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Monte Carlo simulation of harmonic generation in InP

D. PERSANO ADORNO, M. ZARCONE, AND G. FERRANTE
Istituto Nazionale di Fisica della Materia and Dipartimento di Fisica e Tecnologie Relative,
Viale delle Scienze, 90128 Palermo, Italy

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Abstract

Harmonics spectra in an *n*-type InP bulk semiconductor showing up to the 13th harmonic are reported. The external field is linearly polarized with the frequency $\nu = 100$ GHz. Calculations are based on a Monte Carlo simulation for the electron motion in the conducting band and on an electrodynamics equation for the harmonics generation. The effect of a significant reduction of efficiency in the generation of particular harmonics is found, and is traced back to a sort of alternating field Gunn effect. A short analysis of the different physical mechanisms giving rise to harmonics generation is presented.

Harmonics generation arising from the interaction of intense radiation fields in the frequency range from microwave to far-infrared with bulk semiconductors has become recently an interesting field of investigations. Both experimental (Keilmann *et al.*, 1990; Urban *et al.*, 1995) and theoretical (Urban *et al.*, 1996; Brazis *et al.*, 1998; Persano Adorno *et al.*, 2000) analyses have shown high conversion efficiency for the third and fifth harmonics in Si, GaAs, and InP crystals in the frequency range 30–500 GHz and temperature range 80–400 K. We report harmonics spectra in an *n*-type InP bulk semiconductor showing harmonics up to the 13th order with relatively high efficiency, going from 2×10^{-2} for the 3rd harmonic to 2×10^{-6} for the 13th one. The above highly nonlinear electromagnetic processes in semiconductors are due mainly to the nonlinear response of the free carriers interacting with the external field. The nonlinear interaction of the field with the lattice and with bound carriers is neglected. Nonlinear effects are included through the velocity dependence of the effective electron mass $m^*(v)$ (due to nonparabolicity of the conducting band) and through the carriers relaxation time $\tau_c \simeq 1/\nu_c$ with ν_c being the total collision frequency. In fact, the work per unit time performed by the external electric field on the free electron is given by $W = \vec{j} \cdot \vec{E}$. Since the velocity $\vec{v}$ and consequently the current density j oscillate at the frequency ν of the external electric field E, the work W and the average energy $\langle \varepsilon \rangle$ will oscillate at frequency 2ν. Then the total collision frequency $\nu_c(\varepsilon)$ is modulated at frequency 2ν and in turn the free electrons drift velocity will acquire a component oscillating at frequency 3ν, that will give rise to the third harmonic generation. Further collisions will give rise to a modulation of the drift velocity at higher frequency, consequently generating harmonics of a higher order.

In this article we limit our interest to the III–V class of semiconductor compounds, taking as its representative an *n*-type InP bulk sample interacting with a $\nu = 100$ GHz radiation field.

The propagation of an electromagnetic wave along the z direction in the medium is described by the Maxwell equation

$$\frac{\partial^2 \vec{E}}{\partial z^2} - \frac{1}{c}\frac{\partial^2 \vec{E}}{\partial t^2} = \mu_0 \frac{\partial^2 \vec{P}}{\partial t^2}, \tag{1}$$

where

$$\vec{P} = \varepsilon_0(\chi_1 + \chi_2 E + \chi_3 E^2 + \ldots)\vec{E} \tag{2}$$

is the polarization of the free electron gas in terms of the linear χ_1 and nonlinear $\chi_2 \chi_3, \ldots$ susceptibilities.

The source of the nonlinearity is the current density

$$\vec{j} = -ne\vec{v}(\vec{E}), \tag{3}$$

related to the polarization $\vec{P}$ by

$$\vec{j} = \frac{\partial \vec{P}}{\partial t}. \tag{4}$$

Address correspondence and reprint requests to: Michelangelo Zarcone, Di partimento di Fisica e Tecnologie Relative, Università di Palermo, Viale delle Scienze, 90128 Palermo, Italy. E-mail: zarcone@unipa.it

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Expanding the electrons velocity and the electric field in terms of their Fourier components as

$$\vec{v} = \sum_q \vec{v}_q \exp\{-iq(\omega t - kz)\} \tag{5}$$

$$\vec{E} = \sum_q \vec{E}_q \exp\{-iq(\omega t - kz)\}, \tag{6}$$

and taking only the leading term in the nonlinear part of the qth component of the polarization $\vec{P}$

$$\vec{P}_q^{NL} = \varepsilon_0 \chi_q \vec{E}_1^q, \tag{7}$$

we obtain a relation between the qth component of the velocity and the susceptibility χ_q given by

$$\chi_q = -\frac{inev_q}{q\omega E_1^q}. \tag{8}$$

To neglect the field-dependent absorption in this article, we limit our analysis to thin samples and we do not consider the complex form of the dielectric function $\varepsilon(\omega)$. With these assumptions, substituting the expansion for the electric field and using the above relation in the Maxwell equation, the qth harmonic intensity normalized to the fundamental one is given by (Persano Adorno *et al.*, 2000):

$$\frac{Iq}{I_1} = \frac{1}{q^2}\frac{v_q^2}{v_1^2}. \tag{9}$$

The coefficients v_q are given by the Fourier transform of the electron velocity, which is obtained via multiparticle MC simulation of the electron motion in the semiconductor.

In our model we considered the following scattering mechanisms: scattering with acoustic phonons, ionized impurities, acoustic piezoelectric phonons, polar optical phonons, and optical nonpolar phonons.

The InP crystal parameters are the same as that used by Brazis *et al.* (1998). The conduction bands of InP are represented by the Γ valley, by four equivalent L valleys and by three equivalent X valleys. At thermal equilibrium the electrons are expected to be in the conduction band of the InP bulk located in the central Γ valley.

We have performed our calculations with a free electrons concentration $n = 10^{19}$ m^{-3} and temperature $T = 80$ K. The MC simulation has been done for different field amplitudes in the range 5–60 kV/cm.

For such values of parameters, the Fermi temperature is much smaller than the electron temperature $T_F \ll T$, and degeneracy does not play any important role. Moreover, in our calculations we neglect electron–electron collisions, which usually do not affect transport properties in semiconductors. In fact, in such collisions the total momentum and the total energy of the two colliding particles are conserved and no dissipation occurs. Their main effect is in the redistribution of energy and momentum among the particles with a change in the shape of the distribution function.

Electrons are expected to exhibit an enhanced nonlinearity in high electric fields due to the equivalent and nonequivalent intervalley scattering among the three (Γ, L, X) valleys (Persano Adorno *et al.*, 2000). The spectra of harmonics generation at an oscillating field of amplitudes $E = 5$ kV/cm, $E = 25$ kV/cm, and $E = 60$ kV/cm are given in Figure 1. The efficiency of the harmonics for a given intensity of the field decreases with the order, but their relative efficiency is strongly dependent on the field intensity. For a weak field $E = 5$ kV/cm and for a strong field $E = 60$ kV/cm the ratio of the efficiencies between two adjacent harmonics is almost constant with a value of about an order of magnitude. At intermediate values of the external electric field a significant reduction of the efficiency of selected harmonics is observed. For a field intensity of $E = 25$ kV/cm, the ratio of the third to the fifth harmonic is about 100, and the seventh harmonic practically disappears. To better understand this

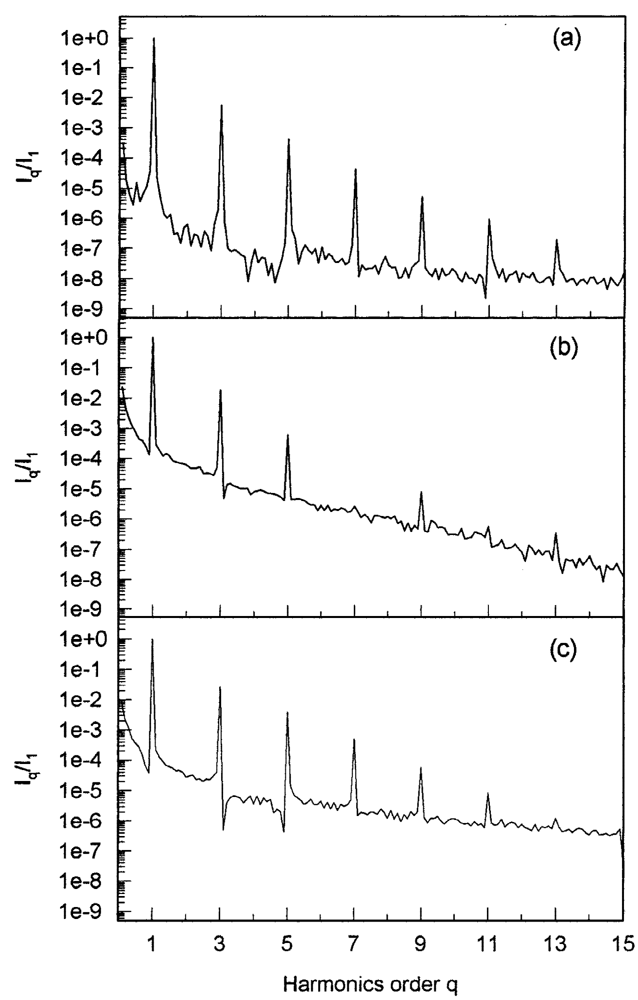

Fig. 1. Harmonics generation efficiency versus their order with $\nu = 100$ GHz, $T = 80$ K, $n = 10^{19}$ m^{-3}. (a) $E = 5$ kV/cm, (b) $E = 25$ kV/cm, (c) $E = 60$ kV/cm.

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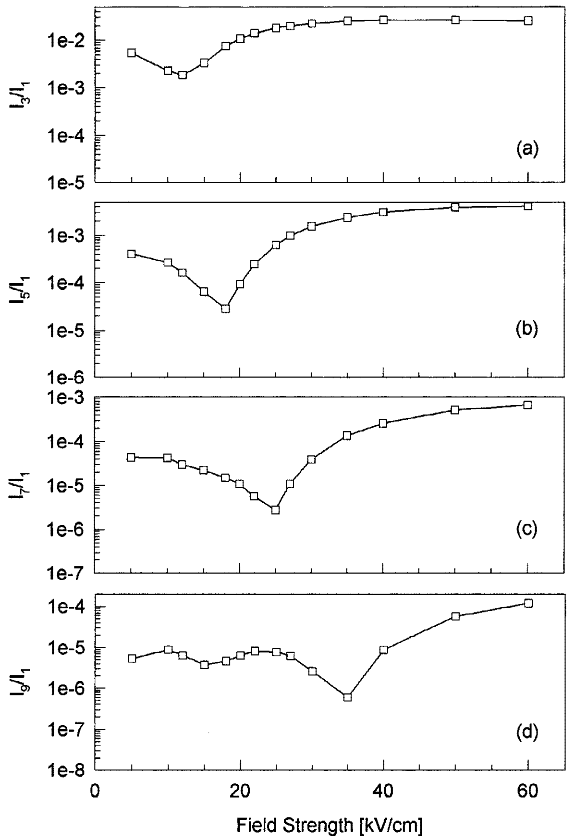

Fig. 2. Efficiency of the first four odd harmonics as a function of the field strength. $\nu = 100$ GHz, $T = 80$ K, $n = 10^{19}$ m^{-3}.

behavior we report in Figure 2 the efficiencies of the first four odd harmonics of the frequency $\nu = 100$ GHz as a function of the electric field amplitude. Besides reduction of particular harmonics efficiency, the reported calculations show additional novel features of interest: for high power levels, the harmonics generation efficiency exhibits saturation while for weak fields, the efficiency is decreasing with the intensity of the field and presents a deep minimum that for the third harmonic corresponds to a value of $E = 12$ kV/cm. This minimum is shifted toward higher fields for higher orders and for the ninth order is situated at $E = 35$ kV/cm. Moreover a small secondary minimum at lower fields is present for the ninth order harmonic. These minima arise for electric field values such that the energy gained by the electrons in the field is large enough to permit transitions between the Γ and the L valley. When it takes place, the electron velocity suffers a sudden decrease, together with the energy available for harmonics generation. This picture holds for relatively weak fields, when most of the electrons are in Γ valley and start to be transferred in the other valleys. This behavior is reminiscent of the Gunn effect occurring in the presence of a static field (for InP $E \simeq 10$ kV/cm). In the presence of an oscillating electric field, the critical value of the field strength at which a given harmonic suffers the largest reduction of generation efficiency is expected to

Table 1. *Comparison between the results for the ratio $E_q/E_{q'}$ obtained from our MC simulation and the estimates. q and q' are harmonics order numbers.*

$E_q/E_{q'}$	q/q'	Simulation
E_5/E_3	1.67	1.50
E_7/E_3	2.33	2.27
E_7/E_5	1.40	1.39
E_9/E_3	3.00	2.92
E_9/E_5	1.80	1.94
E_9/E_7	1.29	1.29

depend on the radiated frequency; in particular, increasing with it. In our case, the period of the fundamental radiation $\tau = 10^{-11}$ s is larger than the average collision time $\tau_c \simeq 10^{-13}$ s, by two orders of magnitude, roughly implying hun-

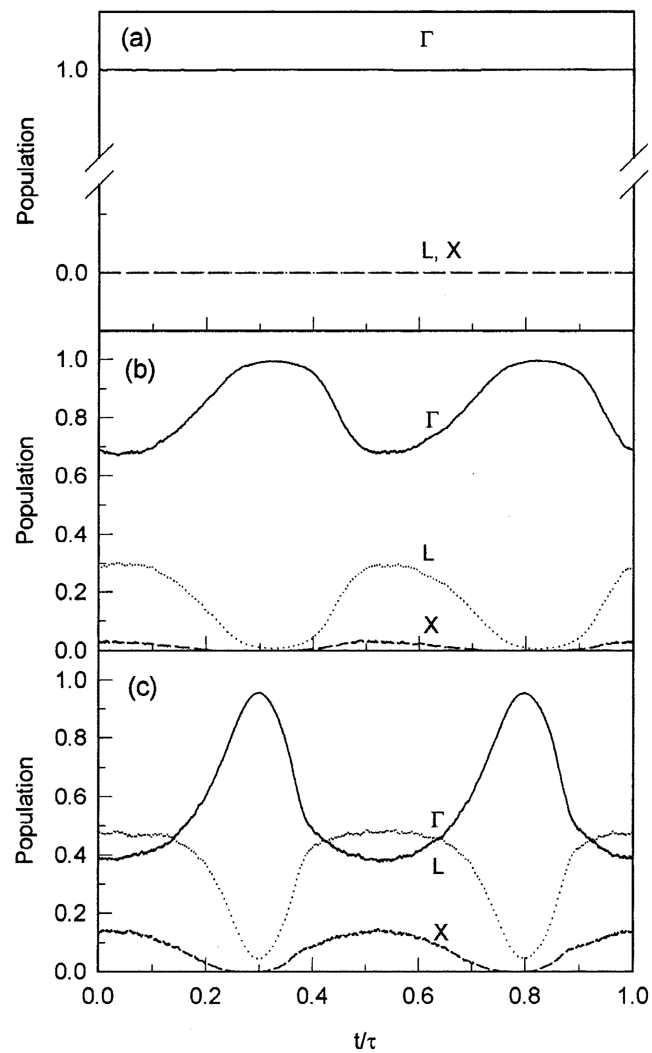

Fig. 3. Population of the three different valleys (Γ, L, X) as a function of time (in units of the external field period τ). Other input data as in Figures 1 and 2. (a) $E = 5$ kV/cm, (b) $E = 25$ kV/cm, (c) $E = 60$ kV/cm.

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dreds of collisions during a cycle of the external field. In such a case, we can exploit quasistatic considerations, and assume that the field transfers to the electron in the time τ in average energy $\langle \varepsilon \rangle = m\overline{v_E^2(t)}/2 = e^2E^2/4m(2\pi\nu)^2$ where $v_E(t) = v_E \sin 2\pi\nu t$ with $v_E = eE/2\pi m\nu$ being the amplitude of the electron quiver velocity. From this follows that the ratio between the values of the intensity of two fields with different frequencies ν and v' that transfer the same amount of energy $\langle \varepsilon \rangle$ is given by the ratio of the two frequencies $E_\nu/E_{\nu'} = \nu/\nu'$. Consequently we can expect that the ratio between the field values corresponding to the minima of efficiency of two different harmonics should be, roughly, equal to the ratio between the harmonic order numbers. In our simulation we get $E_5/E_3 = 1.5$; $E_7/E_3 = 2.27$; $E_7/E_5 = 1.39$; $E_9/E_3 = 2.92$; $E_9/E_5 = 1.94$; and $E_9/E_7 = 1.29$, which are in good agreement with our electron average energy considerations (see Table 1). The secondary minimum for the high order efficiencies is due to the switching on of the scattering mechanism with polar optical phonons, which induce a small minima in the total scattering probability.

To gain more insight into the scattering mechanisms which give the greater contribution to the efficiency of the harmonics generation, we report (Fig. 3) the population of the three different valleys (Γ, L, X) as a function of time after a transient time of 2 ps. For weak fields, almost all the electrons are still in the Γ valley and the only process involved in harmonics generation is the intravalley scattering. For strong fields, the population of the valleys L and X becomes more important and intervalley scattering among nonequivalent valleys as well as the intravalley processes in all three valleys come into play. Moreover the populations are modulated by the external field and oscillate around average values given roughly at $E = 25$ kV/cm by 0.85 for the Γ, 0.14 for the L, and 0.01 for the X valleys and at $E = 60$ kV/cm by 0.70 for the Γ, 0.25 for the L, and 0.05 for the X valleys. The valley Γ always remains the most populated one even at the strongest field. For this reason only a limited number of electrons undergo transitions to the upper valleys. Consequently the population curves, in a field period, present a symmetry with the rise time of the satellite valleys not very different from the decay one, despite the fact that due to the larger density of states of the satellite valleys, one could expect that the return time from the satellite valleys to the Γ valley should be longer than the up transition time.

We conclude by saying that the harmonics generation process appears to be mainly due to intravalley scattering in the Γ valley for weak fields and that for strong fields to intervalley scattering among nonequivalent valleys (mainly $\Gamma \to L, X$) in which electrons undergo a sudden change of their effective mass, amounting to a significant change of state and energy. An important interplay takes place between the intervalley transitions $\Gamma \to L, X$ and the intravalley transitions $L \to L'$, and $X \to X'$. We hope our results prompt new efforts, both experimental and theoretical, to fully understand and exploit the physical processes of high-order harmonics generation in semiconductors. Besides, our results may prove useful in shedding light on common features of such highly nonlinear processes of radiation–matter interaction in different media.

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