Novel infrared generation in low-dimensional semiconductor heterostructures via quantum coherence

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A new scheme for infrared generation without population inversion between subbands in quantum-well and quantum-dot lasers is presented and documented by detailed calculations. The scheme is based on the simultaneous generation at three frequencies: optical lasing at the two interband transitions which take place simultaneously, in the same active region, and serve as the coherent drive for the IR field. This mechanism for frequency down-conversion does not rely upon any ad hoc assumptions of long-lived coherences in the semiconductor active medium. And it should work efficiently at room temperature with injection current pumping. For optimized waveguide and cavity parameters, the intrinsic efficiency of the down-conversion process can reach the limiting quantum value corresponding to one infrared photon per one optical photon. Due to the parametric nature of IR generation, the proposed inversionless scheme is especially promising for long-wavelength (far-infrared) operation.

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Low-dimensional semiconductor heterostructures are almost ideally suited for generation in the mid- to far-infrared range (denoted below as IR for brevity), because the spacing between levels of dimensional quantization can be conveniently manipulated over the region from several to hundreds of microns, and injection pumping is possible. There is however a major problem: strong non-resonant losses of the IR field due to free-carrier absorption and diffraction, which become increasingly important at longer far-IR wavelengths. Due to very short lifetime of excited states, it is difficult to maintain a large enough population inversion and high gain at the intersubband transitions, necessary to overcome losses. There were many suggestions to solve this problem by rapid depletion of the lower lasing state using, e.g., the resonant tunneling to adjacent semiconductor layers or transition to yet lower subbands due to phonon emission [1,2], or even stimulated interband recombination [3]; see [4,5] for recent reviews. The successful culmination of these studies is the realization of quantum cascade lasers [2], in which the lower lasing state is depopulated either by tunneling in the superlattice or due to transition to lower-lying levels separated from the lasing state by nearly the energy of a LO-phonon; see e.g. [6] and references therein.

We here put forward another possibility, allowing us to achieve IR generation without population inversion at the intraband transition. This becomes possible with the aid of laser fields simultaneously generated at the *interband* transitions (called optical fields for brevity), which serve as the coherent drive for the frequency down-conversion to the IR. Employing self-generated optical lasing fields provides the possibility of injection current pumping and also removes the problems associated with external drive (beam overlap, drive absorption, spatial inhomogeneity), which were inherent in previous works on parametric down-conversion in semiconductors, see [4] for a review. The second important feature is the great enhancement of nonlinear wave mixing near resonance with intersubband transitions. The third feature is the possibility of canceling the resonance one-photon absorption for the generated IR field due to coherence effects provided by self-generated driving optical fields [7]. The processes incorporating these three features constitute an important field of research with a variety of physical effects and promising applications. We consider just one example of such processes of IR generation and nonlinear mixing with self-generated optical fields in semiconductor heterostructures.

To avoid misunderstanding, we note that the mechanism of IR generation discussed in this paper is different from the approach in which the resonant tunneling and Fano-type interference are used to establish a large coherence at intersubband transitions [8]. These quantum interference ideas imply usually the presence of a long-lived coherence at the intraband transitions. Our approach does not require long dephasing times. We here focus on nonlinear wave mixing phenomenon, which is greatly enhanced near the resonance with intersubband transition. Note that this process has some common features with recently observed generation of coherent IR emission in rubidium vapor in four-wave mixing experiments [9]. Also, coherent microwave generation at the difference frequency under the action of two resonant external optical fields was observed in cesium vapor [10].

Generic three-level scheme. As the simplest case, consider the situation when only three levels of dimensional quantization are involved in generation: one (lowest-lying) heavy-hole level, and two electron levels; see Fig. 1. Of course, this scheme also describes the situation when there are two hole levels and one electron level involved. We need all three transitions to be allowed by selection rules. In a quantum well (QW) this will generally require using asymmetric structures, e.g. rectangular well with different barrier heights. For example, in a $Al_{0.3}Ga_{0.7}As/GaAs/Al_{0.2}Ga_{0.8}As$ QW parametric IR generation is possible either between two electron subbands $1e$ and $2e$ separated by 98 meV ($\lambda \simeq 13$ μm) or between two lowest heavy-hole subbands ($\lambda \simeq 60$ μm).

Symmetric QWs can also be employed. e.g. in the case of a strong coupling between different subbands of heavy and light holes. A typical example is $Al_{0.3}Ga_{0.7}As/GaAs/Al_{0.3}Ga_{0.7}As$ QW in which the second subband of heavy holes $2hh$ and the first subband of light holes $1lh$ happen to be very close to each other (within homogeneous linewidth) in the Γ-point for a wide range of thicknesses $\sim 5-8$ nm, and therefore are strongly mixed. In this case two optical fields correspond to $1e \to 1hh$ and $1e \to 1lh$ transitions. And IR field is generated via the $1hh \to 2hh$ transition.

A third configuration of interest is the quantum dot (QD). For example in a self-assembled InAs/GaAs quantum dot (QD) the three-level scheme can be easily realized with all three transitions allowed [11].

When the injection current density reaches the threshold value $j_{\rm th}$, optical generation starts due to recombination transitions between ground electron and hole states. Upon increasing the pumping current, optical generation can start also from excited states and the laser can be completely switched to lasing from the excited-state which has higher maximum gain due to a larger density of states. The effect of excited-state lasing was studied both in QW and QD lasers [4,11–13]. It was found that with optimized laser parameters the region of simultaneous ground-state and excited- state lasing can be around $j \sim 2j_{\rm th}$ [12,13]. In order to have the region of two-wavelength lasing sufficiently broad ($\Delta j \sim (0.1-0.2)j_{\rm th}$), gains for the two wavelengths should be close to each other.

The presence of one or two strong optical driving fields in the cavity gives rise to a rich variety of *resonant* coupling mechanisms by which the IR field can be produced. Here we will concentrate on one such scheme in which the two coherent optical fields having frequencies ω_1 and ω_2 excite the induce electronic oscillation at the difference frequency $\omega_2-\omega_1$. It is important to note that the coherent IR polarization is parametrically excited independent of the sign of population difference at the IR transition.

The resulting output intensity of IR radiation depends on the coupling coefficient between the IR polarization and the cavity modes. It is clear that polarization wave has longitudinal wavenumber k_x equal to the difference $k_{2x} - k_{1x}$ of longitudinal wavenumbers of the two optical fields. Therefore, only the mode having the above wavenumber is efficiently excited. The field intensity is maximized when the frequency of this mode is equal to the difference frequency of optical fields. This requires special waveguide design since refractive indices of bulk semiconductor materials for optical and IR frequencies are different. For far-IR generation, there is more flexibility due to efficient manipulation of the refractive index by a slight doping.

Basic model. To quantify the above ideas, we have calculated the excited IR polarization and field by solving the coupled electronic density-matrix equations and electromagnetic Maxwell equations for the three fields, assuming steady-state. It is convenient to expand all fields in an orthonormal set of cavity modes $\mathbf{F}_\lambda$ and to introduce slowly varying complex amplitudes of fields and polarizations. For example, for the IR field at the $3 \to 2$ transition we can write

$$\mathbf{E}(\mathbf{r},t) = \sum_\lambda \frac{1}{2}\mathcal{E}(t)\mathbf{F}_\lambda(\mathbf{r})\exp(-i\omega t) + \text{c.c.} \qquad (1)$$

We will assume that the mode has a simple $\exp(\pm i k_x x)$ dependence in the propagation direction x with the refractive index $\mu = k_x c/\omega$ and the transverse structure defined by a specific waveguide.

After introducing complex Rabi frequency $e(t) = d\mathcal{E}(t)/2\hbar$, the wave equation can be written as

$$\frac{de}{dt} + (\kappa + i(\omega_c - \omega))e = \frac{2\pi i\omega d^2 N}{\hbar\mu^2}\int_{V_c}\sum_j \sigma_{32}^j F_\lambda(\mathbf{r})\, d\mathbf{r}. \qquad (2)$$

Here d is the dipole moment of the IR transition, N is the total volume density of electron states in the active region, ω_{32} the central frequency of $3 \to 2$ transition, ω_c the frequency of the IR cavity mode with given k_x, κ the cavity losses, V_c the cavity volume. The variable σ_{32}^j is the slowly varying amplitude of the element ρ_{32}^j of density matrix, index j labels different electron states contributing to the inhomogeneously broadened line. For a system of QDs, j is simply the dot label. In QWs, index j labels different $\mathbf{k}_\parallel$ states with respect to longitudinal quasimomentum. Only $\mathbf{k}_\parallel$-conserving transitions are considered, where $\mathbf{k}_\parallel$ lies in the plane of the wells (xy-plane).

The same representation is assumed for the two optical fields $\mathbf{E}_{1,2}$, with off-diagonal density matrix elements σ_{21}^j, σ_{31}^j on the right-hand side, and corresponding parameters μ, d, ω, ω_c, κ, k_x, g having index 1 or 2. Note that the optical fields can have *arbitrary* polarization with respect to the parametrically excited IR field. In particular, for a QW laser the IR field should be z-polarized in the general case, while the optical fields are preferentially y-polarized.

Expressions for σ_{ik} and population differences $n_{ik} = \rho_{ii} - \rho_{kk}$, $i, k = 1, 2, 3$, are found from the density matrix equations with phenomenological rates of relaxation and pumping. For simplicity, we will assume bipolar injection with equal injection rates of electrons to level 3 and holes to level 1, so the total particle density is conserved. In the limit $|e| < \gamma_{ik}$ the resulting amplitude of the IR field is found to be

$$e \simeq \frac{ig^2 e_1^* e_2}{\kappa} \sum_j \left(\frac{n_{12}(\nu_j)}{\Gamma_{21}^* \tilde{\Gamma}_{32}} + \frac{n_{13}(\nu_j)}{\Gamma_{31} \tilde{\Gamma}_{32}} \right), \tag{3}$$

where $g^2 = 2\pi\omega d^2 NG/(\hbar\mu^2)$,

$$\begin{aligned}
\Gamma_{21} &= \gamma_{21} + i(\omega_{21} + \nu_j - \omega_1), \\
\Gamma_{31} &= \gamma_{31} + i(\omega_{31} + \nu_j - \omega_2), \\
\Gamma_{32} &= \gamma_{32} + i(\omega_{32} + \nu_j - \omega_2 + \omega_1), \\
\tilde{\Gamma}_{32} &= \Gamma_{32} + |e_1|^2/\Gamma_{31} + |e_2|^2/\Gamma_{21}^*.
\end{aligned}$$

Here G is the optical confinement factor for the IR field, ν_j is the difference between the transition frequency for a given jth state and the central frequency ω_{21}, ω_{31}, or ω_{32}.

The resonance $\omega_c = \omega_2 - \omega_1$ with a given cavity mode having the wavenumber $k_x \simeq k_{2x} - k_{1x}$ is assumed.

The value of the dipole moment at the IR transition is typically $d \sim (1-3)$ nm, while it is $(0.3-1)$ nm for the optical transitions [4,11]. The relaxation rates γ_{ik} of both optical and IR polarizations are of order 5-10 meV in QW lasers at room temperature (i.e., the relaxation time $\lesssim 0.1$ ps) [14–16] and can be several times lower in self- assembled QDs (1 ps). In the IR range the cavity losses are mainly due to free carrier absorption. At the high carrier densities $N \gtrsim 10^{18}$ cm^{-3} necessary for the excited-state lasing in QW's the material losses in a bulk active medium are of order 100 cm^{-1} at $\lambda \simeq 6$ μm, and grow as λ^2 or λ^3 depending on the dominant scattering mechanism [17]. Thick cladding layers have smaller doping density of order $4 \times 10^{16} - 10^{17}$ cm^{-3}, but can contribute significantly to the losses due to the large overlap factor $G \sim 1$. Large losses are a major problem in all proposed far-IR lasing schemes involving free carriers in semiconductors. Note, however, that increasing losses is not a principal limitation in the present parametric scheme since they do not prohibit generation itself, but rather decrease the IR field intensity. This possibility of far-infrared operation is an important advantage of the proposed mechanism. Note also that in QD lasers the excited state lasing can be achieved at lower carrier densities due to the state filling effect, and the intrinsic losses in active medium are lower.

Homogeneous broadening. This case is relevant for QWs at low temperatures. For room temperature, the quasi-Fermi energy for electrons is expected to be larger than the homogeneous bandwidth of 7-10 meV, and inhomogeneous broadening cannot be neglected. As for the self-assembled QDs, present-day structures have a large inhomogeneous broadening $\gtrsim 20$ meV associated with the spread of dot sizes, which is definitely much larger than the homogeneous linewidth.

The resulting expression for the IR field is

$$|e| \simeq \frac{|e_1||e_2|}{\gamma_{32}} \left(\frac{\omega}{\omega_1} \frac{d^2}{d_1^2} \frac{\kappa_1}{G_1} \frac{G}{\kappa} + \frac{\omega}{\omega_2} \frac{d^2}{d_2^2} \frac{\kappa_2}{G_2} \frac{G}{\kappa} \right), \tag{4}$$

where G, G_1 and G_2 are the IR and optical confinement factors. At a given wavelength, the crucial parameter in Eq. (4), which governs the efficiency of down-conversion, is $\eta = (\kappa_{1,2}/G_{1,2})(G/\kappa)$. The main source of IR losses is free-carrier absorption. For the optical fields the ratio $\kappa_{1,2}/G_{1,2}$ in which the material gain at the optical transition, which is of order 10^3 cm^{-1} in QWs and 10^4 cm^{-1} in QDs. Therefore, even for very high IR material losses of order $10^3 - 10^4$ cm^{-1}, the η parameter can still be close to unity. As we already mentioned, IR losses can be dominated by absorption in doped cladding layers. Let us take as an example $2\kappa \simeq 150$ cm^{-1} as measured in quantum cascade lasers at 17 μm wavelength. If $2\kappa_1/G_1 \simeq 1500$ cm^{-1}, then we have $\eta \sim 1$ for $G \sim 0.1$. Evidently, the value of η decreases rapidly with increasing wavelength due to growing κ and decreasing G.

Inhomogeneous broadening. We will assume here that the inhomogeneous widths u_{ik} of all transitions are much larger than all homogeneous bandwidths γ_{ik} and Rabi frequencies $|e_{1,2}|$ of optical laser fields that are at exact resonance with the centers of inhomogeneously broadened lines: $\omega_1 = \omega_{21}$ and $\omega_2 = \omega_{31}$. We consider two different situations in which explicit formulas can be obtained: when the optical field intensities are (i) much smaller and (ii) much greater than the saturation values.

(i). In this case the populations have the spectral distributions as supported by pumping in the absence of generated fields (no spectral hole burning). When $u_{ik} \gg \gamma_{ik}$, the precise shape of the inhomogeneous line is not important and simple expression for the IR field can be obtained:

$$|e| \simeq \frac{2|e_1||e_2|}{(\gamma_{32} + \gamma_{21})} \frac{u_{21}}{u_{32}} \frac{\omega}{\omega_1} \frac{d^2}{d_1^2} \frac{\kappa_1}{G_1} \frac{G}{\kappa}, \tag{5}$$

or, in terms of intensities,

$$|\mathcal{E}|^2 \simeq |\mathcal{E}_1|^2 \frac{|\mathcal{E}_2|^2}{|\mathcal{E}_2|_s^2} \left(\frac{2\gamma_{32}}{(\gamma_{32} + \gamma_{21})} \frac{d}{d_1} \frac{\omega}{\omega_1} \frac{u_{21}}{u_{32}} \frac{\kappa_1}{G_1} \frac{G}{\kappa} \right)^2. \tag{6}$$

Here $|\mathcal{E}_2|_s^2 = \hbar^2 \gamma_{32}^2 / d_2^2$. Equation (5) is similar to (4) except for the ratios of inhomogeneous linewidths that are expected to be of the order of unity. As we see, the ratio of the IR to optical intensity does not change much as compared with homogeneously broadened transitions, but, of course, the threshold conditions do change, so the value of injection current is now much larger.

(ii). In this case the population distributions have narrow spectral holes burned at the spectral positions of optical field modes. This can be relevant for QD lasers or powerful pulsed multiple QW lasers. To simplify calculations, let us assume that the absolute values of the Rabi frequencies of two optical fields are equal, $|e_1| = |e_2|$, all γ_{ik} and r_i are equal to the same value γ, and also $u_{21} = u_{31} = u$. After somewhat lengthy calculations, we arrive at a surprisingly simple result:

$$|e| \simeq \frac{|e_1||e_2|u}{\gamma u_{32}} \left| 0.9 \frac{\omega}{\omega_1} \frac{d^2}{d_1^2} \frac{\kappa_1}{G_1} \frac{G}{\kappa} - 0.1 \frac{\omega}{\omega_2} \frac{d^2}{d_2^2} \frac{\kappa_2}{G_2} \frac{G}{\kappa} \right| . \quad (7)$$

The requirement $|e| < \gamma$ employed in the derivation of Eq. (7) means that $|e| \ll |e_1|$ in the asymptotics (6), since the latter was obtained in the limit $|e_1| \gg \gamma$.

Expressions (4)-(7) predict the rapid growth of the IR intensity, proportional to the product of optical field intensities. Of course, this tendency holds only until the IR intensity reaches the saturation value. Above this value, the IR field begins to deplete the electron populations, and its growth becomes nonlinearly saturated. In the optimal case, the maximum efficiency of down-conversion approaches the limiting quantum value corresponding to one IR photon per one optical photon. For the mid-IR range $5-10$ μm the maximum IR power is $10-50$ mW if we take the value of 100 cm^{-1} for IR losses. Here we assumed the laser with parameter $\eta \sim 0.1$; see the discussion after Eq. (4). Beyond the restsrahlen region of strong phonon dispersion ($\lambda \gtrsim 50$ μm for AlGaAs/GaAs structure) the expected IR power is $\lesssim 1$ mW due to rapidly growing losses. We note, however, that progress in fabricating low-loss IR waveguides for semiconductor lasers is impressive. For example, in [18] waveguide losses as low as 14 cm^{-1} at 9.5 μm wavelength were reported.

Our calculations demonstrate the generation of coherent IR emission at intersubband transitions due to nonlinear wave mixing in standard multiple QW or QD laser diodes. The prerequisite for this is simultaneous lasing at two optical wavelengths which provide the necessary drive fields. This mechanism does not require population inversion at the IR transition, and its threshold current is determined by the minimum injection current necessary for the excited-state lasing.

It is important to note that the proposed parametric scheme seems to be viable in the far-IR region ($\lambda \sim 10-100$ μm) using transitions between hole subbands in the valence band. Indeed, in conventional lasers the increase of IR losses with wavelength is a significant problem for generation since it makes it difficult to reach the lasing threshold. In our case, due to the parametric nature of IR generation, the increase in losses will only lead to a decrease in output intensity; not a complete loss of IR generation as is the case in the usual below threshold laser behavior. For a practical device, the goal is to maximize the factor $(\kappa_{1,2}/G_{1,2})(G/\kappa)$. Also, in the far-IR spectral range it is relatively easy to manipulate the refractive index of the IR mode and provide phase matching by only a modest doping of waveguide layers.

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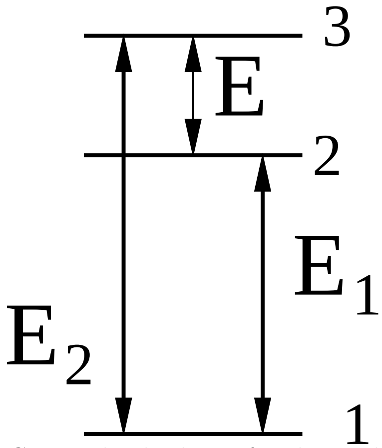

FIG. 1. Generic level scheme for three-color generation in a field biased quantum well: two strong lasing fields E_1 and E_2 excited at adjacent interband transitions $2 \rightarrow 1$ and $3 \rightarrow 1$ generate coherent IR radiation E at the beat frequency. Wavefunctions in such a skewed quantum well are indicated.