot in logarithm scale of $\mathrm{Im}(\chi'')$ versus $|E(\omega)/E_s|^2$ in the case of degenerate four-wave mixing $(\delta_1=0)$ without taking Auger recombination into account (see Fig. 4). On the other hand, as shown in Fig. 5, the exact dispersive susceptibilities $\mathrm{Re}(\chi)$ and $\mathrm{Re}(\chi'')$ are not very well fitted by the two-

TABLE II. Equivalent two-level parameters as a function of semiconductor characteristics.

Population relaxation time	$T_{1,2N}=T_1$
Auger constant	$A_{2N}=A$
Density	$N_{2N}=2N_\infty$
Normalized laser frequency detuning	$\delta=N_0'/N_0$
Absorption cross section	$\sigma_{2N}=\sigma(1+\delta^2)\dfrac{N_0}{2N_\infty}$
Saturation intensity	$I_{S,2N}=\dfrac{I_S}{1+\delta^2}\dfrac{2N_\infty}{N_0}$

TABLE III. Semiconductor parameters used for the calculation of the "exact" susceptibilities (m_0, free-electron mass; m_c, conduction-electron mass; m_v, hole mass; E_g, band-gap energy; and T, temperature).

$\dfrac{m_v}{m_0}$	$\dfrac{m_c}{m_0}$	E_g (eV)	T_2 (s)	Δ_1	T (K)
0.61	0.13	1.8	10^{-13}	20	273

level system. The discrepancy which will be explained in Sec. IV C is much more pronounced when $\Delta_0 = 10$ than for $\Delta_0 = -1$, where the equivalence is almost correct for the description of dispersion up to $|E(\omega)/E_s|^2 = 100$. Nevertheless, this misfit between the two descriptions of the dispersive properties of the semiconductor is not important for the four-wave mixing process. Indeed, as demonstrated by Fig. 6, the modulus of χ' (or χ'') which is of interest in four-wave mixing is well fitted by the two-level model whatever the laser-band-gap normalized detuning is. As is demonstrated in Sec. IV C, this is due to the fact that the dispersive contribution is of importance only for Δ_0 smaller than one when two-level model properly accounts for the saturation of the real part of χ' (or χ'').

The same conclusions can be made concerning the equivalence in the case of nearly degenerate four-wave mixing taking Auger recombination into account. As shown in Fig. 7, the equivalence is good in any case for the absorption coefficient α and the modulus of the mixing susceptibility. On the other hand, this figure demonstrates that, as expected, Auger recombination shortens the lifetime of free carriers; indeed, the saturation of absorption occurs for larger intensities and the nearly degenerate mixing susceptibility is higher when $A_0 = 200$, since in this case the medium can relax faster than the field modulation moves.[14]
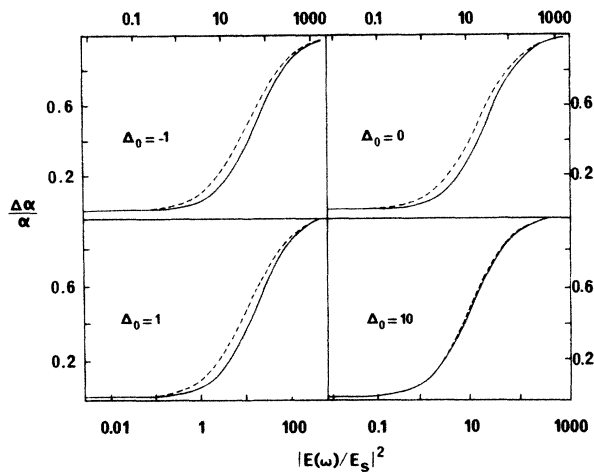

FIG. 4. Intensity dependence of Im(χ'') (dashed lines) and Im(χ''_{2N}) (solid lines) for $\Delta_0 = 0.1, 0, 1$, and 10 ($\delta_1 = 0$, $A_0 = 0$).

C. Discussion of the equivalence

The misfit between the exact and the two-level description of the saturation of the dispersive susceptibilities can easily be qualitatively understood. The exact dispersion is calculated through integration over the wave number k of the function $F(k) = k^2 \Delta g(\Delta)$. This function is represented in Fig. 8 together with the reduced bands both for $\Delta_0 = 0$ [Fig. 8(a)] and $\Delta_0 = 10$ [Fig. 8(b)]. For $\Delta_0 = 0$, the laser is exactly resonant with the band-gap energy; $F(k)$ is always positive and saturation occurs together with band filling when the pump intensity is increased. On the other hand, in the case in which the laser-band-gap normalized detuning is large, $F(k)$ is nearly antisymmetric with respect to k_0. As a conse-

FIG. 3. Intensity dependence of $\Delta\alpha/\alpha$ for $\Delta_0 = -1, 0, 1$, and 10 ($\delta_1 = 0$, $A_0 = 0$). The dashed and solid lines represent the "exact" and "two-level" calculations, respectively.
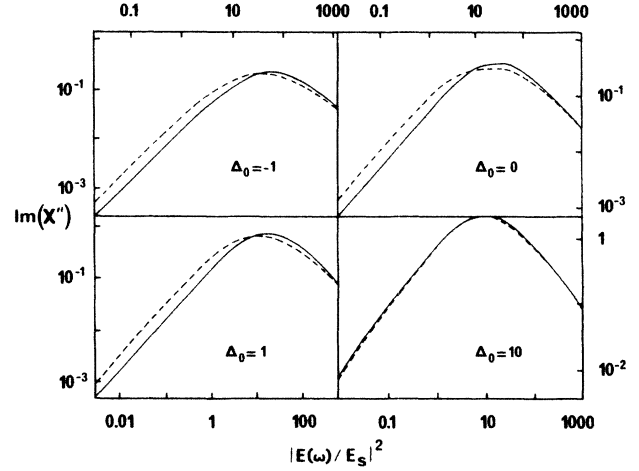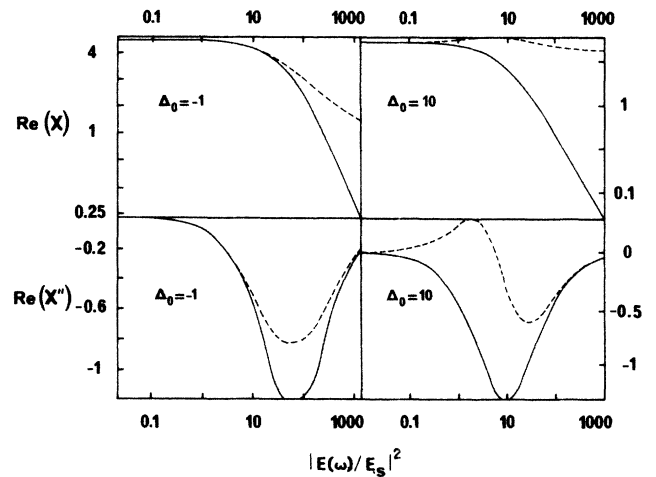

FIG. 5. Intensity dependence of Re(χ) and Re(χ'') (dashed lines) and Re(χ_{2N}) and Re(χ''_{2N}) (solid lines) for $\Delta_0 = -1$ and 10 ($\delta_1 = 0$, $A_0 = 0$).
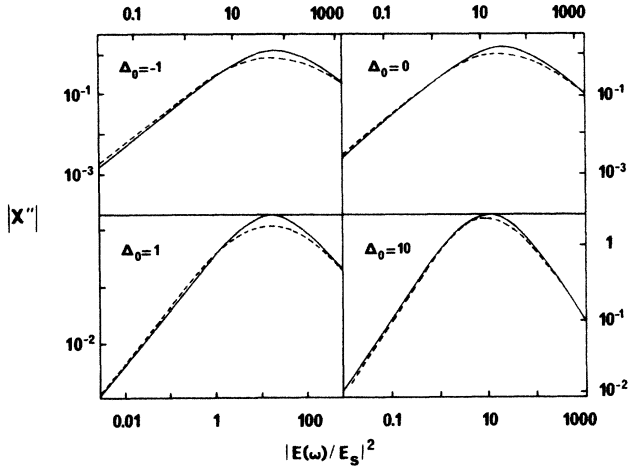
FIG. 6. Intensity dependence of $|X''|$ (dashed lines) and $|X''_{2N}|$ (solid lines) for $\Delta_0=-1, 0, 1$, and 10 ($\delta_1=0$, $A_0=0$).

quence, the band filling generated through the intensity increase decreases the magnitude of the negative part of $F(k)$, thus providing an increase in the dispersive susceptibility Re(X) (see Fig. 5 for $\Delta_0=10$). The two-level model cannot be applied in this case, but, as $F(k)$ is antisymmetric with respect to k_0, negative and positive contributions to the integral compensate each other and the equivalent two-level system is resonant in this case. This fact actually explains why the modulus of the mixing susceptibility shown in Fig. 6 is always well
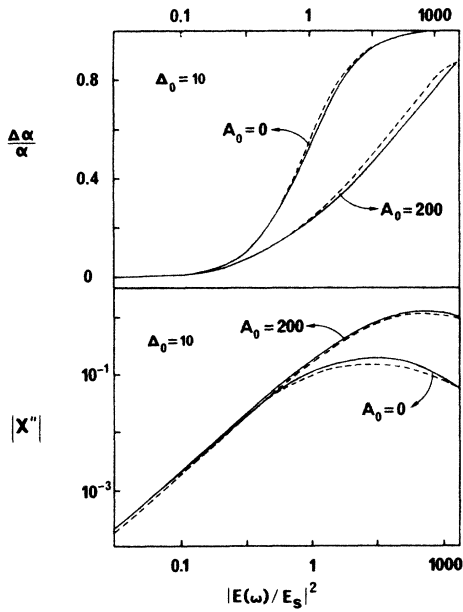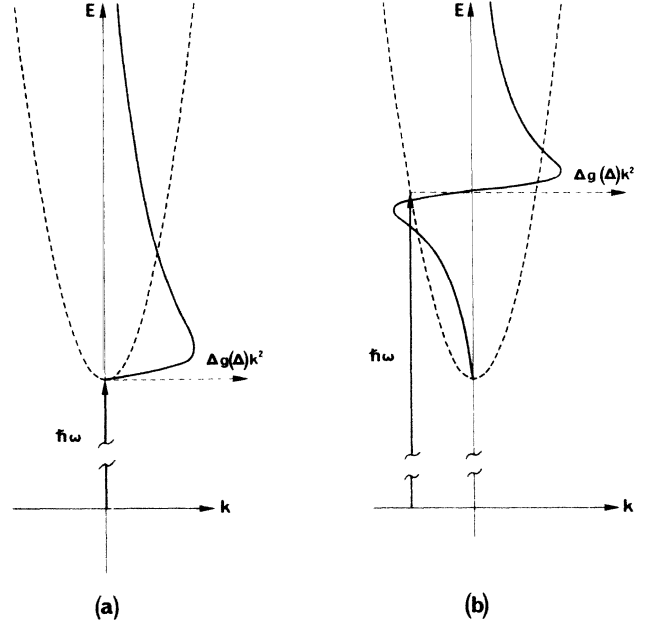

FIG. 8. Dispersion function $F(k)$ for the semiconductor in the reduced band scheme. (a) and (b) correspond to the laser frequency in or above band-gap resonance, respectively.

represented by the equivalent two-level system. Indeed, the absorptive susceptibilities which involve the symmetric function $G(k)=k^2g(\Delta)$ do not suffer from the same problem and are, therefore, always well represented by the equivalent two-level system.

D. Use of the equivalence

For each value of Δ_0 the calculated value of the two-level normalized detuning $\delta=N'_0/N_0$ is found to be quite nearly equal to $\Delta_c^\infty+\Delta_v^\infty$. This equality means that the energy levels $|1\rangle$ and $|2\rangle$ of the equivalent two-level system lie at the quasi-Fermi-levels, as shown in Fig. 9. Knowing the band-gap energy and the effective masses of the semiconductor, the two-level parameters can easily be related to the absorption cross section and saturation intensity of the electron-hole pair in the semiconductor.

The two-level normalized detuning δ is calculated by performing two integrations in order to determine N_0 and N'_0; nevertheless, in the case in which Δ_0 is much larger than one, $N_0=(1/2\pi^2)(2m/\hbar T_2)^{3/2}(\Delta_0)^{1/2}$ and as demonstrated in Sec. IV C the pump frequency is reso-

FIG. 7. Intensity dependence of $\Delta\alpha/\alpha$ and $|X''|$ for $\Delta_0=10$ in the case of nearly degenerate four-wave mixing ($\delta_1=50$) with and without Auger recombination. The dashed and solid lines represent the exact and two-level calculations, respectively.
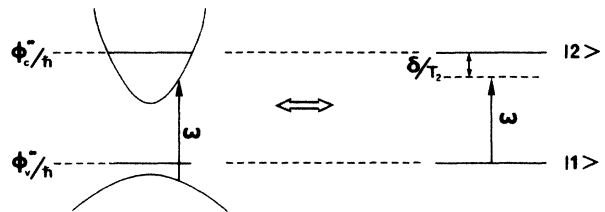

FIG. 9. Two-level picture of the semiconductor.

nant with the equivalent two-level system ($\delta = 0$).

The number of equivalent two-level systems $N_{2N} = 2N_\infty$ can be obtained through the solution of the coupled equations

$$\phi_c^\infty - \phi_v^\infty = \hbar \left[\omega - \frac{\delta}{T_2} \right] , \tag{22a}$$

$$N_c(\phi_c^\infty) = N_v(\phi_v^\infty) = N_\infty . \tag{22b}$$

The first equation is related to the position of the energy levels of the equivalent two-level system, and the second one is used to ensure the neutrality of the medium. As integrals giving N_c and N_v are tabulated,[9] an iterative technique allows us to rapidly determine N_∞ without any numerical integration.

The absorption cross section and saturation intensity of the equivalent two-level system are then easily calculated by using the expression used in Table II.

V. CONCLUSION

This paper has developed a quantitative description of the saturation characteristics of absorption and degenerate or nearly degenerate four-wave mixing processes in semiconductors. The calculation which takes intraband relaxation into account is valid when all characteristic times are much larger than the intraband relaxation time (10^{-13} s), this condition being very easily fulfilled in most experiments. As the results given by this theory are not very easy to handle when incorporated into the propagation equations, an equivalence with a two-level system has also been developed. This equivalence is always valid for absorptive properties of the semiconductor whatever the pump wavelength is. Concerning dispersion, the equivalence is nearly appropriate for pump wavelengths near or below the band gap. Nevertheless, as the dispersive contribution is much smaller than the absorptive one when the pump wavelength is much larger than that of the band gap, the equivalence is also valid for degenerate or nearly degenerate four-wave mixing whatever the pump wavelength is. A very simple instruction guide has also been given which makes the equivalence very straightforward to use.

As a demonstration, this equivalence has already been successfully used in a theoretical interpretation of the influence of Auger recombination in the relaxation process of semiconductor-doped glasses.[15] Let us emphasize in a final statement that this equivalence can also be used in the case of semiconductor lasers, provided some changes are made in order to take the electrical pumping into account.

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