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Higher–energy resonant second harmonic generation in step–graded asymmetric semiconductor quantum wells

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In this paper we describe an optimization procedure applied to step–graded semiconductor structures in order to maximize intraband second–order susceptibility. It is based on simultaneous solving of nonlinear equations by varying chosen structure parameters and thus extracting the remaining parameters so that the condition of double resonance could be met. In the first case, all three states are bound. Then, we consider a resonant state in continuum as the third state while the other two are bound. This permits the use of a wider range of input photon energies. Numerical results, obtained for $Al_xGa_{1-x}N$ and $Al_xGa_{1-x}As$ quantum wells with pump photon energies of $\hbar\omega = 240$ meV are presented.

I Introduction

Double–resonant intraband second harmonic generation (SHG) has been studied in various asymmetric quantum wells (QW's) in the recent few years. In such structures it is possible to tune the potential shape and layer widths in order to achieve resonance conditions [1]. The second order nonlinear effects are maximal when consecutive energy states are equidistant, with the separation between them equal to the input photon energy. The aim is to find the optimal structure parameters which maximize the cyclic product of the three dipole matrix elements since it determines the second order susceptibility $\chi^{(2)}$. Evidently, this product is equal to zero for symmetric structures.

Most of the papers published so far describe resonant SHG for $\hbar\omega = 116$ meV [1] – [12] which corresponds to CO_2 laser input radiation. This refers to the case where all three states are bound and have to be placed below the barrier top (we have considered only the lowest three states). It is obvious that the conduction band offset between semiconductors limits the depth of the well and the highest photon energies that can be frequency–doubled. One way of overcoming this problem may be the use of semiconductor heterojunction with larger band offsets, such as aluminum nitride and gallium nitride [13] – [15]. The other possibility is to consider bound–continuum transitions. States above the barrier are double–degenerated and those close to resonance could be favourable as the third state.

The dependence of $\chi^{(2)}$ on the triple product of the matrix elements changes in comparison to the situation where all states are bound, but optimization procedure remains similar.

We have analysed $Al_xGa_{1-x}N$ and $Al_xGa_{1-x}As$ step–graded quantum wells in the attainable range of parameters. The input photon energy is set to $\hbar\omega = 240$ meV (corresponding to CO laser radiation). The method is based on solving a system of nonlinear equations which contains the dimensions of QW and state energies as parameters. The double–resonance condition is imposed by replacing energies in this system with E_0, $E_0 + 240$ meV and $E_0 + 480$ meV, where E_0 represents the energy of the ground state. The dipole matrix elements obtained by these calculations in case of three bound states are quite large for given material and input photon energy. On the other hand, introduction of bound–continuum resonance transitions gives us the possibility of accomplishing higher–energy SHG in QW's based on conventional $Al_xGa_{1-x}As$ alloy.

II Theoretical considerations

We consider an n–doped QW structure based on direct band gap semiconductors, and take the band gap throughout it to be large enough that interband transitions, caused by radiation present in the structure, may be neglected. The polarization response of the structure to the pump field with photon energy $\hbar\omega$ is then mainly governed by intraband transitions between quantised conduction band states E_i. Nonlinear polarization at twice the frequency of the pump field, acting as the source of second harmonic field is described by the second–order susceptibility $\chi^{(2)}$. Under the conditions stated above $\chi^{(2)}$ is significant only for both the pump and harmonic polarized perpendicular to the QW plane (z–axis), i.e. $\chi^{(2)} \equiv \chi^{(2)}_{zzz}$. It is given by the general expression (e.g. Ref. [1]):

$$\chi^{(2)}_{zzz} = \frac{e^3}{L_z\epsilon_0\hbar}\sum_i\sum_k \frac{1}{(2\hbar\omega - \Delta E_{ki}) - i\hbar,_{ki}} \times$$
$$\sum_l M_{ik}M_{kl}M_{li}\left[\frac{\rho_{ii}-\rho_{ll}}{\hbar\omega + \Delta E_{li} - i\hbar,_{li}} - \frac{\rho_{ll}-\rho_{kk}}{\hbar\omega - \Delta E_{kl} - i\hbar,_{kl}}\right] \qquad (1)$$

where $M_{ij} =< \Psi_i|z|\Psi_j >$ are the transition dipole matrix elements, ΔE_{ij} the transition energies between states i and j, ρ_{ii} denotes the electron sheet density in the state i, $,_{ij}$ the off-diagonal relaxation rates and L_z the length of the structure. In majority of feasible structures almost all electrons normally reside on the lowest state (i.e. $\rho_{ii} \ll \rho_{00}$ for $i > 0$), and QW is tailored so that the lowest three of its states (0,1 and 2) are spaced by approximately the pump photon energy. Eq. (1) then grossly simplifies to

$$\chi^{(2)}_{zzz} = \frac{e^3\rho_{00}}{L_z\epsilon_0\hbar}\frac{M_{01}M_{12}M_{20}}{(2\hbar\omega - \Delta E_{02} - i\hbar,_{02})(\hbar\omega - \Delta E_{01} - i\hbar,_{01})} \qquad (2)$$

The largest value occurs in exactly the double-resonance regime, $\hbar\omega = \Delta E_{01} = \Delta E_{12} \equiv \Delta E$, i.e. with strictly equispaced states, amounting to

$$\chi^{(2)}_{zzz} = \frac{e^3\rho_{00}}{L_z\epsilon_0}\frac{M_{01}M_{12}M_{20}}{(\hbar,)^2} \qquad (3)$$

where we take $,_{01} = ,_{02} = ,$ (though this is not essential).

In case of having free states contributing to the process, these expressions have to be slightly modified in order to include the density of free states. The second order susceptibility of such quantum system remains determined by Eq. (1). As we have said before, because of the denominators with energy differences, this expression will simplify under resonance conditions, i.e. when some of the states are spaced by about the "pump" photon energies, with just one term with these "properly spaced" states remaining as important (resonantly enhanced). Taking that only the ground state is significantly

populated with electrons (which is justified in realistic QW's), the second order susceptibility is then found to be:

$$\chi^{(2)}_{zzz} = \frac{e^3}{L_z\epsilon_0}\rho_{00}M_{01}\sum_k \frac{M_{0k}M_{1k}}{(\hbar,\ )^2 + [(2\Delta E) - (E_k - E_0)]^2} \tag{4}$$

where $(\hbar,\ )^2$ is the linewidth (dephasing rate), being typically 5 meV in QW systems (and is here taken to be common to all transitions). The summation is performed over all energies E_k belonging to the continuum. Wave functions corresponding to the states above the barrier are normalized by taking into consideration the box–boundary conditions. The previous expression should then be transformed in the following way:

$$\chi^{(2)}_{zzz} = \frac{e^3}{L_z\epsilon_0}\rho_{00}M_{01}\frac{1}{L_z}\sum_k \frac{\hat{M}_{0k}\hat{M}_{1k}}{(\hbar,\ )^2 + [(2\Delta E) - (E_k - E_0)]^2}\frac{\Delta k_B}{\Delta k_B} \tag{5}$$

where $\hat{M}_{ij}$ represent matrix elements calculated with normalised bound state and non–normalised contiuum state wave functions and $\Delta k_B = \pi/L_z$. When $L_z \to +\infty$, then Δk_B becomes dk_B and $\sum \to \int$. Specializing to the case we consider, that of two bound and one resonance state having about proper spacings, the second order susceptibility acquires the form:

$$\begin{aligned}\chi^{(2)}_{zzz} &= \frac{e^3}{L_z\epsilon_0}\rho_{00}M_{01}\sqrt{\frac{2m_{B0}}{\hbar^2}}\frac{1}{\pi} \\ &\times \int_{U_B}^{+\infty} \frac{\hat{M}_{02}\hat{M}_{12}}{(\hbar,\ )^2 + [(2\Delta E) - (E_2 - E_0)]^2}\left(\frac{1}{2\sqrt{E_2 - U_B}} + \frac{1}{\sqrt{E_{gB}}}\right)dE_2 = \frac{e^3}{L_z\epsilon_0}\rho_{00}\Pi^*\end{aligned} \tag{6}$$

where m_{B0} is the effective mass in the barrier at the conduction band edge. The matrix elements with state 2 (belonging to continuum) are to be calculated twice, because of the double degeneracy (i.e., with both wave functions corresponding to energy E_2). These two wave functions are taken in form of scattering states (i.e., are orthogonal), which prevents under - or overcompleteness in summing over all continuum states in (6). Clearly, with bound states wave functions localized in the well, one expects that the wave functions close to the resonances will give the largest contributions in (6), because of the largest matrix elements, provided the resonance conditions are also met. We may note that, due to different types of normalization of bound and free states wave functions, the triple product of matrix elements in (6) is not (even in terms of physical dimensions) equivalent to the corresponding product in case of all three bound states. To make the comparison between the two cases possible, the former should be multiplied by $(\hbar,\ )^2$ ($\hat{\Pi}^{(2)} = (\hbar,\ )^2\Pi^*$).

In order to maximize $\chi^{(2)}$ one should clearly maximize the corresponding products of dipole matrix elements $\Pi^{(2)} = M_{01}M_{12}M_{20}$ in numerator of (3), by appropriate tailoring of QW profile (and hence the wave functions) while preserving the levels spacing. In case of $\chi^{(2)}$, the presence of M_{20} rules out symmetric QW's, because of the definite parity of wave functions, so one should consider asymmetric structures only. To find the best potential shape, one should vary the potential $U(z)$, and (related to it in ternary alloys) effective mass $m(z)$, subjected to the constraint that states spacing should be as desired, i.e. only the wave functions and hence the matrix elements should be affected.

Quantised electron states in QW structure with position-dependent effective mass $m(z)$ may be found by solving the envelope function Schrödinger equation of the form [16]

$$-\frac{\hbar^2}{2}\frac{d}{dz}\left(\frac{1}{m(z)}\frac{d\Psi}{dz}\right) + U(z)\Psi = E\Psi \tag{7}$$

where $\Psi(z)$ is the envelope wave function, $U(z)$ the potential and E the energy. Effects of bulk dispersion nonparabolicity may be conveniently described by energy-dependent effective mass, according to the two-band Kane model [17]

$$m(z,E) = m^*(z)\left[1 + \frac{E - U(z)}{E_g(z)}\right] \tag{8}$$

where $E_g(z)$ is the material composition- (and hence the position-) dependent band gap, and $m^*(z)$ denotes the parabolic (band edge) effective mass. These effects become increasingly important at higher energies. They are technically significant in QW's designed for SHG of CO_2 laser radiation, and even more so at higher energies.

Consider an asymmetric step QW with stepwise constant potential and effective mass (Fig. 1(a)), which is frequently used in resonant SHG. Its optimization has been considered in Ref. [1]. The

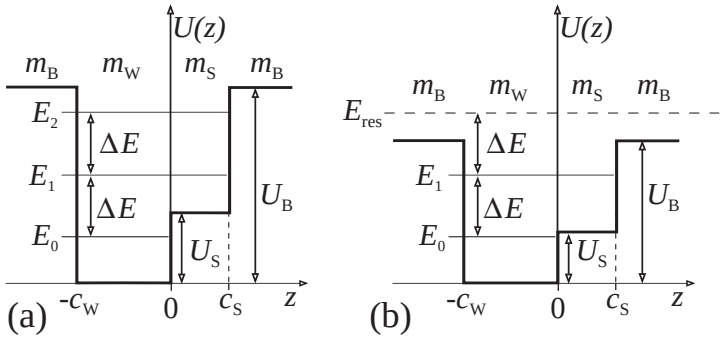

FIG. 1. The potential (conduction band edge) in single–step QW in case of (a) three bound states (b) two bound and one resonant state. The structure design parameters, used in the main text, are all denoted.

authors first assumed an idealized model with infinite barriers and constant effective mass, with nonparabolicity neglected, allowing for analytic solution. Having optimized the parameters of such a QW, it was then modified to take a finite barrier height into account. A single composition of the barrier material was assumed, regardless of the pump photon energy $\hbar\omega$ that QW was designed for, i.e. the barrier height was not considered as a parameter for optimization. Yet, data presented in Ref. [1] for infinite and finite barrier QW's indicate that the matrix elements product $\Pi^{(2)}$ in the latter exceeds the value in the former by a factor which depends on $\hbar\omega$ but is always very significant, e.g. $\Pi^{(2)}$ may more then double due to finite barriers. The effect is ascribed to the fact that finite barriers allow for wave function penetration inside them, and more extended wave functions lead to larger matrix elements. With enhancement of $\Pi^{(2)}$ as large as that (i.e. such difference between optimized idealized QW, and modified but nonoptimized realistic QW) one may wonder whether the barrier height should be considered as free parameter, on equal grounds with others, and perform the full optimization of this system. This is the problem we consider below, taking also the nonparabolicity (neglected in Ref. [1]) into account.

Eqs. (7-8) should be solved, with observing the boundary conditions (continuity of $\Psi(z)$ and $(1/m(z))d\Psi/dz$) at $z = -c_W$, $z = 0$ and $z = c_S$. With the conventional exponential or plane wave type of solutions in separate layers of the structure (Fig. 1(a)) we get a system of six homogeneous equations. Nontrivial solution of that system requires that

$$\Phi(E) = \sin(k_W c_W)\left[\sinh(k_S c_S)\frac{k_B}{m_B}\left(\frac{k_W^2}{m_W^2} - \frac{k_S^2}{m_S^2}\right) + \cosh(k_S c_S)\frac{k_S}{m_S}\left(\frac{k_W^2}{m_W^2} - \frac{k_B^2}{m_B^2}\right)\right]$$
$$- \cos(k_W c_W)\frac{k_W}{m_W}\left[\sinh(k_S c_S)\left(\frac{k_B^2}{m_B^2} + \frac{k_S^2}{m_S^2}\right) + 2\cosh(k_S c_S)\frac{k_B k_S}{m_B m_S}\right] = 0 \qquad (9)$$

in the energy range $(0 < E < U_S)$, where m_B, m_W and m_S are the energy-dependent nonparabolic effective masses in the barrier, well and the step layers, respectively, and the corresponding wave vectors are $k_B = [2m_B(U_B - E)/\hbar^2]^{1/2}$, $k_W = [2m_W E/\hbar^2]^{1/2}$, and $k_S = [2m_S(U_S - E)/\hbar^2]^{1/2}$. In the energy range above the step $U_S < E < U_B$ we define $k_S^* = ik_S = [2m_S(E - U_S)/\hbar^2]^{1/2}$ and Eq. (9) is modified by substituting $\sinh(k_S c_S) \to \sin(k_S^* c_S)$, $\cosh(k_S c_S) \to \cos(k_S^* c_S)$, $k_S \to k_S^*$, $k_S^2 \to -k_S^{*2}$. This defines the function $\Phi(E)$,its zeros being the energies of quantised states in asymmetric single

step QW, with nonparabolicity included. The corresponding wave functions are then simply derived from the boundary conditions and the normalization condition $\int_{-\infty}^{+\infty}|\Psi(z)|^2dz = 1$.

Having chosen the alloy system to work with it is reasonable to take the well layer to comprise pure "well-type" semiconductor, because, with $\Pi^{(2)}$ roughly scaling as effective mass to power 3/2 (Ref. [1]) there is no benefit from allowing the well layer to be made of the alloy. Thus, m_W is defined from the start, and in the step and barrier layers, which are made of the alloy, with suitable compositions x_S and x_B, the effective mass and potential are uniquely related to each other, i.e. $m_{S,B} = m_{S,B}(x_{S,B})$ and $U_{S,B} = U_{S,B}(x_{S,B})$. Therefore, $\Phi(E)$ is a nonlinear function of four independent parameters, say the widths c_W and c_S, and potentials U_S and U_B. All possible QW shapes, i.e. the values of the four parameters, which result in three states spaced by a specified amount $\Delta E = \hbar\omega$, may be obtained from the system of three nonlinear equations

$$\begin{aligned}\Phi(U_B, U_S, c_W, c_S, E_0) &= 0\\ \Phi(U_B, U_S, c_W, c_S, E_0 + \Delta E) &= 0\\ \Phi(U_B, U_S, c_W, c_S, E_0 + 2\Delta E) &= 0\end{aligned} \tag{10}$$

where E_0, the ground state energy measured from the well bottom, is an additional free parameter. Eq. (10) may then be solved for three parameters out of five, the remaining two being "input" parameters to be used for the QW shape variation, with values of all the five parameters subject either to obvious physical constraints or to limitations imposed by the chosen alloy system (also the technological feasibility of the structure may impose some additional constraints). By evaluation of the wave function and the matrix elements (all that can be done analytically, though via rather cumbersome expressions) for each individual solution, it is quite straightforward to search the entire two-dimensional free-parameters space and find the best of all step QW's, which maximizes $\Pi^{(2)}$.

An analogous procedure may also be used to explore the possibility of designing QW's with two bound states and a resonant state, belonging to the continuum,for the second harmonic generation. The bound-continuum transitions have previously been considered in other contexts, e.g. for infrared absorption and photodetectors [18, 19], but not for the harmonic generation.

We consider the class of QW's with the structure given in Fig.1(b). The state energies and the corresponding wave functions can be found from the effective-mass Schrödinger equation, which is valid for the conduction band electrons, and may take account of nonparabolicity via the energy-dependent effective mass Eq. (8), according to the two-band Kane model [17]. States above the barrier are double degenerate, and most interesting are those close to resonances. Resonances correspond to local maxima of transmission, which generally do not exactly equal unity in this asymmetric system (only at quite high energies will the transmission become close to unity). Positions of transmission coefficient extrema coincide with extrema of the following function:

$$\begin{aligned}F(E) &= A\sin^2(k_W c_W)\sin^2(k_S^* c_S) + B\sin^2(k_W c_W)\cos^2(k_S^* c_S)\\ &+ C\cos^2(k_W c_W)\sin^2(k_S^* c_S) + \frac{1}{2}D\sin(2k_W c_W)\sin(2k_S^* c_S)\\ &+ 2\sin^2(k_W c_W)\sin^2(k_S^* c_S) - 2\sin^2(k_W c_W) - 2\sin^2(k_S^* c_S)\end{aligned} \tag{11}$$

where $A = (\frac{m_W k_S^*}{m_S k_W})^2 + (\frac{m_S k_W}{m_W k_S^*})^2$, $B = (\frac{m_W k_B^*}{m_B k_W})^2 + (\frac{m_B k_W}{m_W k_B^*})^2$, $C = (\frac{m_S k_B^*}{m_B k_S^*})^2 + (\frac{m_B k_S^*}{m_S k_B^*})^2$, $D = (\frac{m_B^2 k_W k_S^*}{m_W m_S k_B^{*2}} + \frac{m_W m_S k_B^{*2}}{m_B^2 k_W k_S^*} - \frac{m_W k_S^*}{m_S k_W} - \frac{m_S k_W}{m_W k_S^*})$, $k_B^* = [2m_B(E - U_B)/\hbar^2]^{1/2}$. The wave function amplitudes inside the well are larger at resonance energies then off them, but there is no big difference outside the well, due to the normalization condition.

Within the class of single step asymmetric QW's, all possible shapes that provide resonance conditions are accessed by solving the system of three nonlinear equations which demand that the two bound and the resonance state are spaced by exactly $\Delta E = \hbar\omega$:

$$\begin{aligned}\Phi(U_B, U_S, c_W, c_S, E_0) &= 0\\ \Phi(U_B, U_S, c_W, c_S, E_0 + \Delta E) &= 0\\ E_{\rm res}(U_B, U_S, c_W, c_S) - (E_0 + 2\Delta E) &= 0\end{aligned} \tag{12}$$

where $E_{\rm res}$ is the first continuum resonance energy, corresponding with the first minimum of the function $F(U_B, U_S, c_W, c_S, E)$ (Eq. (11)).

These equations contain the QW structure parameters, and also the ground state energy E_0 , with its value alone being irrelevant for the process we consider, so it is also taken as a parameter on equal footing with layer widths (c_W,c_S) and potentials (U_S,U_B). With a total of five parameters and three equations (12), two of them are really free "QW design" input parameters, while the remaining three can be determined by solving (12). Therefore, not only that a QW may be designed for a chosen $\hbar\omega$ (within some limits), but there is even room for the QW shape optimization in order to give maximal nonlinearity. The optimal QW shape may be found by defining a two dimensional parameter space, to be searched by first solving (12) for the remaining parameters, and then (provided that the solution is physically and technologically acceptable) calculating the matrix elements and the nonlinear susceptibility corresponding to a particular solution.

Similarly, the described procedure may be employed for optimization of (also frequently encountered) coupled QW's in respect to $\chi^{(2)}$. QW's intended for other nonlinear processes which may not require equispaced states (off-resonant harmonic generation, parametric down-conversion etc.) can be optimized in the same fashion, as well.

The described procedure is systematic in the sense that it allows the entire search of the free-parameters space defining the QW profile within a given class (i.e. of the same general shape), and does not include intuition or elements of luck in spotting the "best potential shape". On practical side, it may be implemented with reasonable effort and computation time only for structure comprising not more than a few layers of different widths and compositions. Yet, exactly such simple structures are of the largest technical importance at present.

III Numerical results

To illustrate this procedure we have performed the optimization of ternary $A_xB_{1-x}C$ alloys based QW's in respect to resonant intersubband nonlinearity. The step-type QW's are based on $Al_xGa_{1-x}N$ (which may be useful for higher energy SHG) in case of three bound states as displayed in Fig. 1(a), and on the conventional $Al_xGa_{1-x}As$ alloy (with a third state in continuum), Fig. 1(b). In a single step QW made of alloys of AC and BC compounds, i.e. with the structure $A_{x_1}B_{1-x_1}C/BC/A_{x_2}B_{1-x_2}C/A_{x_1}B_{1-x_1}C$, the effective mass in separate layers is given by

$$\begin{aligned} m_B &= [m_{AC}x_1 + m_{BC}(1-x_1)]\left[1 + \frac{E-U_B}{E_{g\,B}}\right] \\ m_S &= [m_{AC}x_2 + m_{BC}(1-x_2)]\left[1 + \frac{E-U_S}{E_{g\,S}}\right] \\ m_W &= m_{AC}\left[1 + \frac{E}{E_{g\,W}}\right] \end{aligned} \qquad (13)$$

as follows from Vegard's law and the way chosen to introduce nonparabolicity, with band gaps in the step and barrier layers $E_{g\,B} = E_{g\,AC}x_1 + E_{g\,BC}(1-x_1)$ and $E_{g\,S} = E_{g\,AC}x_2 + E_{g\,BC}(1-x_2)$. The step and barrier heights are $U_{S,B} = x_{2,1}\Delta E_c$, where ΔE_c is the conduction band offset between AC and BC compounds.

We first consider the wurtzite semiconductor $Al_xGa_{1-x}N$ QW's, both compounds having direct band gaps, and use the following parameters [13] – [15],[20]: $m_{GaN} = 0.18m_0$, $m_{AlN} = 0.27m_0$, $E_{g\,GaN} = 3.45$ eV, $E_{g\,AlN} = 6.28$ eV. There is a dispersion of data on ΔE_c between GaN and AlN in literature. We have used the value $\Delta E_c = 2$ eV, based on recent photoemission spectroscopy measurements [21, 22] and calculations [23] – [25] which all suggest the valence band offset about 0.8 eV.

Choosing initially $\hbar\omega = 240$ meV (this corresponds to 5.1μm CO laser or approximately to frequency doubled CO_2 used as pump for the next SHG), we performed the step QW optimization, via

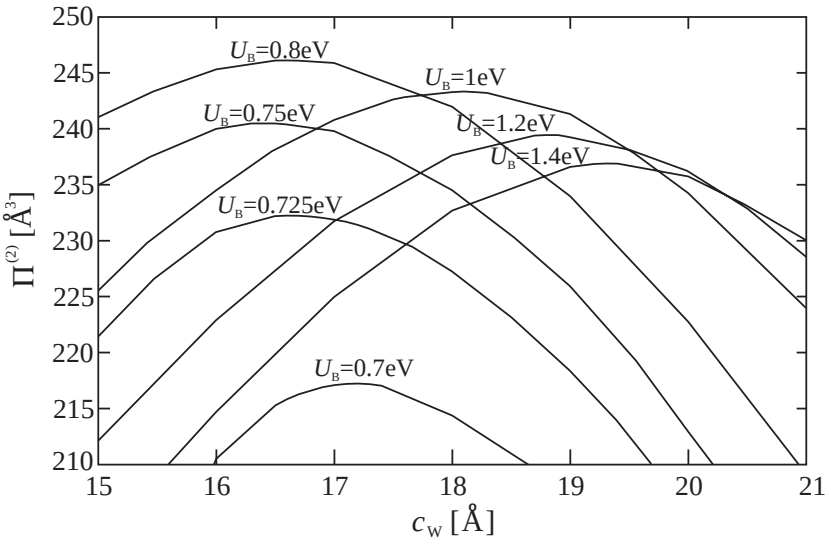

FIG. 2. The product of matrix elements $\Pi^{(2)} = M_{01}M_{12}M_{20}$ *in* $Al_xGa_{1-x}N$ *step QW from Fig. 1(a) as it depends on the choice of the lower well width* c_W *for various values of barrier height* U_B*, calculated under double resonance condition* $\hbar\omega = 240$ *meV.*

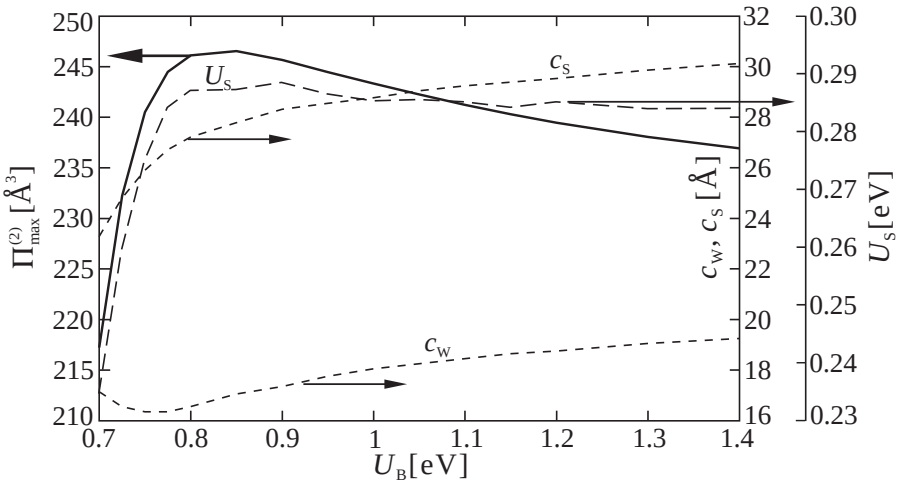

FIG. 3. The maximal values of $\Pi^{(2)}$ *in* $Al_xGa_{1-x}N$ *single step QW at double resonance,* $\hbar\omega = \Delta E = 240$ *meV, achievable with various values of barrier height* U_B*, together with other corresponding structural parameters.*

solving Eq. (10), by taking the width of the well c_W and barrier height U_B as free parameters. These were varied in the range 15 Å$\leq c_W \leq$ 30 Å and 0.7 eV$\leq c_W \leq$ 1.4 eV, respectively. Other parameters of the structure (c_S, U_S, E_0) were coming out as the solution of Eq. (10). By calculating the product of matrix elements $\Pi^{(2)}$, relevant for SHG, we find that the best results are obtained if the first level happens to be below the step ($E_0 < U_S$) and the other two above it, as is indeed generally accepted in the literature. Some results for this case are given in Fig. 2, which clearly shows the importance of proper choice of the barrier height, along with other parameters, in designing the optimized QW. Maximum values of $\Pi^{(2)}$ achievable with specified values of U_B (dictated by technology-related constraints, for instance), together with values of optimally designed QW parameters, may be read from Fig. 3. The largest value of $\Pi^{(2)}$ achievable in this structure amounts to 247 Å^3 with the QW parameters U_B = 850 meV, U_S = 287 meV, c_W = 17 Å, c_S = 28 Å (energies of the three states are E_0 = 195 meV, E_1 = 435 meV, and E_3 = 675 meV).

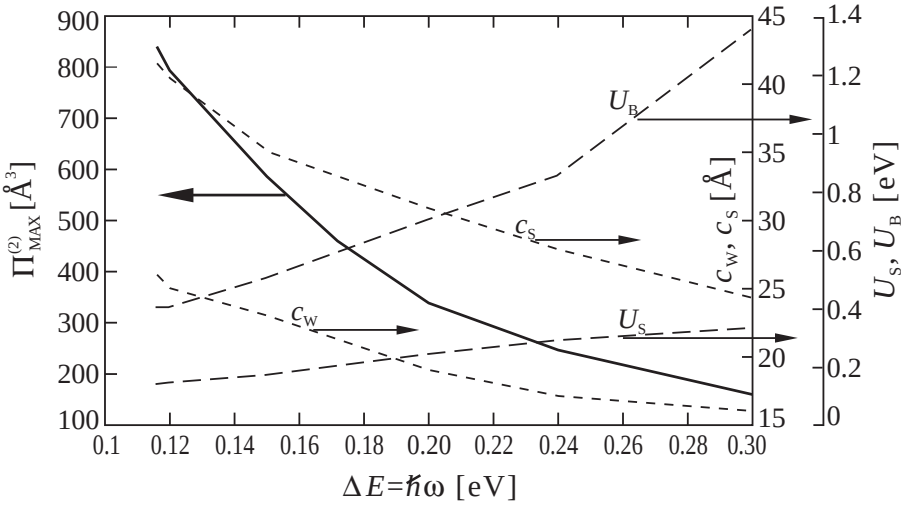

FIG. 4. Maximum achievable $\Pi^{(2)}$ *in* $Al_xGa_{1-x}N$ *single step QW (Fig. 1(a)), together with the structural parameters, given for a range of pump photon energies under double resonance conditions.*

The procedure was then repeated for various values of pump photon energy in the range $\hbar\omega$ = 100 – 300 meV. The fully optimized value $\Pi^{(2)}_{MAX}$ and the corresponding QW parameters, as they depend on $\hbar\omega$, are presented on Fig. 4. One may note that $\Pi^{(2)}_{MAX}$ decreases with $\hbar\omega$, which is in qualitative agreement with the fact that simple idealized structures, like constant mass linear harmonic oscillator, have dipole matrix elements scaling as $\Delta E^{-1/2}$, hence one normally expects that $\Pi^{(2)}_{MAX} \sim \Delta E^{-3/2}$ [11].

The theory described in the previous section was also employed for the design and optimization of QW's based on the $Al_xGa_{1-x}As$ alloy , to be used for resonant second harmonic generation of $\hbar\omega$ = 240 meV radiation (corresponding to, say, CO laser). The material parameters are taken as [16]: $m_{GaAs} = 0.067m_0$, $m_{AlAs} = 0.15m_0$, $E_{g\,GaAs}$ = 1.42 eV, $E_{g\,AlAs}$ = 2.67 eV (direct gap), and ΔE_c = 750 meV. Due to comparatively large photon energies involved, this technologically favourable alloy does not provide sufficient band offset to enable QW's with three bound states spaced by this

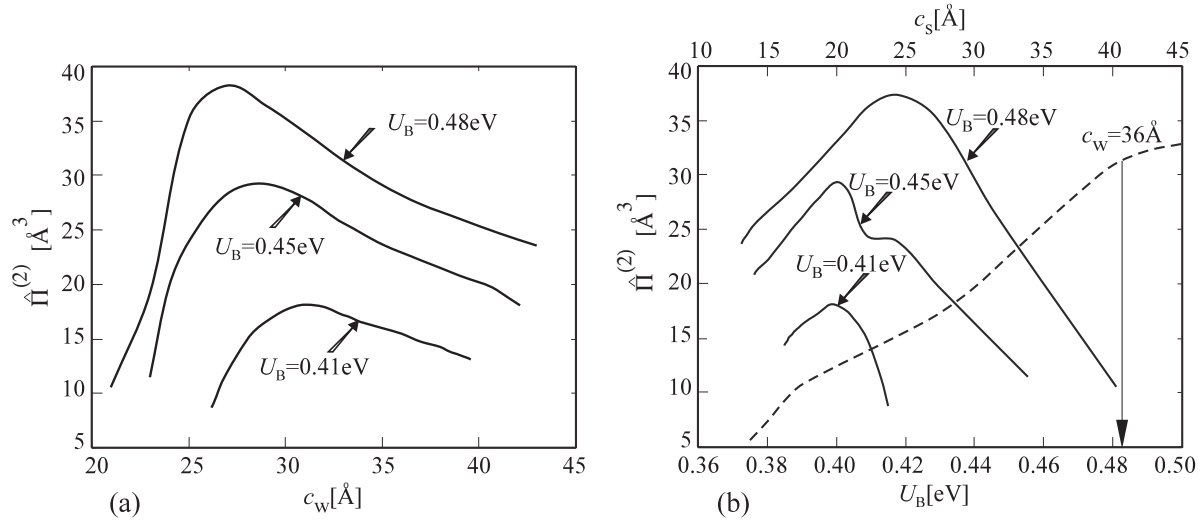

FIG. 5. The product of dipole matrix elements as it depends on the QW design parameters:(a) on the well width c_W *and (b) on the barrier height* U_B *(dashed line) and the step layer width* c_S *(solid lines).*

amount, so one cannot take advantage of double resonance enhancement of nonlinear susceptibility in this case. The difficulty may be circumvented, at least in principle, by allowing the highest of these states to be a resonance.

The well layer width c_W and barrier height U_B were taken as the input parameters, and the others came out as the solution of Eq. (12) . The cyclic product of matrix elements, as it depends on the choice of the QW parameters, is given in the Fig. 5(a). To obtain as large nonlinearity as possible, it is clearly advantageous to choose the highest technologically reasonable barrier height, and, upon fixing this value, there is an optimal well width. In Fig. 5(b) the dependence of the matrix elements product on U_B and the step layer width c_S is given. The largest value $\Pi^{(2)}_{MAX} = 38$ Å^3 is obtained in QW with $U_B = 480$ meV, $U_S = 190$ meV, $c_W = 27$ Å, $c_S = 24.5$ Å .

IV Conclusion

A systematic method for the optimization of ternary semiconductor alloys based QW's in respect to nonlinear optical susceptibilities was discussed.

It is applicable to step–graded QW's, like asymmetric step QW, coupled QW and similar. It allows the variation of a potential of specified general shape throughout its free-parameters space, in such a way to keep quantised states energies as required, while looking for maximal nonlinearity. We have demonstrated it on the design of $Al_xGa_{1-x}N$ and $Al_xGa_{1-x}As$ based QW's intended for resonant SHG in case of all three bound states, and two bound plus one resonant state. The calculations for AlGaN QW's have provided rather large dipole matrix elements products.

Even though the values of the dipole matrix elements products in semiconductor quantum wells with two bound states and the resonant state in the continuum as the third are not very large, it is important to know that it is possible to obtain resonantly enhanced nonlinear optical susceptibility, corresponding to higher values of pump photon energies, in conventional technologically favourable $Al_xGa_{1-x}As$ based QW's.

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