Resonant generation of difference harmonics due to intersubband transitions in a biased superlattice

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The difference harmonic susceptibility due to intersubband transitions in a superlattice subjected to uniform electric field is calculated. The transitions between uncoupled ground states and tunnel-coupled excited states are considered in the framework of the effective mass approximation. The spectral dependencies of the susceptibility and modification of the nonlinear response with variation of the applied voltage are presented. The efficiency of transformation of the double-frequency near-IR pump into THz radiation is also discussed and the enhancement of transformation due to the THz waveguide effect has been found.

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I. INTRODUCTION

The examination of nonlinear response due to intersubband transitions in various semiconductor heterostructures has been intensively carried out over the past decade. The giant second-order response appears in nonsymmetric heterostructures both due to the big dipole matrix elements of the confined electrons and to possibility for realization of double resonance conditions. The frequency doubling process at mid-IR wavelengths was reported for QW under an electric field[1] and asymmetric QWs.[2] The cases of transitions in the valence band[3] and in the self-assembled quantum dots[4] were also examined. Recently, the difference-harmonic generation has been investigated both due to intersubband (see experimental data and theoretical calculations in Refs. 5 and 6, respectively), and interband transitions.[7] To the best of our knowledge, the difference-harmonic generation due to intersubband transitions in a superlattice subjected to uniform electric field (biased superlattice, BSL) has not yet been considered. We have examined this variant of nonlinear response for BSL with tunnel uncoupled ground states and weakly coupled excited states described in the tight-binding approximation. The efficiency of the down-conversion process under consideration is also discussed for the case of BSL placed in the center of the resonator for THz radiation.

The energy band diagram of BSL being pumped by two beams with photon energies $\hbar\omega_1,\hbar\omega_2$ and the scheme of excitation of BSL subjected in the waveguide to resonant frequencies corresponding to the difference-harmonic frequency are shown in Fig. 1. The difference-harmonic susceptibility was calculated under double resonant conditions and the induced nonlinear polarization appear to be oriented along the SL growth direction. Both spectral and bias-voltage dependencies of the susceptibility occur as sharp nonmonotonic functions of the difference between the energies of pumping photons and the level-splitting energy. This energy is equal to the Bloch energy, $\varepsilon_f=|e|F_\perp l$, which determines a discrete set of equidistant levels in the BSL with the period l (the Wannie-Stark ladder under a homogeneous electric field $F_\perp$).[8]

Thus, we consider the down-conversion transformation of a two-color mid-IR pump into the THz signal. An enhancement of the above-described transformation due to the THz waveguide effect is also discussed.[9] The THz resonator can be formed by the heavy-doped top and back gates, which are transparent for the mid-IR pump and which reflect the THz radiation [see Fig. 1(b)], or by the top and bottom metallic mirrors. The efficiency of transformation is estimated based on the electrodynamical description of such ideal resonator with the BSL placed in the center of the structure and with the model of energy losses introduced through the complex dielectric permittivity inside the THz waveguide.

The paper is organized as follows. In Sec. II we evaluate the third-order susceptibility of electrons using the tight-binding excited states and the uncoupled ground states; the spectral and gate-voltage dependencies are also presented. The numerical results for efficiency of down-conversion transformation based on the electrodynamical consideration of the THz waveguide is performed in Sec. III. In conclusion, we discuss the assumptions used in the paper, as well as the possibility of using superlattices for THz generation.

II. NONLINEAR SUSCEPTIBILITY

The calculations below are based on the one-particle description of electron states in BSL. The general expression for the nonlinear susceptibility tensor of the third-order, $\chi_{\alpha\beta\gamma}$, which describes the generation of the difference harmonic, is written below on the basis of the eigenstates problem $\hat{h}|\nu\rangle=\varepsilon_\nu|\nu\rangle$. Here $\hat{h}$ is the effective mass Hamiltonian for BSL (see Refs. 8 and 10), $|\nu\rangle$ and ε_ν are the eigenstate vector and the energy in SL under a uniform electric field, respectively. Consideration of the second order response on perturbation $(ie/\omega_{1,2})\mathbf{E}_{1,2}\cdot\hat{\mathbf{v}}$, where $\mathbf{E}_{1,2}$ are the strengths of electric field of the first and the second beams and $\hat{\mathbf{v}}$ is velocity operator, gives us the susceptibility tensor, $\chi_{\alpha\beta\gamma}(\omega_1,\omega_2)$, in the form (see Refs. 11 and 7)

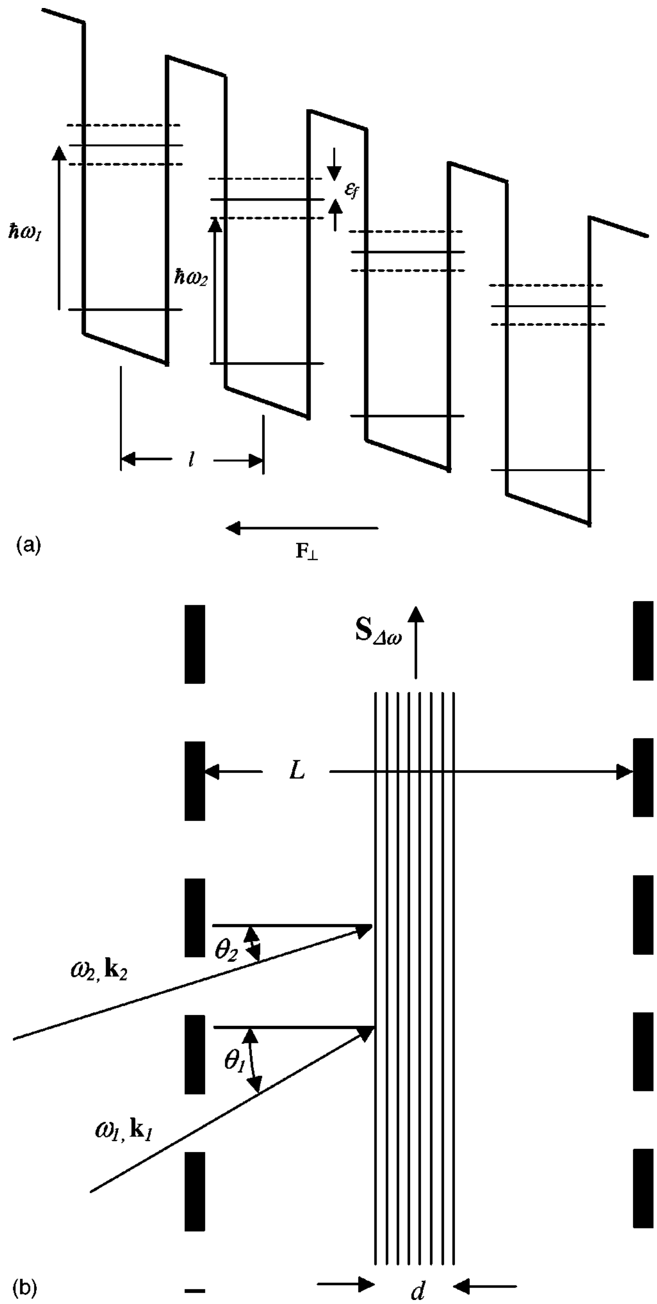

FIG. 1. The energy diagram of BSL under resonant two-frequency pump (a) and a geometry of the THz waveguide with excitation scheme (b).

$$\chi_{\alpha\beta\gamma}(\omega_1,\omega_2)=\frac{i2e^3}{\Delta\omega\omega_1\omega_2L^3}\sum_{\nu\nu'\nu''}(f_\nu-f_{\nu'})\times\left\{\Theta_{\alpha\beta}(\nu,\nu',\nu'')\frac{\langle\nu'|\hat{v}_\gamma|\nu\rangle}{\varepsilon_{\nu'\nu}+\hbar\omega_2-i\gamma}+\Theta_{\alpha\gamma}(\nu,\nu',\nu'')\frac{\langle\nu'|\hat{v}_\beta|\nu\rangle}{\varepsilon_{\nu'\nu}-\hbar\omega_1-i\gamma}\right\}. \quad (1)$$

Here, $\Delta\omega=\omega_1-\omega_2$ is the difference frequency ($\Delta\omega>0$), $\langle\nu'|\hat{v}_\alpha|\nu\rangle$ is the velocity matrix element, α,β,γ are the Cartesian coordinate indices, and L^3 is the normalization volume. Besides the summations over ν,ν' and ν'' include all the BSL states and $\varepsilon_{\nu'\nu}\equiv\varepsilon_{\nu'}-\varepsilon_\nu$. The $\Delta\omega$-depending factor, $\Theta_{\alpha\beta}$, has the form:

$$\Theta_{\alpha\beta}(\nu,\nu',\nu'')=\frac{\langle\nu|\hat{v}_\alpha|\nu''\rangle\langle\nu''|\hat{v}_\beta|\nu'\rangle}{\varepsilon_{\nu''\nu}-\hbar\Delta\omega-i\gamma}-\frac{\langle\nu|\hat{v}_\beta|\nu''\rangle\langle\nu''|\hat{v}_\alpha|\nu'\rangle}{\varepsilon_{\nu'\nu''}-\hbar\Delta\omega-i\gamma}, \quad (2)$$

where the transition broadening, γ, is a phenomenological parameter which is supposed to be independent of the quantum numbers, i.e., the broadening is the same for all transitions.

The wave function for the tunnel-uncoupled ground states (labeled 1 below) is written as $\varphi^{(1)}_{z-sl}$, where $\varphi^{(1)}_z$ is the orbital of the ground state in the quantum well centered at $z=0$ and $s=0,\pm1,\pm2\ldots.$ The index s means as well the number of the quantum level for the case of tunnel-uncoupled ground states and the energy of sth level is written as $\varepsilon_s=s\varepsilon_f$. Taking into account the in-plane kinetic energy of electrons, we write the dispersion law in the form $\varepsilon_{sp}=s\varepsilon_f+\varepsilon_p$ with $\varepsilon_p=p^2/2m$, where $\mathbf{p}$ is the 2D-momentum and m is the electron effective mass. The wave function for the weakly-coupled excited states (labeled 2 below) is written as a superposition of the orbitals: $\Sigma_r\Psi^{(q)}_r\varphi^{(2)}_{z-rl}$. Here $r=0,\pm1,\pm2\ldots,\varphi^{(2)}_z$ is the orbital of the excited electron state in the quantum well centered at $z=0$, and q are the quantum numbers of the excited states under consideration. The column-vector $\Psi^{(q)}$ is determined by the matrix eigenvalue problem, and the wave function is written as (see Ref. 7,8 for details)

$$N_q\sum_r J_{q-r}(2T/\varepsilon_f)\varphi^{(2)}_{z-rl}, \quad (3)$$

where $J_r(x)$ is the Bessel function, T is the tunnel matrix element for the weakly-coupled states in the adjacent quantum wells, and N_q is the normalization factor (since $N_q^2\simeq1$ for the weak interwell tunneling case this factor is omitted below). The dispersion law for the excited states is given by $\varepsilon_{qp}=\varepsilon_{21}+q\varepsilon_f+\varepsilon_p$, where $q=0,\pm1,\pm2,\ldots$ and ε_{21} is the interlevel energy in a single QW.

The nonzero matrix elements of transverse velocity between excited states q and q' are given by

$$\langle q|\hat{v}_z|q'\rangle=-i\frac{Tl}{\hbar}(q-q')\delta_{|q-q'|,1}, \quad (4)$$

while the matrix elements between the ground and excited states (s and q, respectively) are

$$\langle q|\hat{v}_z|s\rangle=-\langle s|\hat{v}_z|q\rangle\simeq i\frac{\varepsilon_{21}Z_{21}}{\hbar}J_{q-s}(2T/\varepsilon_f). \quad (5)$$

Here $Z_{21}\equiv\int dzz\varphi^{(2)}_z\varphi^{(1)}_z$ is the interwell matrix element of coordinate. Since the problem is translation-invariant along the 2D-plane, the zzz-component of susceptibility tensor is nonzero only. Using the double resonant condition, for the pump frequencies close to the interlevel energy distance ($\hbar\omega_{1,2}\approx\varepsilon_{21}$), and taking into account the matrix elements of the transverse velocity Eqs. (4) and (5), we transform Eqs. (1), (2) into

$$\chi_{zzz}(\omega_1,\omega_2)\simeq\frac{|e|^3\varepsilon_{21}^2Z_{21}^2T}{\hbar^3\Delta\omega\omega_1\omega_2}\frac{n_e l}{\varepsilon_f-\hbar\Delta\omega-i\gamma}\times\sum_q J_q(2T/\varepsilon_f)J_{q-1}(2T/\varepsilon_f)\times\{(\hbar\omega_1-\varepsilon_{21}-q\varepsilon_f+i\gamma)^{-1}-(\hbar\omega_2-\varepsilon_{21}-(q-1)\varepsilon_f-i\gamma)^{-1}\},\quad(6)$$

where n_e is the electron concentration in BSL.

The numerical calculations of nonlinear susceptibility (6) have been performed for $GaAs/Al_{0.33}Ga_{0.67}As$ BSL with period $l=14$ nm and barrier width 8.5 nm (so that the energy distance between the ground state and excited states without tunnel-coupling is $\varepsilon_{21}\simeq120$ meV), with the electron concentration 7.1×10^{17} cm^{-3} and broadening energy γ supposed by equal to 2 meV. For the parameters under consideration, we obtain $Z_{21}\simeq15$ nm and $T\simeq5$ meV. The spectral dependencies for the real and imaginary parts of χ_{zzz} and for the absolute value of the susceptibility versus the difference energy $\hbar\Delta\omega$ and versus the detuning energy, which are introduced as $\delta\varepsilon\equiv(\hbar\omega_1+\hbar\omega_2)/2-\varepsilon_{21}$, are shown in Figs. 2(a), 2(b) and 2(c), respectively. These spectral dependences are symmetric with respect to $\delta\varepsilon$, if we neglect the factor $1/\Delta\omega$ in Eq. (6). A set of the peaks at $\hbar\Delta\omega=\varepsilon_f$ and $\delta\varepsilon=(q+1/2)\varepsilon_f$ [which correspond to $\hbar\omega_1=\varepsilon_{21}+(q+1)\varepsilon_f$ and $\hbar\omega_2=\varepsilon_{21}+q\varepsilon_f$] with oscillator strengths proportional to $J_q(2T/\varepsilon_f)J_{q+1}(2T/\varepsilon_f)$ has been obtained. For the weak coupling case, the nonlinear response at the difference harmonic is only found to be substantial for pump frequencies close to ε_{21} and $\varepsilon_{21}\pm\varepsilon_f$.

The dependence of the absolute value of χ_{zzz} versus the difference energy $\hbar\Delta\omega$ and the Bloch energy ε_f is presented in Fig. 3. The only major level-splitting peak at $\varepsilon_f=\hbar\Delta\omega$ and some oscillations are obtained due to transformation of the discrete energy levels into minibands under decreasing ε_f. As one can see from Figs. 2 and 3, the generation of a difference harmonic in BSL is very sensitive to the pump frequencies and to the biased voltage. The maximum value of susceptibility obtained in our calculation is about $|\chi_{\max}|\approx2.4\times10^{-4}$ cm/V. This maximum susceptibility appears to be up to two orders of magnitude higher than the result for the case of interband excitation.[7] This is due to decrease of the pump quanta energy, which gives an additional factor $(\varepsilon_g/\varepsilon_{21})^2$ (ε_g is the interband gap), and due to greater contributions under the double resonant condition.[12] The maximum susceptibility also exceeds the experimental result for the structures measured due to the absence of the wide passive layers between the tunnel-coupled quantum wells used in Ref. 5.

III. EFFICIENCY OF DOWNCONVERSION

In this section we study the efficiency of transformation of the intersubband pump into the THz signal, taking into account the waveguide-induced enhancement of the downconversion process. We consider two IR beams with incidence angles θ_1 and θ_2 ($\theta_1\simeq\theta_2$) which excite the intersubband transitions in biased SL with width d replaced into the resonator with the dielectric permittivity ϵ and with width L [see Fig. 1(b)]. The THz waveguide is formed by the doped regions $|z|>L/2$ with the plasma frequency Ω_p; we suppose $\Delta\omega\ll\Omega_p\ll\omega_{1,2}$ so that the structure is transparent for mid-IR radiation while THz radiation is confined inside $|z|<L/2$. An another possibility is to use the metallic mirrors and introducing IR-pump through an oblique edge of the sample. The THz wave is propagated along in-plane direction OX and a transverse distribution of THz field $E_z\exp(i\Delta\omega t-i\Delta kx)$ is determined from the wave equation

$$\left[\frac{d^2}{dz^2}-\Delta k^2+\epsilon\left(\frac{\Delta\omega}{c}\right)^2\right]E_z=\begin{cases}-4\pi(\Delta\omega/c)^2P_\perp, & |z|<d/2\\ 0, & |z|>d/2\end{cases}\quad(7)$$

with the boundary conditions: $E_{z=\pm L/2}=0$. The in-plane wave vector Δk is determined as follows $\Delta k=k_1\sin\theta_1-k_2\sin\theta_2$ and induced polarization $P_\perp$ is expressed through the susceptibility χ_{zzz} according to $P_\perp=\chi_{zzz}E_1E_2$, where E_1 and E_2 are the z-components of the electric field strengths for first and second beams. The energy losses in the resonator is introduced through a small complex addition to dielectric permittivity $\epsilon=\epsilon'+i\epsilon''$ at the difference frequency $\Delta\omega$.

Introducing $\kappa^2=\Delta k^2-\epsilon(\Delta\omega/c)^2$ and using the additional boundary conditions at biased SL (under the inequality $|\kappa|d\ll1$) we obtain the system of ordinary differential equations inside the waveguide regions:

$$\left(\frac{d^2}{dz^2}-\kappa^2\right)E_z=0,\quad\frac{d}{2}<|z|<\frac{L}{2}\quad(8)$$

with the four boundary conditions:

$$\left.\frac{dE_z}{dz}\right|_{-0}^{0}=-4\pi\left(\frac{\Delta\omega}{c}\right)^2\int_{(SL)}dzP_\perp,\quad E_z|_{-0}^{0}=0,$$

$$E_{z=\pm L/2}=0,\quad(9)$$

where we have used $d\to0$. The straightforward calculations give us the distribution of the transverse component of THz field

$$E_z=\frac{\overline{E}}{\cosh(\kappa L/2)}\begin{cases}-\sinh(\kappa(z+L/2)), & 0>z>-L/2\\ \sinh(\kappa(z-L/2)), & L/2>z>0\end{cases},\quad(10)$$

where we have introduced the characteristic field $\overline{E}=2\pi(\Delta\omega/c)^2d\overline{\chi}E_1E_2/\kappa$.

In order to estimate the maximum efficiency of transformation, we use in Eq. (9) the peak value of susceptibility $\overline{\chi}$, which is realized under double-resonant conditions. The

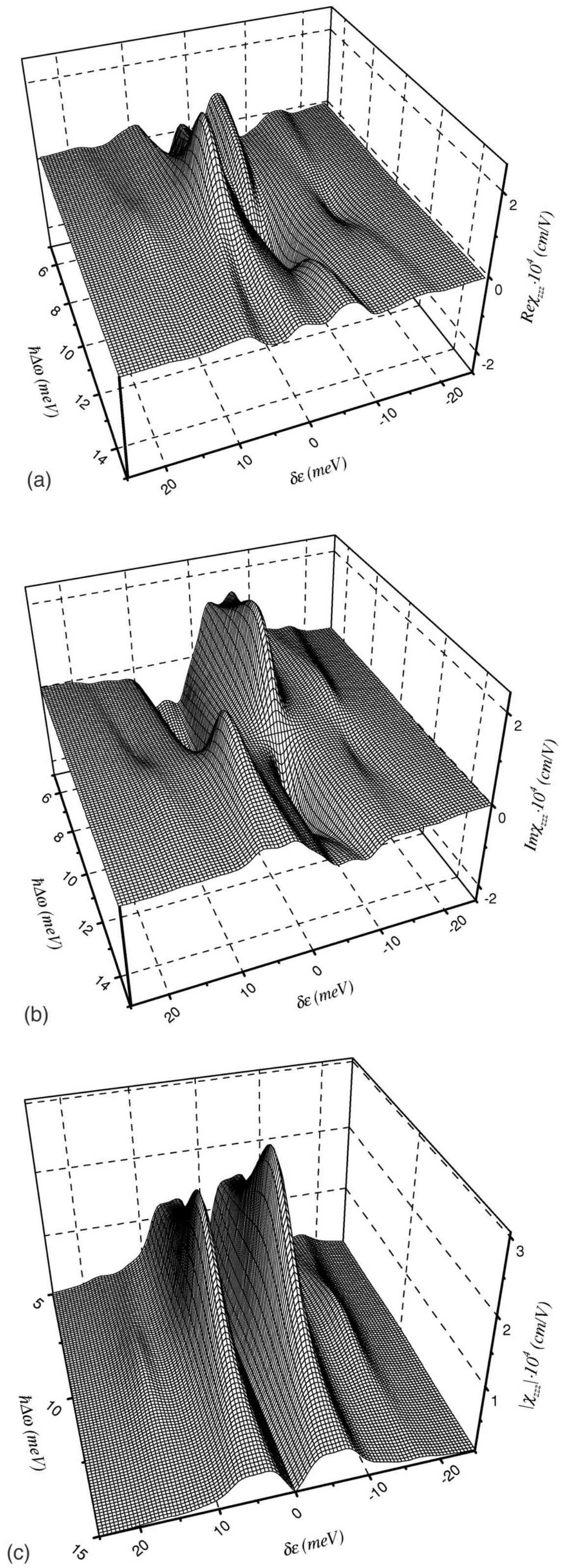

FIG. 2. The spectral dependencies of a real part (a) and an imaginary part (b) of χ_{zzz} and $|\chi_{zzz}|$ (c) versus the difference energy $\hbar\Delta\omega$ and the detuning energy $\delta\varepsilon$.

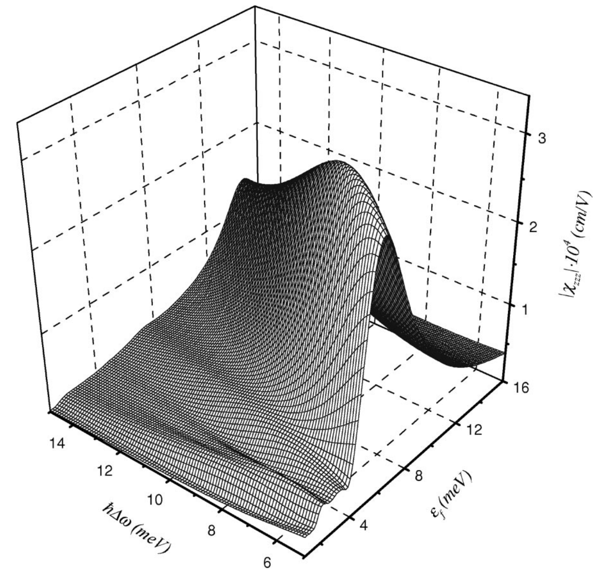

FIG. 3. The spectral dependence of $|\chi_{zzz}|$ versus the difference energy $\hbar\Delta\omega$ and the Bloch energy ε_f.

Poynting vector for the propagated along OX axis THz radiation is given by (see Ref. 7)

$$S_z = \frac{c^2 \Delta \mathbf{k}}{2\pi\Delta\omega}|E_z|^2. \tag{11}$$

The total energy flow along BSL, $\overline{S} = \int_{-\infty}^{\infty} dz S_z$, is obtained from Eqs. (9)–(10) as follows (this value is measured in W/cm while the Poynting vectors of pump beams, $S_{1,2}$ are measured in W/cm^2)

$$\overline{S} = \frac{c^2 \Delta k}{2\pi\Delta\omega}\int dz |E_z|^2 = \frac{c^2 \Delta k}{2\pi\Delta\omega}\frac{|\overline{E}|^2}{\kappa'}\mathcal{F}(\kappa L), \tag{12}$$

where dimensionless function $\mathcal{F}(\kappa L)$ is written as

$$\mathcal{F}(\kappa L) = \frac{\sinh(\kappa' L) - (\kappa'/\kappa'')\sin(\kappa'' L)}{\cosh(\kappa' L) + \cos(\kappa'' L)}, \tag{13}$$

and we have used the complex factor $\kappa' + i\kappa'' \equiv \sqrt{\Delta k^2 - \epsilon(\Delta\omega/c)^2}$. The function (13) versus $\kappa' L$ and $\kappa'' L$ is shown in Fig. 4. If $\kappa' L \gg 1$, the function $\mathcal{F}(\kappa L)$ is replaced by unity. The simplified expression for $\mathcal{F}(\kappa L)$ is obtained for the case of high-quality resonator, if $\sqrt{\epsilon'(\Delta\omega/c)^2 - \Delta k^2} \equiv \tilde{\kappa} \gg \sqrt{\epsilon''}\Delta\omega/c$. Under this inequality $\kappa' \simeq -\epsilon''(\Delta\omega/c)^2/2\tilde{\kappa}$ and Eq. (13) is transformed into

$$\mathcal{F}(\kappa L) \simeq \frac{\kappa'}{\tilde{\kappa}}\frac{\tilde{\kappa}L - \sin(\tilde{\kappa}L)}{2\cos^2(\tilde{\kappa}L/2) + (\kappa')^2/2}. \tag{14}$$

As a result, the maximal value of the energy flow, under the resonant condition $\tilde{\kappa}L/2 = \pi n$ with $n = 0,1,\ldots$, is determined by

$$S_{\max} = \frac{c^2 \Delta k}{2\pi\Delta\omega}\frac{|\overline{E}|^2}{\kappa'^2 L}\alpha_n \tag{15}$$

with $\alpha_n = (2\pi n - 1)/2\pi n$.

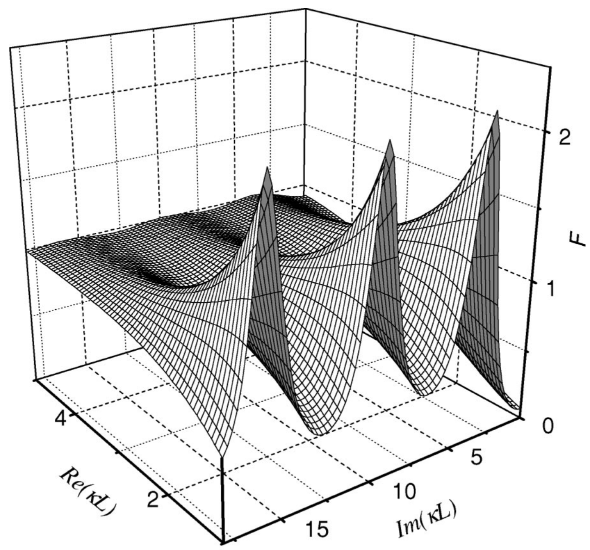

FIG. 4. The function $\mathcal{F}(\kappa L)$ of Eq. (13) versus real part $\kappa' L$ and imaginary part $\kappa'' L$ of an argument κL.

The output signal appears to be essentially dependent on the THz waveguide effect due to the modulation of spectral dependences of susceptibility caused by the function $\mathcal{F}(\kappa L)$. In Fig. 5(a), we present $\overline{S}$ versus the difference energy $\hbar\Delta\omega$ for the 100-period BSL with the total width $d=1.4\times10^{-4}$ cm under different ε_f. The incident angles, Θ_1 and Θ_2 are chosen as 15° and 10°, respectively. These spectral dependencies are plotted for the same BSL parameters as in Sec. II, $\hbar(\omega_1+\omega_2)/2\simeq125$ meV, and the pump powers $S_{1,2}$ are equal 1.7 kW/cm^2. The width of resonator is chosen as $L=26.7$ μm, while the dielectric losses were determined by the ratio $\epsilon''/\epsilon'\approx0.03$. The presented results demonstrate an essential [more than one order of magnitude; see, the dotted-dashed curve for $L\to\infty$ in Fig. 5(a)] enhancement of output power due to the THz waveguide effect under the resonant condition $\hbar\Delta\omega\simeq\varepsilon_f$. In addition, Fig. 5(b) shows an essential sensitivity of the output power on parameters used for the case of wide resonator with $L=400$ μm. The maximal output energy flow appears to be up to 0.12 W/cm under the optimal conditions.

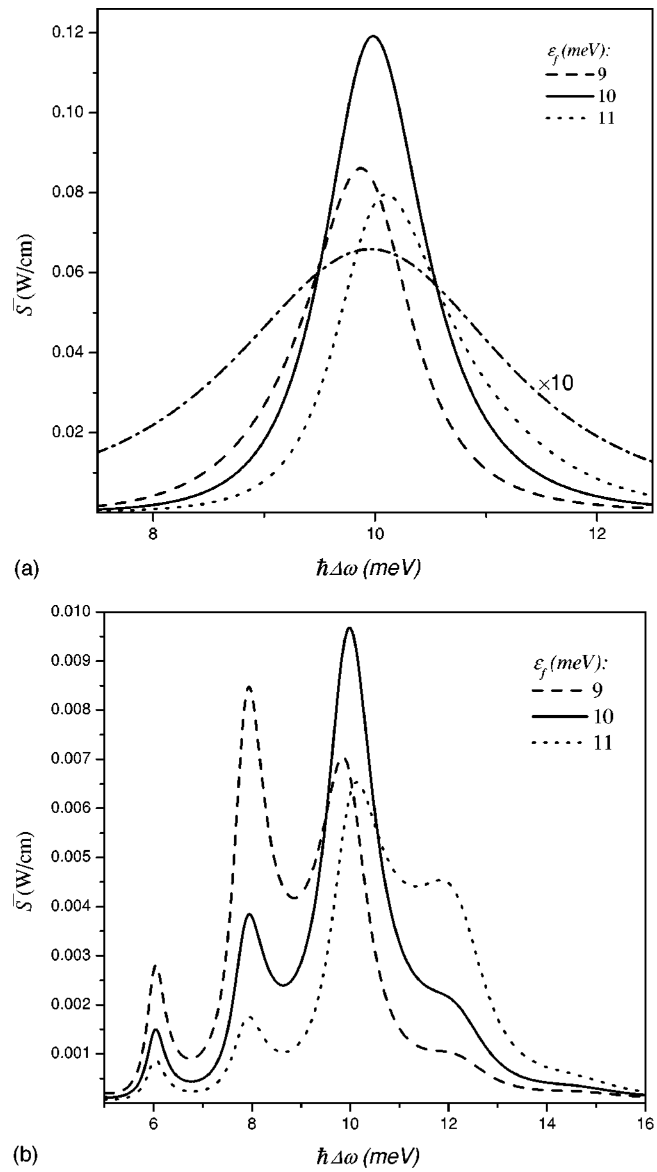

FIG. 5. The spectral dependence of the output power, $\overline{S}$, versus the difference energy $\hbar\Delta\omega$ at different Bloch energies ε_f for two THz waveguide widths: 26.7 μm (a); 400 μm (b).

IV. CONCLUSION

In summary, we have obtained and analyzed the difference harmonic susceptibility due to intersubband transitions in the biased superlattice. Both the double-resonant enhancement of the susceptibility and the THz waveguide enhancement of the down-conversion process were considered. The typical values of the susceptibility is substantially higher than for the other mechanisms reported in Refs. 5–7 and the efficiency of transformation increases substantially around the resonances of THz waveguide. As a result the output THz signal appears to be noticeable both due to an intensive mid-IR pump from the CO_2 laser and due to enhancement of the efficiency of down-conversion in a high-quality resonator. Note, that the similar enhancement of the down-conversion process due to the THz waveguide effect is possible for the case of the interband pump considered in Ref. 7.

Let us discuss the main assumptions used in the calculations. Both the effective mass approximation under the description of the energy spectrum and of the matrix elements (4), (5) as well as the phenomenological consideration of the broadening with the characteristic energy γ, which is the same for all transitions under consideration, are generally accepted. We consider the ground states as uncoupled because the corresponding tunnel matrix element is about 1 meV for the BSL parameters used in Sec. II. The restrictions of the tight-binding approach for consideration of the excited electron states are also well known[8] and errors are small in the case under consideration. We can also neglect all the electron-electron effects on the intersubband transitions, if the broadening energy $\gamma\geqslant2$ meV. According to Ref. 13, the splitting of intersubband transitions in BSL due to a nonlocal exchange interaction is possible if $\gamma \leqslant 1$ meV and more complicated consideration should be pursued for this case. Thus, except of the high-mobility BSL, the above listed approximations do not change typical the character of spectral and electric field dependencies for the susceptibility under consideration. Next, we have used the simplified THz resonator model with complex dielectric permittivity and ideal boundary condition at $z = \pm L/2$. The ratio, $\epsilon''/\epsilon' = 3 \times 10^{-2}$, we used, overestimates the phonon contributions to damping[14] but this value is used to include other possible damping mechanisms such as: (a) mirror losses due to electron absorption; (b) elastic scattering of modes caused by the geometrical imperfection of the structure; and (c) losses in the active region due to the absorption of electrons in BSL. A special consideration of these processes is necessary for a more precise description of the THz waveguide enhancement effect.

In conclusion, we believe that the presented results have demonstrated that the BSL in the THz waveguide is a promising structure for efficient down-conversion of two-color mid-IR pump from CO_2 laser into THz radiation, and it is substantially more efficient than earlier suggested schemes.[5–7]

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