Quantum Interference Control of Carriers and Currents in Zincblende Semiconductors based on Nonlinear Absorption Processes

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Quantum interference between optical absorption processes can excite carriers with a polarized distribution in the Brillouin zone depending on properties of the incident optical fields. The polarized distribution of carriers introduces a current that can be controlled by the phases and polarizations of the incident optical fields. Here we study the quantum interference of 2- and 3-photon absorption processes in AlGaAs. We present theoretical predictions for carrier and current injection rates considering different frequencies, phases, and polarizations of the incident fields. We also discuss the important features that result from only nonlinear optical processes being involved, which leads for instance to a sharper distribution of carriers in the Brillouin zone.

I. INTRODUCTION

Quantum interference between different optical processes arises when two optical beams of different frequencies can lead to the same transition. Quantum interference between different excitation processes has been first used for photoionization of molecular systems, and asymmetric photoionization in semiconductors. In a crystal, amplitudes for different optical processes leading to electron-hole excitations can interfere constructively in some regions of the Brillouin zone (BZ), and destructively in others. By controlling the polarizations and phases of the incident fields, it is possible to excite carriers in selected localized regions of the BZ. Such Quantum Interference Control (QuIC), using 1- and 2-photon absorption processes, has been used for current injection in semiconductors, graphene, topological insulators, and transition metal dichalcogenides, as well as spin current injection in semiconductors. It has also been theoretically investigated for current injection in graphene nanoribbons, spin currents in topological insulators, and spin and valley currents in transition metal dichalcogenides.

In crystalline materials, every instance of QuIC studied to date has involved 1- and 2-photon absorption processes, partly because phase related optical fields of frequencies \( \omega \) and \( 2\omega \) can be conveniently achieved by second harmonic generation, while fractional ratios of the frequencies are harder to obtain. Currents injected via 1+2 QuIC have been exploited to determine parameters of the optical fields responsible for their injection. This method has found application in the measurement and stabilization of the carrier-envelope phase of a train of octave-spanning laser pulses. However, phase-coherent frequency combs can also be used to study more general \( m+n \) QuIC, especially for fractional ratios \( n/m < 2 \) as that only requires a narrower frequency range of the comb. Thus the use optical frequency combs for QuIC experiments presents an opportunity to separately study several non-linear optical processes in semiconductors, which cannot easily be done using simple harmonic generation as it only produces frequencies that are integer multiples.

In this article we present a theoretical study of QuIC with 2- and 3-photon processes in AlGaAs. We derive expressions for the optical injection coefficients, and evaluate them for different stoichiometries of AlGaAs using a 30-band model. We compute all the symmetry-allowed injection coefficients corresponding to different polarizations of the incident fields, and analyze their frequency dependence over a range where the injection of carriers that do not contribute to the current is avoided. That is, considering 3-photon absorption of photons at energy \( 3\hbar\omega/2 \), we require \( 2\hbar\omega \) to be less than the band gap. The alloy AlGaAs is an ideal material for 2+3 QuIC, as its stoichiometry can be chosen to yield a band gap appropriate for the available laser wavelengths. Experiments demonstrating 2+3 QuIC of photocurrent in AlGaAs are being reported in another article.

As expected, 2+3 QuIC has some qualitative differences compared to 1+2 QuIC. For instance, there is a change of sign in the current injection coefficient for different frequencies, which is due to an interplay between intraband and interband processes contributing to 3-photon absorption. We also find that 2+3 QuIC leads to sharper distributions of carriers in the BZ than 1+2 QuIC. The sharper distribution of carriers leads to a higher swarm velocity, which is a desirable feature for photocurrents, and it also opens the possibility of exciting carriers in semiconductors in a tailored fashion.

The outline of this article is the following: In Sec. II we present a method to compute the optical injection rates for a generic material. In Sec. III we describe the model used for AlGaAs. In Sec. IV we present our results for carrier and current injection from 2- and 3-photon absorption (2PA and 3PA) processes in AlGaAs. We also discuss the efficiency of the current injection by analyzing the swarm velocity, and computing the optimal laser intensities. In Sec. V we discuss the implications of our results and present our conclusions. We list the inde-
pended components of the optical injection tensors for zincblende lattice symmetry in the Appendix.

II. OPTICAL INJECTION RATES

Assuming the independent particle approximation, we consider a system described by a Hamiltonian $\mathcal{H}_0$ in the absence of any external perturbation, so the full Hamiltonian $\mathcal{H}(t)$ in the presence of the external perturbation $\mathcal{V}_{\text{ext}}(t)$ is $\mathcal{H}(t) = \mathcal{H}_0 + \mathcal{V}_{\text{ext}}(t)$, where in the basis of eigenstates of $\mathcal{H}_0$,

$$\mathcal{H}_0 = \sum_{nk} \hbar \omega_{nk} a_{nk}^\dagger a_{nk}, \quad (1)$$

$$\mathcal{V}_{\text{ext}}(t) = \sum_{mnk} a_{nk}^\dagger V_{mnk}(t) a_{nk}(t). \quad (2)$$

In the interaction picture, the creation and annihilation fermion operators are $a_{nk}^\dagger t = a_{nk}^\dagger e^{i\omega_{nk}t}$ and $a_{nk}(t) = a_{nk} e^{-i\omega_{nk}t}$, and the time-evolution operator can be expanded as

$$\mathcal{U}(t) = 1 + \sum_{N=1}^\infty \frac{1}{i\hbar} \int_0^t dt_N \mathcal{V}_{\text{ext}}(t_N) \cdot \mathcal{U}_N(t_N), \quad (3)$$

and the terms of each order in $\mathcal{V}_{\text{ext}}$ can be obtained from the previous one by

$$\mathcal{U}_N(t) = \int_0^t \frac{dt_N}{i\hbar} \sum_{mnk} a_{nk}^\dagger V_{mnk}(t_N) a_{nk} e^{i\omega_{mn}t} \mathcal{U}_{N-1}(t_N), \quad (4)$$

where $\omega_{mnk} = \omega_{mk} - \omega_{nk}$, and $\mathcal{U}_0(t) = 1$. We are interested in the excitation of an electron from a valence band $v$ to a conduction band $c$ due to the external field. This excited state is $|cvk\rangle = a_{vk}^\dagger |gs\rangle$, where $|gs\rangle$ is the ground state of $\mathcal{H}_0$ with filled valence bands. The state of the system is described by

$$|\psi\rangle = \mathcal{U}(t) |gs\rangle = \gamma_0 |gs\rangle + \sum_{cvk} \gamma_{cvk}(t) |cvk\rangle + \ldots \quad (5)$$

where the coefficients

$$\gamma_{cvk}(t) = \langle cvk | \mathcal{U}(t) | gs \rangle \quad (6)$$

contain the information we want. The expectation value of the density $\langle M \rangle$ of a quantity associated with an operator $M = \sum_{mnk} a_{mk}^\dagger M_{mnk} a_{nk}$ due only to the excitation of states $|cvk\rangle$ is

$$\langle M \rangle = \frac{1}{L^D} \sum_{cv'v'k} \gamma_{cv,k}^* \gamma_{cv,k} \langle cv'k | M | cvk \rangle$$

$$= \frac{1}{L^D} \sum_{cv'v'k} \left( M_{cv'k} \delta_{v'v} - M_{v'vk} \delta_{v'v} \right) \times \gamma_{cv,k}^* \gamma_{cv,k} e^{-i\omega_{cv',vk}t} e^{i\omega_{cvk}t}, \quad (7)$$

where $L$ is a normalization length, and $D$ is the spatial dimension of the system. For a full Hamiltonian $\mathcal{H}(t)$ that follows from a Hamiltonian for a single particle of the form

$$\mathcal{H}(x, p; t) = \frac{1}{2m} (p - eA(t))^2$$

$$+ \mathcal{H}_{\text{SO}}(x, p - eA(t)) + \mathcal{Y}_{\text{latt}}(x), \quad (8)$$

where $x$ and $p$ are position and momentum operators, $H_{\text{SO}}$ is the spin-orbit term, and $\mathcal{Y}_{\text{latt}}(x)$ is the lattice potential energy. Here we neglect a contribution to the interaction that is solely a function of time ($\sim |A(t)|^2$), for it will not lead to any transitions, and we work in a gauge where the electric field $E(t)$, assumed independent of position, is fully described by the vector potential $A(t)$. The interaction term in the Hamiltonian takes the form $\mathcal{V}_{\text{ext}}(t) = -e \mathbf{v} \cdot A(t)$, where $e = -|e|$ is the charge of the electron and $\mathbf{v} = -e^{-1} \partial \mathcal{H}/\partial A$ is the velocity operator. Indeed, the interaction is of the form we consider for any unperturbed Hamiltonian for a single particle that is at most quadratic in the momentum.

We take the vector potential to be

$$A(t) = \sum_\alpha A_\alpha e^{-i(\omega_\alpha t + \epsilon t)} = -\sum_\alpha \frac{i}{\omega_\alpha} E_\alpha e^{-i(\omega_\alpha t + \epsilon t)}, \quad (9)$$

with $\omega_\alpha = \pm \omega, \pm 3\omega/2$; here $\epsilon \to 0^+$ describes turning on the field from $t = -\infty$. The $\gamma_{cvk}(t)$ coefficients can be expanded as $\gamma_{cvk}(t) = \langle cvk | \mathcal{U}(t) | gs \rangle$ following the expansion (4) of $\mathcal{U}(t)$ for an incident optical field, so we can write the coefficients $\gamma_{cvk}(t)$ as

$$\gamma_{cvk}(t) = \mathcal{R}_{cvk}^{(N)} e^{-i(\Omega_N - \omega_{cvk} + \epsilon) t}, \quad (10)$$

where $\Omega_N = \omega_1 + \ldots + \omega_N$. The coefficients $\mathcal{R}_{cvk}^{(N)}$ involve the electric field amplitudes $E_\alpha$ according to

$$\mathcal{R}_{cvk}^{(N)} = R_{cvk}^{(N) ab} (\omega_\alpha, \ldots, \omega_\beta) E_\alpha^* \ldots E_\beta^*, \quad (11)$$

where repeated indices are summed; here superscripts refer to Cartesian indices and subscripts to incident frequency components. For the lower orders we have

$$R_{cvk}^{(1) a} (\omega_\alpha) = \sum_\alpha \frac{i\epsilon}{\hbar \omega_\alpha} \mathcal{R}_{cvk}^{(N)} \cdot (\omega_\alpha), \quad (12)$$

and

$$R_{cvk}^{(2) ab} (\omega_\alpha, \omega_\beta) = \sum_\alpha \frac{-e^2}{\hbar^2 \omega_\alpha \omega_\beta} \times \left( \sum_{cv'k} \frac{v_{cvk}^a v_{cv'k}^b}{\omega_\beta - \omega_{cvk}} + \sum_{cv'k} \frac{v_{cvk}^b v_{cv'k}^a}{\omega_\beta - \omega_{cvk}} \right), \quad (13)$$

and
We are interested in the non-oscillatory response of the system, so we focus on the $\Omega_N = \Omega_{N'}$ contributions to Eq. (7). In order to compute the injection rate $d\langle M \rangle/dt$ associated with that equation, it is important to realize that

$$
\frac{d}{dt} \langle \gamma^{cv,c'}_{\nu'}(k) e^{-i\omega_{\nu'} t} e^{i\omega_{cv} k t} \rangle_{t=0,t \to 0} = \sum_{\Omega_N = \Omega_{N'}} \frac{2\pi}{\hbar^2 \Omega_{cw}} R_{c,c'}^{(N)} R_{c'cv}^{(N)} \Big|_{\omega_{\nu'} = -\omega_{cv}} . \tag{14}
$$

The fact that the $R_{c'cv}^{(N)}$ coefficients are always accompanied by $\delta (\Omega_N - \omega_{cv})$ in the expression for the response allows for substitutions $3\hbar \omega - \omega_{cv} = 0$ that were used to write $R_{c'cv}^{(3)}$ in a simpler way in Eq. (13). The expression for the injection rate of $\langle M \rangle$ due to the interference of an $N'$ photon process with an $N$ photon process is

$$
d\langle M \rangle = \mu_{\alpha\beta\gamma \delta ...} (\Omega) E^a \omega^b \omega^c \omega^d \ldots E^q + c.c., \tag{15}
$$

where there are $N'$ frequency labels ($\alpha, \beta, \gamma, \delta, ...$) and $N$ frequency labels ($\rho, \sigma, ...$). The injection rate coefficient $\mu_{\alpha\beta\gamma \delta ...} (\Omega)$ is assembled from extracting the terms multiplying the electric field amplitudes in $R_{c,c'}^{(N)} R_{c'cv}^{(N)}$ of Eq. (14) together with the appropriate matrix elements appearing in Eq. (7) for $\langle M \rangle$. We give examples below.

Quantum interference of 2- and 3-photon processes

The processes of 3PA with frequency $\omega$ and 2PA with frequency $3\omega/2$ can interfere since the total frequency for each of them is $\Omega = 3\omega$. For such processes the frequencies are all equal in the equations (12) and (13) for the coefficients $R_{c'cv}^{(2)}$ and $R_{c'cv}^{(3)}$, and symmetrizing their components leads to some simplifications. Using $\omega_{\beta} = \omega_{c'cv} - \omega_{\alpha}$ and $\omega_{\alpha} = \omega_{\beta} = 3\omega/2$ in Eq. (12), the second order coefficient simplifies to

$$
R_{c'cv}^{(2)ab} \left( \omega_{\alpha}, \omega_{\beta}, \omega_{\delta} \right) = \sum_{\alpha \beta \gamma} \frac{\hbar^2}{\omega_{\alpha} \omega_{\beta} \omega_{\delta}} \left[ \sum_{c'} \frac{v^a_{c'cv} v^{b \dagger}_{c'cv} k^{c'cv} e^{i\omega_{c'cv} \delta k}}{\omega_{\alpha} - \omega_{c'cv} k} \right] \left[ \sum_{c'} \frac{v^{b \dagger}_{c'cv} v^{c \dagger}_{c'cv} k^{c'cv} e^{i\omega_{c'cv} \delta k}}{\omega_{\beta} - \omega_{c'cv} k} \right] - \sum_{c'} \left( \sum_{c'} \frac{v^a_{c'cv} v^{b \dagger}_{c'cv} k^{c'cv} e^{i\omega_{c'cv} \delta k}}{\omega_{\alpha} - \omega_{c'cv} k} \right) \left( \sum_{c'} \frac{v^{b \dagger}_{c'cv} v^{c \dagger}_{c'cv} k^{c'cv} e^{i\omega_{c'cv} \delta k}}{\omega_{\beta} - \omega_{c'cv} k} \right) \left( \sum_{c'} \frac{v^{d \dagger}_{c'cv} v^{c \dagger}_{c'cv} k^{c'cv} e^{i\omega_{c'cv} \delta k}}{\omega_{\delta} - \omega_{c'cv} k} \right) \left( \sum_{c'} \frac{v^{d \dagger}_{c'cv} v^{c \dagger}_{c'cv} k^{c'cv} e^{i\omega_{c'cv} \delta k}}{\omega_{\delta} - \omega_{c'cv} k} \right) \right] . \tag{13}
$$

and using $\omega_{\beta} + \omega_{\gamma} = \omega_{c'cv} - \omega_{\alpha}$ and $\omega_{\alpha} = \omega_{\beta} = \omega_{\gamma} = \omega$ in Eq. (13), the third order coefficient simplifies to

$$
R_{c'cv}^{(3)ab} \left( \omega, \omega, \omega \right) = R_{c'cv}^{(3)ab} \left( \omega, \omega, \omega \right) = \frac{\hbar^2}{\omega_{\alpha} \omega_{\beta} \omega_{\delta}} \sum_{mn} v^a_{cmk} v^b_{mnk} v^c_{nmk} v^d_{nck} e^{i\omega_{cmk} \delta \omega_{nck}} , . \tag{17}
$$

Notice that the denominators in Eqs. (16) and (17) are minimal for $m, n, c, v$ so the dominant contributions to $R_{c'cv}^{(2)}$ always involve intraband velocity matrix elements, but $R_{c'cv}^{(3)}$ also has contributions from interband velocity matrix elements. Intra-band velocity matrix elements are associated with the corresponding band dispersion, $v^a_{cmk} = \partial_{\omega} \omega_{nm}, which vanishes at the k point corresponding to the bandgap. Thus $R_{c'cv}^{(2)}$ is zero for total photon energies corresponding to the band gap, and increases for larger excess photon energies. The dependence of $R_{c'cv}^{(3)}$ on the total photon energy is different, as it depends on both interband and intraband velocity matrix elements. For total photon energies just above the gap, $R_{c'cv}^{(3)}$ is determined mainly by the interband matrix elements, but as the photon excess energy increases $R_{c'cv}^{(3)}$ becomes dominated by the intraband matrix elements, since the electronic transitions occur at k points with larger band dispersion.

The injection rate coefficients corresponding to the interference of 2- and 3-photon processes can then be computed as

$$
\mu_{2+3,fg} (\Omega) = 2\pi \int \frac{d\mathbf{k}}{(2\pi)^3} \delta_{c'c} \langle M_{c'cv} \delta_{c'cv} - M_{c'cv} \delta_{c'cv} \rangle \times \delta_{\omega_{\alpha} = -\omega_{\gamma}, \Omega} R_{c'cv}^{(3)ab} R_{c'cv}^{(2)fg} \delta (\Omega - \omega_{cv}) , \tag{18}
$$

following Eqs. (7) and (15), where we have taken the continuous momentum limit. For the plots in the next sections we use a frequency broadening $\Delta$ corresponding to $h\Delta = 13 \text{meV}$. The factor $R_{c'cv}^{(3)ab} R_{c'cv}^{(2)fg}$ changes sign under a transformation $\mathbf{k} \rightarrow -\mathbf{k}$, resulting in constructive versus destructive interference in opposite points of the Brillouin zone. In Fig. 1 we illustrate constructive versus destructive interference of 2- and 3-photon processes at opposite points in the Brillouin zone.
III. ELECTRONIC MODEL OF AlGaAs

We use a 30-band $k \cdot p$ model for computing the electronic bands. The model has free parameters associated with energies and momentum matrix elements at the $\Gamma$ point, and the parameters are adjusted to match the experimental results for band energies from $-5 \, \text{eV}$ to $4 \, \text{eV}$, such that computations of optical absorption coefficients are expected to be reliable for photon energies up to $6 \, \text{eV}$.

Using the $\Gamma$ point as the expansion point for a $k \cdot p$ model, the effective Hamiltonian that acts only on the periodic part of an energy eigenfunction of crystal momentum $k$ is

$$H_{\text{eff}} = H + \frac{\hbar}{m} k \cdot p + \frac{\hbar^2 k^2}{2m},$$

where $H$ is the Hamiltonian (8) with the vector potential set equal to zero; in this model we neglect the $k$ dependence of the effective spin-orbit term. The second term on the right-hand-side is the usual $k \cdot p$ contribution, and the last term is the contribution to the kinetic energy only due to the lattice momentum. The basis of states has 8 sets, 4 of them corresponding to the $\Gamma_1$ representation of the point group $T_d$ (or $43m$), 3 corresponding to the $\Gamma_4$ representation, and 1 to the $\Gamma_3$ representation. The $\Gamma_1$ representation has only 1 state, $\Gamma_4$ has 3 states, and $\Gamma_3$ has 2 states, so in total we have $4 \times 1 + 3 \times 3 + 1 \times 2 = 15$ states before considering spin; we denote these states as $|A\rangle, |B\rangle$, etc. Tensor products of these are taken with spin states to get 30 states in all. Terms $\langle A | H_{\text{eff}} | B \rangle$ are then $2 \times 2$ matrices, and take the form

$$\langle A | H_{\text{eff}} | B \rangle = E_A \delta_{AB}\sigma_0 + \frac{i}{3} \Delta_{AB} \cdot \sigma$$

$$+ i P_{AB} \cdot k \sigma_0 + \frac{\hbar^2 k^2}{2m} \delta_{AB} \sigma_0,$$

where $\sigma_0$ is the unit $2 \times 2$ matrix and the components of $\sigma$ are the usual Pauli matrices. The free parameters of the model are the energies $E_A$, the matrix elements of the spin-orbit term $\Delta_{AB}$, and the matrix elements of the momentum operator $P_{AB}$. Since the basis for the states is the same at every $k$ point, the corresponding $2 \times 2$ matrices corresponding to the velocity operator $\langle A | v | B \rangle$ are diagonal in the spin sector,

$$\langle A | v^a | B \rangle = \frac{1}{\hbar} \frac{\partial}{\partial k^a} \langle A | H_{\text{eff}} | B \rangle = \left( \frac{i}{\hbar} P^a_{AB} + \frac{\hbar k^a}{m} \delta_{AB} \right) \sigma_0,$$

from which the matrix elements of the velocity operator between the energy eigenstates can be determined.

For GaAs$^{39}$ and AlAs$^{41}$ we use reported parameters adjusted for room temperature, while the parameters for Al$_{x}$Ga$_{1-x}$As are obtained from a linear interpolation according to the stoichiometry. This approximation is accurate within an energy tolerance corresponding to room temperature$^{42}$. The chosen parameters lead to effective masses and $g$-factors that are in good agreement with experimental data. More important for the problems we consider, the band structures and linear optical absorption spectra are also in good agreement with experimen-
tal data. In Fig. 2 we show the relevant electronic bands for two different stoichiometries, and in Fig. 3 we show the imaginary parts of the corresponding dielectric functions, which are related to the 1-photon absorption rates (or carrier injection) by $\text{Im} \varepsilon(\Omega) = \hbar \epsilon^{xx}(\Omega)/2\epsilon_0$.

As already mentioned, the 30-band model allows us to perform reliable calculations for total photon energies up to $6 \text{ eV}$. However, for the energies we are most interested – below $3 \text{ eV}$ – we can get accurate results for the optical absorption coefficients even if only 6 valence and 2 conduction bands are included in the model, and only the valley including the $\Gamma$ point of the BZ are considered. We also point out that the model we use is applicable to most zincblende semiconductors, where each specific material corresponds to a particular set of parameters. Since the bandstructures of zincblende semiconductors are qualitatively similar, our results presented in the next sections are qualitatively valid for most direct-gap zincblende semiconductors.

![FIG. 3. Imaginary part of the dielectric function for two stoichiometries $\alpha$.](image)

**IV. QUANTUM INTERFERENCE CONTROL USING TWO- AND THREE-PHOTON ABSORPTION IN AlGaAs**

We consider two incident fields of different frequencies with amplitudes $E_\omega = E_\omega e^{i\phi_\omega} \hat{e}_\omega$ and $E_{3\omega/2} = E_{3\omega/2} e^{i\phi_{3\omega/2}} \hat{e}_{3\omega/2}$, where $E_\omega > 0$ and $E_{3\omega/2} > 0$ are the field magnitudes, the unit vectors $\hat{e}_\omega$ and $\hat{e}_{3\omega/2}$ indicate their polarizations, and $\phi_\omega$ and $\phi_{3\omega/2}$ indicate their phases. We also define the phase parameter $\Delta \phi = 2\phi_{3\omega/2} - 3\phi_\omega$, which will be useful later. We assume that the field at $3\omega/2$ has a weaker intensity than the field at $\omega$, and we demand that the frequencies satisfy $2\omega < \Delta g < 3\omega$, where $E_g = \hbar \Delta g$ is the electronic gap. Therefore only 3PA processes are important for the lower frequency field $E_\omega$, while only 2PA processes are relevant for the higher frequency field $E_{3\omega/2}$; the 3PA associated with $E_{3\omega/2}$ is weaker due to the lower intensity of the field, and we neglect it.

We focus on Al concentrations $\alpha$ such that $0.18 \lesssim \alpha \lesssim 0.38$, since $\text{Al}_{\alpha} \text{Ga}_{1-\alpha} \text{As}$ with $\alpha$ too small has a band gap smaller than $2\hbar \omega$ for telecommunication wavelengths ($\hbar \omega \sim 0.8 \text{ eV}$), and $\text{Al}_{\alpha} \text{Ga}_{1-\alpha} \text{As}$ with $\alpha$ too large is too reactive.

![FIG. 4. Two-photon carrier injection coefficients for two stoichiometries.](image)

**A. Carrier injection**

We track the number of injected carriers by calculating the number of electrons in the conduction bands, which corresponds to the operator

$$N = \sum_{ck} a_{ck}^\dagger a_{ck},$$

(22)

so we use $n_c \delta = \delta_{cc'}$ and $n_{cc'} = 0$ for the carrier density matrix elements in Eq. (18). The optical injection of
We show the frequency dependence of the independent components of the injection tensor coefficients in Appendix A. In Figs. 4, 6 and 5, we show the frequency dependence of the independent components of the coefficients $\xi_{2+3}^{abcd}(3\omega)$, $\xi_{2+3}^{abcde}(3\omega)$, and $\xi_{2+3}^{abdef}(3\omega)$ respectively. Notice that the 3PA coefficient is large for frequencies right above the band gap, while the coefficient for 2PA nearly vanishes for similar frequencies. As discussed below Eqs. (16) and (17), the dominant contribution to 2PA always involves intraband velocity matrix elements, which correspond to the band dispersion, so they vanish at the $\Gamma$ point of the Brillouin zone. The 3PA has contributions from interband velocity matrix elements, which in general do not vanish at $\Gamma$.

### B. Current injection

The injected current density ($J$) due to the quantum interference between two- and three-photon absorption processes is given by

$$\frac{d}{dt} \langle n \rangle_{2+3} = \xi_{2+3}^{abcd}(3\omega) E_{-\omega}^{a} E_{-\omega}^{b} E_{\omega}^{c} E_{\omega}^{d} + c.c.,$$

where $\hbar \Omega = 3h\omega$ is the total transition energy\(^{43}\), and the coefficients are calculated as

$$\xi_{2}^{abcd}(3\omega) = 2\pi \int \frac{dk}{(2\pi)^{D}} \sum_{cv} R_{cvk} R_{cvk}^{de} \delta(3\omega - \omega_{cv}),$$

$$\xi_{3}^{abcde}(3\omega) = 2\pi \int \frac{dk}{(2\pi)^{D}} \sum_{cv} R_{cvk}^{abcd} R_{cvk}^{de} \delta(3\omega - \omega_{cv}),$$

$$\xi_{2+3}^{abdef}(3\omega) = 2\pi \int \frac{dk}{(2\pi)^{D}} \sum_{cv} R_{cvk}^{abcd} R_{cvk}^{de} \delta(3\omega - \omega_{cv}).$$

The symmetries of the zincblende lattice, corresponding to the point group $T_d$ (or 43m), strongly restrict the number of independent non-zero components of the tensors $\xi_2$, $\xi_{2+3}$, and $\xi_3$. We list the independent components of the injection tensor coefficients in Appendix A. In Figs. 4, 6 and 5, we show the frequency dependence of the independent components of the coefficients $\xi_{2}^{abcd}(3\omega)$, $\xi_{3}^{abcde}(3\omega)$, and $\xi_{2+3}^{abdef}(3\omega)$ respectively. Notice that the 3PA coefficient is large for frequencies right above the band gap, while the coefficient for 2PA nearly vanishes for similar frequencies. As discussed below Eqs. (16) and (17), the dominant contribution to 2PA always involves intraband velocity matrix elements, which correspond to the band dispersion, so they vanish at the $\Gamma$ point of the Brillouin zone. The 3PA has contributions from interband velocity matrix elements, which in general do not vanish at $\Gamma$. 

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where $\hbar \Omega = 3h\omega$ is the total photon energy. We list the independent components of the injection tensor coefficient $\eta_{2+3}^{abdef}(3\omega)$ in Appendix A. In Fig. 7 we show the frequency dependence of the independent components of the coefficient $\eta_{2+3}^{abdef}(3\omega)$ for different stoichiometries. The plots show that some components change sign as the frequency increases. This sign flip is due to the competing contributions due to intraband and interband velocity matrix elements to the $R_{cvk}^{(3)}$ coefficients. For low excess photon energies, the excited carriers are close to the $\Gamma$ point in the BZ, and the interband contribution is the most important, as the band dispersion is small. For larger photon excess energies, the excited carriers are located further from the $\Gamma$ point in the BZ, so the band dispersion is large and the intraband contributions are more important.
To illustrate some aspects of the different tensor components, in Fig. 8 we plot the injection current for different polarizations of the incident fields in a typical experimental scenario. We assume that the sample has electrodes mounted such that they always measure the current along the [100] crystal direction, which we denote by \( \hat{x} \). In the first case we keep either \( \hat{e}_\omega \) or \( \hat{e}_{3\omega/2} \) fixed along the \( \hat{x} \) direction, while the other field is rotated in the \( \hat{x}-\hat{y} \) plane and points along the direction \( \theta = \hat{x} \cos \theta + \hat{y} \sin \theta \), where \( \hat{y} \) corresponds to the [010] crystal direction. In the second scenario, the polarizations of both incident fields are rotated in the \( \hat{x}-\hat{y} \) plane and they are kept either parallel or perpendicular to each other. In Fig. 8, we show that the current is largely along the \( \hat{e}_\omega \) direction regardless of the \( \hat{e}_{3\omega/2} \) direction. However, the magnitude of the current depends significantly on the \( \hat{e}_{3\omega/2} \) direction, and it is maximal for \( \hat{e}_{3\omega/2} = \hat{e}_\omega \).

Since the excited carriers respond to the induced voltage due to the injected current, and usually screen it at least partially, a good measure of the efficiency of the current injection is the swarm velocity, defined as \( v_{\text{swarm}} = \frac{d}{dt} \langle J \rangle / e \frac{d}{dn} \langle n \rangle \), which represents the average contribution to the injection current due to one excited electron.\(^{44}\) Since \( \langle n \rangle_{2+3} \ll \langle n \rangle_2 + \langle n \rangle_3 \), the total density of carriers is \( \langle n \rangle \approx \langle n \rangle_2 + \langle n \rangle_3 \), and for both light beams polarized along the \( \hat{x} \) direction we have a swarm velocity of magnitude

\[
 v_{\text{swarm}} = \frac{2 \left| J_{2+3} \right| E_\omega^3 E_{3\omega/2}^2}{|e| \left( \varepsilon^{xxxxxx}_3 (3\omega) E_\omega^6 + \varepsilon^{xxxx}_2 (3\omega) E_{3\omega/2}^4 \right)},
\]

(30)

where we have chosen \( \Delta \phi = \pi/2 \) to optimize the magnitude of the numerator. The whole expression is optimized
by choosing the intensities of the two beams appropriately; the condition to be satisfied is $\xi_2^{xxx} (3\omega) E_{3\omega/2}^4 = \xi_3^{xxxx} (3\omega) E_{3\omega/2}^3$, which corresponds to an equal number of carriers injected by 2-photon absorption and 3-photon absorption. If this holds,

$$v_{\text{swarm}} = \frac{|\eta_{3\omega/2}^{\text{xxx}}(3\omega)|}{|\epsilon| \sqrt{\xi_3^{xxxx}(3\omega) \xi_2^{xxx}(3\omega)}}. \quad (31)$$

In Fig. 9 we plot this expression, together with the expression that would result if the beam of frequency $\omega$ were polarized in the $\hat{x}$ direction while the one of frequency $3\omega/2$ in the $\hat{y}$ direction, which is the same as $E_1^{\text{xxx}}$ but with $\eta_{3\omega/2}^{\text{xxx}}(3\omega)$ replaced by $\eta_{3\omega/2}^{\text{yyyy}}(3\omega)$; as well, $\xi_2^{xxx}(3\omega)$ should also be replaced by $\xi_2^{yyyy}(3\omega)$, but they are equal. We see that different stoichiometries give similar values for the swarm velocity if the frequency is adjusted according to the electronic band gap of the system. The fact that higher Ga concentrations lead to larger injected currents (see Fig. 7) is only due to a higher carrier injection. Yet with appropriate laser intensities it is possible to reach the same levels of injected current densities with any Al concentration, although the laser frequencies and intensities at which the maximum is achieved depend on the Al concentration.

We point out that the 2+3 QuIC swarm velocity is about twice its equivalent for 1+2 QuIC. This is an indication that the distribution of carriers injected in the BZ is sharper for 2+3 QuIC compared to 1+2 QuIC. We further confirm that by computing the variance of the lattice momentum $k$ of the electrons injected in the conduction band for both 1+2 QuIC, $\sigma_{1+2}^a = \langle (k^a)^2 \rangle_{1+2} - \langle k^a \rangle_{1+2}^2$, with

$$\xi_{1+2}^{xxx} (3\omega) E_{3\omega/2}^{4} = \xi_{3}^{xxxx} (3\omega) E_{3\omega/2}^{3},$$

in which $E_{3\omega/2}^{4}$ and $E_{3\omega/2}^{3}$ are the total photon energies for 2+3 and 1+2 QuIC, respectively.
and 2+3 QuIC, \( \sigma_{2+3}^2 = \langle \epsilon^2 \rangle_{2+3} - \langle \epsilon \rangle_{2+3}^2 \). For the incident fields polarized along the \( \hat{x} \) direction, we find \( \langle \epsilon \rangle_{1+2} = (4.9, 0, 0) \times 10^{-2} \text{A}^{-1} \) and \( \langle \epsilon \rangle_{2+3} = (5.7, 0, 0) \times 10^{-2} \text{A}^{-1} \), as well as \( \sigma_{2+3} = (3.4, 4.3, 4.3) \times 10^{-3} \text{A}^{-2} \) and \( \sigma_{2+3} = (2.8, 2.2, 2.2) \times 10^{-3} \text{A}^{-2} \), which indeed indicates that the distribution of injected electrons in the BZ is sharper for 2+3 QuIC, especially in the directions transverse (\( \hat{y} \) and \( \hat{z} \)) to the polarization of the field.

2. Laser intensities

Our calculations are performed in the perturbative regime, the validity of which requires that the fraction of the injected carrier population density relative to the total density of states \( n_{\text{max}} \) in the range of energies covered by the laser pulse be small. We thus consider our calculations to be valid when

\[
\langle n \rangle_2 + \langle n \rangle_3 < 0.1n_{\text{max}}, \tag{32}
\]

where the fraction 0.1 is chosen somewhat arbitrarily. The carrier injection due to the 2- and 3-photon interference \( \langle n \rangle_{2+3} \) mostly has the effect of concentrating the carrier injection in some regions of the BZ, but it does not contribute significantly to the total number of injected carriers compared to \( \langle n \rangle_2 \) and \( \langle n \rangle_3 \). For the estimates of laser intensities we consider the incident fields to be both polarized along the \( \hat{x} \) direction, so for a laser pulse of duration \( T \) we require

\[
\left[ \frac{d}{dt} \langle n \rangle_2 + \frac{d}{dt} \langle n \rangle_3 \right] T = 0.1n_{\text{max}}, \tag{33}
\]

\[
\left[ \xi_{2}\times (3\omega) \langle E \rangle_{3\omega/2} + \xi_{3}\times (3\omega) \langle E \rangle_{0} \right] T = 0.1n_{\text{max}}. \tag{34}
\]

For optimal interference, there should be equal densities of carriers injected by 2- and 3-photon absorption, \( \langle n \rangle_2 = \langle n \rangle_3 \), which according to Eq. (34) gives

\[
\xi_2 (3\omega) E_{3\omega/2}^4 = \xi_3 (3\omega) E_{0}^6 < 0.05 \frac{k_{\Omega}^2}{\pi v_{\Omega} T^2}. \tag{36}
\]

The maximal amplitudes \( E_{\omega} \) and \( E_{3\omega/2} \) of the incident fields can then be estimated from the extreme of the inequality in the above equation. For the stoichiometry of \( \alpha = 0.2 \), pulses with duration \( T = 150 \text{fs} \), and total photon energy \( h\Omega = 2.4 \text{ eV} \), we have

\[
E_{\omega} = 1.24 \times 10^5 \frac{V}{m}, \tag{37}
\]

\[
E_{3\omega/2} = 6.05 \times 10^7 \frac{V}{m}. \tag{38}
\]

The intensities in the material medium with these field amplitudes are

\[
I_{\omega} = 2\epsilon_0 c n_\omega E_{\omega}^2 = 26.5 \text{ GW cm}^2, \tag{39}
\]

\[
I_{3\omega/2} = 2\epsilon_0 c n_{3\omega/2} E_{3\omega/2}^2 = 6.54 \text{ GW cm}^2. \tag{40}
\]

For these values, the injected current density is

\[
\langle J^x \rangle = 2n_{2+3}^\times (3\omega) E_{3\omega/2}^2 E_{3\omega/2}^2 T = 6.25 \text{ MA cm}^{-2}. \tag{41}
\]

We emphasize that these are just estimates, as the limit of carrier density is set somewhat arbitrarily in Eq. (32). We note that we are ignoring scattering of the injected carriers. This means that the true maximal intensities would be larger than our estimates here, since there is room for more photon absorption as scattering depletes some of the excited states. We also note that in this treatment the electron-electron interaction has been neglected; were it included, the phase parameter would be shifted. However this shift is usually very small for zincblende semiconductors, except for frequencies very close to the band gap.

V. DISCUSSION AND CONCLUSION

One of the main utilities of QuIC in semiconductors is the injection of carriers in localized regions of the BZ. In this respect 2+3 QuIC performs better than 1+2 QuIC, and that can be seen in a higher swarm velocity for 2+3 QuIC, which is a desirable feature for current injection. Another interesting difference between 1+2 and 2+3 QuIC is that in 2+3 QuIC several current injection coefficients change sign as the total photon energy is increased, while in 1+2 QuIC they typically do not. This happens because interband velocity matrix elements are responsible for the largest contribution to the 3PA coefficient at low photon energies, but at higher photon energies the intraband velocity matrix elements dominate. Since only nonlinear optical processes are involved...
in 2+3 QuIC, the laser intensities required for maximal effect are higher than for 1+2 QuIC, but still moderate. Also, the optical fields have a power law attenuation as they propagate through the absorbing material, instead of the exponential attenuation of linear absorption. Thus a waveguide geometry is desirable, and while QuIC in waveguides presents some challenges, as it raises issues of phase- and mode-matching, it also presents opportunities for easy integration with devices on-chip. Since optical frequency combs are routinely propagated through waveguides, other than the problems of phase- and mode-matching, 2+3 QuIC experiments in waveguides has no additional hurdles.

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Appendix A: Nonzero injection coefficient components of zincblende lattices

AlGaAs in the virtual crystal approximation forms a zincblende lattice, which has the symmetry of point group $T_d$ (or 43m). The optical responses we consider in this work involve tensors of rank 4 up to 6. With $T_d$ symmetries, generic rank-4 tensors have 21 non-zero components of which 4 are independent, rank-5 tensors have 60 non-zero and 10 independent components, and rank-6 tensors have 183 non-zero and 31 independent components. However, the tensors representing the optical processes have a few more specific restrictions due to their relation to the optical fields, as the indices associated with the same incident field are symmetrized.

With these considerations the tensor $\xi_2$ has 3 independent components

\begin{align}
\xi_2^{xxx} &= P(x, y, z), \\
\xi_2^{xyy} &= \xi_2^{yxy} = \xi_2^{yxy} = P(x, y, z), \\
\xi_2^{xyx} &= P(x, y, z),
\end{align}

where $P(x, y, z)$ indicates all the possible permutations of $(x, y, z)$ in the indices. The tensor $\xi_{2+3}$ has 3 independent components

\begin{align}
\xi_{2+3}^{xxxyy} &= P(x, y, z), \\
\xi_{2+3}^{xxxyy} &= \xi_{2+3}^{xxxyy} = \xi_{2+3}^{xxxyy} = P(x, y, z), \\
\xi_{2+3}^{xxxyy} &= \xi_{2+3}^{yxyy} = \xi_{2+3}^{yxyy} = \xi_{2+3}^{yxyy} = P(x, y, z), \\
\xi_{2+3}^{xyxx} &= \xi_{2+3}^{xyxx} = \xi_{2+3}^{xyxx} = P(x, y, z), \\
\xi_{2+3}^{xyxx} &= \xi_{2+3}^{xyxx} = \xi_{2+3}^{xyxx} = P(x, y, z),
\end{align}

and $\xi_3$ has 5 independent components

\begin{align}
\xi_3^{xxxxx} &= P(x, y, z), \\
\xi_3^{xyy} &= \xi_3^{xyy} = P(x, y, z), \\
\xi_3^{xyy} &= \xi_3^{xyy} = \xi_3^{xyy} = \xi_3^{xyy} = \xi_3^{xyy} = P(x, y, z), \\
\xi_3^{xyy} &= \xi_3^{xyy} = \xi_3^{xyy} = \xi_3^{xyy} = \xi_3^{xyy} = P(x, y, z), \\
\xi_3^{xyy} &= \xi_3^{xyy} = \xi_3^{xyy} = \xi_3^{xyy} = P(x, y, z).
\end{align}

Finally, the tensor $\eta_{2+3}$ has 9 independent components

\begin{align}
\eta_{2+3}^{xxxxx} &= P(x, y, z), \\
\eta_{2+3}^{xyy} &= P(x, y, z), \\
\eta_{2+3}^{xyy} &= \eta_{2+3}^{xyy} = \eta_{2+3}^{xyy} = P(x, y, z), \\
\eta_{2+3}^{xyy} &= \eta_{2+3}^{xyy} = \eta_{2+3}^{xyy} = \eta_{2+3}^{xyy} = P(x, y, z), \\
\eta_{2+3}^{xyy} &= \eta_{2+3}^{xyy} = \eta_{2+3}^{xyy} = \eta_{2+3}^{xyy} = \eta_{2+3}^{xyy} = P(x, y, z), \\
\eta_{2+3}^{xyy} &= \eta_{2+3}^{xyy} = \eta_{2+3}^{xyy} = \eta_{2+3}^{xyy} = \eta_{2+3}^{xyy} = \eta_{2+3}^{xyy} = P(x, y, z), \\
\eta_{2+3}^{xyy} &= \eta_{2+3}^{xyy} = \eta_{2+3}^{xyy} = \eta_{2+3}^{xyy} = \eta_{2+3}^{xyy} = \eta_{2+3}^{xyy} = P(x, y, z).
\end{align}

The independent components shown here are the ones plotted in the figures in the main text.

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