Optimization of nonlinear optical properties in AlGaAs quantum wells: Inverse spectral theory

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A procedure for the design of quantum well structures optimized with respect to the intersubband resonant second order nonlinear effects is proposed. It relies on the inverse spectral theory, allowing one to start from an arbitrary potential and shift its levels to the positions required for a particular application, in this case such that they become equispaced. Free parameters that appear, and determine the shape of the modified potential, are then varied in order to find the optimal potential shape, that maximizes the nonlinearity, while levels energies are automatically fixed throughout this variation. Furthermore, the procedure is adapted to handle cases of variable effective mass, unlike the conventional inverse spectral theory. The use of this procedure is demonstrated by designing a graded AlGaAs ternary alloy quantum well optimized for the second order nonlinearity at 10.6 μm. Starting with a truncated parabolic potential, the final optimized quantum well potential is obtained, with nonlinearity exceeding values previously obtained in the literature.

I   Introduction

Intersubband transitions in semiconductor quantum wells (QW) have attracted considerable research attention. This is mainly due to large values of dipole transition matrix elements [1] and the possibility of achieving the resonance conditions. Thus, both the linear and, even more so, nonlinear optical processes in these structures are very intense. Large dipole matrix elements are associated with a small effective mass m^* of electrons, scaling approximately as $m^{*\,-1/2}$ (Ref. [2]). Within a given material (i.e., m^*), however, a lot can be done to enhance those matrix elements relevant to a particular type of nonlinearity, by proper shaping of the QW (i.e., its potential), and hence the quantized states' wave functions. While varying the QW shape it is (almost always) essential to keep the level spacing as specified, e.g. equispaced for double resonant second harmonic generation (SHG). Clearly, only asymmetric structures are useful for SHG, and may be realized by either an asymmetric composition grading in a stepwise constant or continuous manner, e.g. Refs. [2] and [3], or electric field biasing, e.g. Refs. [4, 5], or both.

Some considerations of optimizing the QW shape, within the class of simple step-graded QW's, and within a somewhat idealized model, were presented in Ref. [2]. If the search for the best potential shape requires any amount of trial-and-error type calculations, most effort is spent in restoring the level spacing upon changing the potential shape, rather than in checking the values of matrix elements. Within the class of continuously graded QW's this problem may be very serious. For these we have recently used a method based on supersymmetric quantum mechanics [6, 7, 8]. It starts with a (rather arbitrary) initial potential, such that its quantized states are positioned as required, e.g. are equispaced if the QW is intended for resonance SHG, and then generates a family of potentials isospectral to the initial one, their shape being controlled by one or more scalar parameters. By varying these parameters one may easily search for the potential shape which maximizes those matrix elements relevant for e.g. resonance SHG. Here we describe another method for finding the best potential shape of QW's, which is even more versatile than the one based on supersymmetry. It starts with a completely arbitrary initial potential, and, using the inverse spectral theory (IST), e.g. Ref. [9], shifts the states to their desired positions, while, at the same time, it introduces free parameter(s) for varying the shape of this modified potential in an isospectral manner. The use of the method is exemplified by finding the QW shape for optimal second order susceptibilitys, relevant for SHG and optical rectification (OR).

II   The IST tailoring of the potential

The electron motion in a potential well $U(z)$, with a constant effective mass m^* is described by the envelope function Schrödinger equation

$$-\frac{\hbar^2}{2m^*}\frac{d^2\psi_i(z)}{dz^2} + U(z)\psi_i(z) = E_i\psi_i(z).$$ (1)

The discrete eigenenergies of the system, and the corresponding eigenfunctions, are denoted as E_i and $\psi_i(z)$ $(i = 1, 2, \ldots)$, respectively. The IST enables one to construct a modified potential $U_{IST}(z)$, which has the property that one (say, the k'th) of its eigenvalues is shifted by a prescribed amount ϵ from the k-th eigenvalue (E_k) of the initial potential $U(z)$, while all other eigenvalues of $U(z)$ and $U_{IST}(z)$ coincide. It has the form

$$U_{IST}(z; E_k + \epsilon) = U(z) - \frac{\hbar^2}{m^*}\frac{d^2}{dz^2}\ln[\mathrm{W}_k\{\psi_\epsilon, \psi_k\}]$$ (2)

where $\mathrm{W}_k(z)$ is the Wronskian

$$\mathrm{W}_k\{\psi_\epsilon, \psi_k\} = \psi_\epsilon(z)\frac{d\psi_k(z)}{dz} - \psi_k(z)\frac{d\psi_\epsilon(z)}{dz}$$ (3)

with ψ_k being the k-th eigenfunction of the initial potential, and ψ_ϵ any solution of the Schrödinger equation with $U(z)$ corresponding to energy $E_k + \epsilon$. The shift ϵ may take any value in the interval $(E_{k-1} - E_k, E_{k+1} - E_k)$, i.e., the shifted level cannot cross any other level [10].

The function ψ_ϵ, which is clearly not a normalizable eigenfunction, may be written as a linear combination of the two particular solutions, i.e., $\psi_\epsilon = C_1\varphi_1(z) + C_2\varphi_2(z)$, satisfying the fundamental initial conditions $\varphi_1(0) = \varphi_2'(0) = 1$, and $\varphi_2(0) = \varphi_1'(0) = 0$.

Consider a system having non-equispaced states, hence not appropriate for resonant SHG. Its energy spectrum may be "corrected" to provide equal (and just right) spacing between, say, the ground (1) and some other two states (2 and 3) by making shifts (one at a time): $E_2 \rightarrow E_2^{shift} = E_1 + \Delta E$, and $E_3 \rightarrow E_3^{shift} = E_1 + 2\Delta E$, where $\Delta E = \hbar\omega$ is the photon energy of the input laser radiation. The values of shifts of the initial potential states in the above equations are thus $\epsilon_2 = E_2^{shift} - E_2$ and $\epsilon_3 = E_3^{shift} - E_3$.

If the initial potential is symmetric, and lacks the second order nonlinearity, one also has to build in the asymmetrization in this procedure. For this purpose, the transform described above is done in
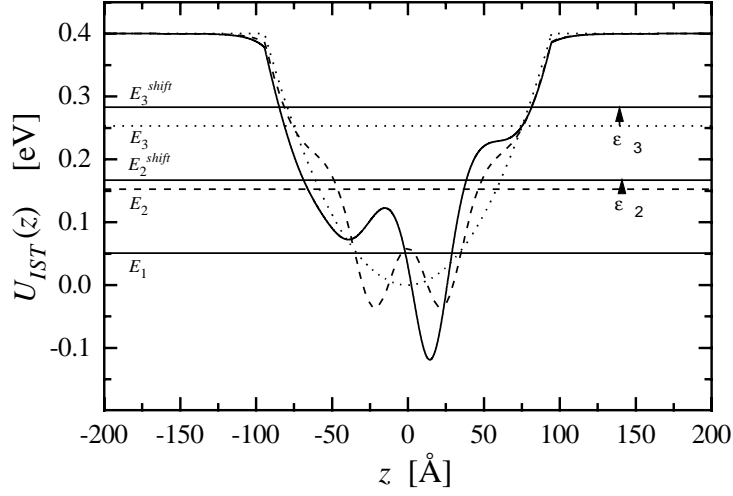
FIG. 1. The procedure of levels shifting and the potential asymmetrization, taking levels 2 and 3 to shift in respect to the fixed level 1.

two steps. First, the wave function ψ_ϵ corresponding to energy $E_3 + \epsilon_3 = E_1 + 2\Delta E$ is chosen, via the constants $C_1 = 0$ and $C_2 = 1$, to have the opposite parity from that of ψ_3, i.e., to be odd, which delivers a new potential

$$\tilde{U}(z; E_3 + \epsilon_3) = U(z) - \frac{\hbar^2}{m^*}\frac{d^2}{dz^2}\ln[\mathrm{W}_3\{\psi_{\epsilon_3}, \psi_3\}] \tag{4}$$

which, however, is still symmetric, as displayed in Fig. 1, but has the third state correctly spaced in respect to the ground state, $E_3^{shift} - E_1 = 2\Delta E$. The normalized wave functions for $i \neq 3$, corresponding to it, are given by

$$\tilde{\psi}_i(z) = \left(1 - \frac{\epsilon_3}{E_i - E_3}\right)^{-1/2}\left[\psi_i(z) - \frac{2m^*\epsilon_3}{\hbar^2}\frac{\psi_{\epsilon_3}(z)}{\mathrm{W}_3\{\psi_{\epsilon_3}, \psi_3\}}\cdot\int_{-\infty}^{z}\psi_i\psi_3 dz'\right] \tag{5}$$

while the wave function for $i = 3$

$$\tilde{\psi}_3(z) = \left(-\frac{\hbar^2}{2m^*\epsilon_3}[\mathrm{F}_{\epsilon_3}^+ - \mathrm{F}_{\epsilon_3}^-]\right)^{-1/2}\frac{\psi_3(z)}{\mathrm{W}_3\{\psi_{\epsilon_3}, \psi_3\}} \tag{6}$$

where $\mathrm{F}_{\epsilon_3}^\pm = \lim_{z\to\pm\infty}\mathrm{F}_{\epsilon_3}(z)$ and $\mathrm{F}'_{\epsilon_3}(z) = 1/\psi_{\epsilon_3}^2(z)$.

Next comes the asymmetrization. In calculating ψ_{ϵ_2} at energy $E_2 + \epsilon_2 = E_1 + \Delta E$, the constants $C_{1,2}$ are chosen so that, along with the particular solution having the opposite parity from ψ_2, the other one having the same parity as ψ_2 (i.e., odd) is also introduced. Indeed, setting $C_1 = 1$ and $C_2 = \alpha$ gives the final potential

$$U_{IST}(z; \alpha, E_3 + \epsilon_3, E_2 + \epsilon_2) = \tilde{U}(z) - \frac{\hbar^2}{m^*}\frac{d^2}{dz^2}\ln[\tilde{\mathrm{W}}_2\{\psi_{\epsilon_2}(\alpha), \tilde{\psi}_2\}] \tag{7}$$

or, after substituting (4) into (7)

$$U_{IST}(z; \alpha) = U(z) - \frac{\hbar^2}{m^*}\frac{d^2}{dz^2}\ln[\mathrm{W}_3\{\psi_{\epsilon_3}, \psi_3\}\cdot\tilde{\mathrm{W}}_2\{\psi_{\epsilon_2}(\alpha), \tilde{\psi}_2\}], \tag{8}$$

which is asymmetric for all $\alpha \neq 0$. The corresponding normalized wave functions for $i \neq 2$ read:

$$\psi_i^{IST}(z; \alpha) = \left(1 - \frac{\epsilon_2}{E_i - E_2}\right)^{-1/2}\left[\tilde{\psi}_i(z) - \frac{2m^*\epsilon_2}{\hbar^2}\frac{\psi_{\epsilon_2}(z; \alpha)}{\tilde{\mathrm{W}}_2\{\psi_{\epsilon_2}(\alpha), \tilde{\psi}_2\}}\int_{-\infty}^{z}\tilde{\psi}_i\tilde{\psi}_2 dz'\right] \tag{9}$$

where now $E_3 \equiv E_3^{shift}$, while the wave function for $i = 2$

$$\psi_2^{IST}(z; \alpha) = \left(-\frac{\hbar^2}{2m^*\epsilon_2}[\tilde{F}_{\epsilon_2}^+ - \tilde{F}_{\epsilon_2}^-] \right)^{-1/2} \frac{\tilde{\psi}_2(z)}{\tilde{W}_2\{\psi_{\epsilon_2}(\alpha), \tilde{\psi}_2\}} \tag{10}$$

where $\tilde{F}_{\epsilon_2}^\pm = \lim_{z \to \pm\infty} \tilde{F}_{\epsilon_2}(z)$ and $\tilde{F}_{\epsilon_2}'(z) = 1/\tilde{\psi}_{\epsilon_2}^2(z)$. Eqs. (8)-(10) are the "design" equations for the final asymmetric potential and the wave functions which enable a finite value of the cyclic product of matrix elements relevant for the second order nonlinearity. The free parameter α appearing in (8)-(10) implies the existence of a family of isospectral potentials. By varying α, one also varies the potential shape and hence the wave functions and the dipole matrix elements, with the final aim of finding the optimal $\alpha = \alpha_{opt}$, i.e., the optimized potential $U_{IST}(z; \alpha_{opt})$ which gives the largest product of relevant matrix elements (and the resonant second order nonlinearity). Throughout this variation of α in (8), the energies of its states, and hence the resonance conditions, remain preserved, i.e., $E_2^{shift} - E_1 = \Delta E$ and $E_3^{shift} - E_1 = 2\Delta E$.

III Supersymmetric quantum mechanics as special case of the IST

Consider, within the IST, the case $\epsilon \to 0$, when the transformed potential is isospectral to the original, just as is the case in supersymmetric quantum mechanics (SUSYQM). Taking, for convenience, that one of the fundamental solutions (say φ_1) has the property $\lim_{\epsilon \to 0} \varphi_1(z) = \psi_k(z)$, the Wronskian may be written as $W\{\varphi_1, \psi_k\} + \alpha W\{\varphi_2, \psi_k\}$, where the first term becomes

$$\lim_{\epsilon \to 0} W\{\varphi_1, \psi_k\} = \frac{2m^*\epsilon}{\hbar^2} \int_{-\infty}^{z} \psi_k^2(z') dz' \tag{11}$$

while the second term becomes a constant αC, where $C = \lim_{\epsilon \to 0} W\{\varphi_2, \psi_k\}$. If α is given a finite value, then $W\{\psi_\epsilon, \psi_k\} = \alpha C$ is constant, which corresponds to identity transform of the potential, $U_{IST}(z) = U(z)$, for any α. However, taking α tends to zero so that $\alpha(\epsilon) C \to 2m^*\epsilon\lambda/\hbar^2$, where λ is an arbitrary constant, gives the Wronskian

$$W\{\psi_\epsilon, \psi_k\} \to \frac{2m^*\epsilon}{\hbar^2} \left[\lambda + \int_{-\infty}^{z} \psi_k^2(z') dz' \right] \equiv \frac{2m^*\epsilon}{\hbar^2} W_{SS}(z) \tag{12}$$

and the potential $U_{IST}(z)$ isospectral to the original $U(z)$ is

$$U_{IST}(z) = U(z) - \frac{\hbar^2}{m^*} \frac{d^2}{dz^2} \ln W_{SS}(z) = U_{SS}(z) \tag{13}$$

where the subscript SS refers to the expressions derived in the SUSYQM [17].

Therefore, we conclude that the optimized potential obtained via IST will never be inferior to that obtained via SUSYQM, provided one starts with the same class of initial potentials in both approaches [11, 12](though the improvement in some examples may not be drastic).

IV Mapping the variable mass into a constant mass Hamiltonian

In semiconductor quantum well structures based upon graded semiconductor alloys, it is not only the potential, but also the electron effective mass, that are position dependent. The results of Sec. II, relying on the constant-mass assumption, are thus likely not to be directly applicable to almost all common QW's (the exception are some specially designed QW's based upon quaternary alloys, where

the effective mass can be kept constant while the potential varies). In technologically more convenient, ternary alloy based QW's, the effective mass is necessarily position dependent, as discussed below, and some additional considerations are due, in order to be able to use the results of Sec. II for these structures. The approach we use is to map the variable mass Hamiltonian onto one with constant mass. Here we extend the considerations presented in Ref. [13], to include the cases of truncated and asymmetric potentials.

Consider a QW based upon a ternary semiconductor alloy $A_x B_{1-x} C$, with the mole fraction varying along the z axis, i.e., $x = x(z)$. We take the conduction band edge in the BC compound to be lower by ΔV than that in the AC compound, while the electron effective masses in the two are m_{BC} and m_{AC}, respectively. Then the potential experienced by electrons (the conduction band edge), and their effective mass both vary along the z axis as $V(z) = \Delta V \cdot x(z)$, and $m(z) = m_{AC} x(z) + m_{BC}[1 - x(z)]$. Thus, in ternary alloys the potential and the effective mass are related as $V(z) = (\Delta V/\Delta m)[m(z) - m_{BC}] = \theta \cdot [m(z) - m_{BC}]$, where $\Delta m = m_{AC} - m_{BC}$ and $\theta = \Delta V/\Delta m$.

With the position dependent effective mass, the Schrödinger equation for the envelope functions has the Ben Daniel - Duke form [14], i.e., with the notation introduced above:

$$\frac{d}{dz}\left(\frac{1}{m(z)}\frac{d\psi(z)}{dz}\right) + a\left[E - \theta\left(m(z) - m_{BC}\right)\right]\psi(z) = 0 \tag{14}$$

where $a = 2m_0/\hbar^2$, and m_0 is the free electron mass. Now we aim at finding the function $m(z)$, and, uniquely related to it, $V(z)$ and $x(z)$, such that eigenstates of Eq. (14) are identical to those of the constant-mass Eq. (8) with the potential $U_{IST}(z)$ specified in advance. For this purpose we introduce a new coordinate y, such that $z = f(y)$, where the function $f(\ldots)$ will be specified later. In terms of this new coordinate, Eq. (11) takes the form

$$\frac{d^2 u(y)}{dy^2} + \left\{A(y) + a\underline{m}(y)f'^2(y)\left[E - \theta\left(\underline{m}(y) - m_{BC}\right)\right]\right\}u(y) = 0 \tag{15}$$

with

$$A(y) = -\frac{1}{4}\left(\frac{d\,\ln[\underline{m}(y)\,f'(y)]}{dy}\right)^2 + \frac{1}{2}\frac{d^2\,\ln[\underline{m}(y)\,f'(y)]}{dy^2} \tag{16}$$

where $u(y) = const \cdot \underline{\psi}(y)/\sqrt{\underline{m}(y)\,f'(y)}$, and $\underline{\psi}(y) = \psi[f(y)] = \psi(z)$, $\underline{m}(y) = m[f(y)] = m(z)$, $f'(y) = df(y)/dy$. Eqs. (14) and (15) clearly have identical spectra.

Now we require the constant mass $(m^* = m_{BC})$ Schrödinger equation with the optimized potential $U_{IST}(y, \alpha_{opt})$, Eq. (8), to coincide with Eq. (15). This results in a system of two equations, from which we find

$$\underline{m}(y) = \frac{1}{4am_{BC}\theta}\nu^{-2}(y) \tag{17}$$

$$z = f(y) = \int_0^y \sqrt{\frac{m_{BC}}{\underline{m}(y')}}\,dy'. \tag{18}$$

where the function $\nu(y)$ in Eq. (17) is found by solving the nonlinear differential equation

$$2\nu(y)\frac{d^2\nu(y)}{dy^2} - \left(\frac{d\nu(y)}{dy}\right)^2 - 4am_{BC}\left[\theta\,m_{BC} + V(y)\right]\nu^2(y) + 1 = 0. \tag{19}$$

Eq. (19) may be solved by first finding the particular solutions $\zeta_{L,R}(y)$ of the characteristic equation [15] (here we specialize to the case of a truncated potential $V(y) = V = const$ for $|y| > \Delta$, and $V(y) = U_{IST}(y, \alpha_{opt})$ for $|y| < \Delta$)

$$\frac{d^2\zeta(y)}{dy^2} - am_{BC}\left[\theta\,m_{BC} + \left\{\begin{array}{c}V\\U_{IST}(y;\alpha_{opt})\end{array}\right\}\right]\zeta(y) = 0, \qquad \begin{array}{c}|y| > \Delta\\|y| < \Delta\end{array} \tag{20}$$

chosen so that their Wronskian squared equals unity. In analogy to scattering theory, the two linearly independent solutions of Eq. (20) may be written as ($\kappa^2 = am_{BC}[\theta m_{BC} + V]$):

$$\zeta_{L,R}(y) = \begin{cases} \exp(\kappa y) + R_L \exp(-\kappa y), & T_R \exp(-\kappa y) & y < -\Delta \\ A_{L,R} s_1(y) + B_{L,R} s_2(y) & -\Delta < y < \Delta \\ T_L \exp(\kappa y), & R_R \exp(\kappa y) + \exp(-\kappa y) & y > \Delta \end{cases} \quad (21)$$

where $s_{1,2}(y)$ are the particular solutions of (20), satisfying the fundamental initial conditions $s_1(0) = s_2'(0) = 1$, and $s_2(0) = s_1'(0) = 0$, and may be found numerically, while the constants $T_{L,R}$, $R_{L,R}$, $A_{L,R}$, and $B_{L,R}$ are determined from the Wronskian properties and the boundary conditions at $y = \pm\Delta$. These constants depend slightly on the choice of Δ, but this dependence becomes diminishingly weak as Δ increases. We also use the fact that $T_L = T_R = T$.

Since the Wronskian squared of ζ_L and ζ_R equals unity, in terms of the two particular solutions of (20) the general solution of Eq. (19) may be written as [15]

$$\nu(y) = [C_e \zeta_L^2(y) \pm \sqrt{1 + 4C_e C_o}\, \zeta_L(y)\zeta_R(y) + C_o \zeta_R^2(y)]/(2\kappa T) \quad (22)$$

where $C_{e,o}$ are constants to be determined. Now, having in mind the physically acceptable values of the QW parameters, i.e., $m(|y| \to \infty) = V(\Delta m/\Delta V) + m_{BC} = m_b$ and $V(|y| > \Delta) = V$ (inside the barrier), and accounting for the asymptotic behaviour of $\exp(\pm\kappa y)$, we find that the condition $\nu(|y| \gg \Delta) \to const. \neq 0$ is satisfied by the choice of constants

$$C_o = \pm\frac{TR_L}{(R_R R_L - T^2)}, \quad C_e = \pm\frac{TR_R}{(R_R R_L - T^2)}. \quad (23)$$

Finally, substitution into Eq. (17) gives the effective mass variation [16]:

$$\underline{m}(y) = \frac{m_b}{\left[1 + \frac{R_{R,L}}{(R_R R_L - T^2)} \exp(\pm 2\kappa y)\right]^2}, \quad |y| > \Delta \quad (24)$$

where subscript $R(L)$ and $+(-)$ in the exponential refer to $y < -\Delta$ ($y > \Delta$), and

$$\underline{m}(y) = \frac{m_b T^2 (R_R R_L - T^2)^2}{[TR_R \zeta_L^2(y) - (R_R R_L + T^2)\zeta_L(y)\zeta_R(y) + TR_L \zeta_R^2(y)]^2}, \quad |y| < \Delta. \quad (25)$$

The real-space variation of the effective mass $m(z) = m[f(y)] = \underline{m}(y)$ may then be found from (18) and (24)-(25) numerically, and then the potential $V(z)$ and the grading function $x(z)$ directly follow, e.g.

$$V(z) = V[z(y)] = \underline{V}(y) = \frac{\Delta V}{\Delta m}[\underline{m}(y) - m_{BC}] \quad (26)$$

The final Hamiltonian has the effective mass following the potential, as corresponds to ternary alloys based QW's, and is fully isospectral to the Hamiltonian with the constant effective mass $m^* = m_{BC}$ and the optimized potential $U_{IST}(y, \alpha_{opt})$, Eq. (8). Its normalized wave functions are given by

$$\underline{\psi}_i(y) = [\underline{m}(y)/m_{BC}]^{1/4} \psi_i^{IST}(y) \quad (27)$$

in parametric form, i.e., with $z = f(y)$, Eq. (18). The wave functions $\psi_i^{IST}(y)$, Eqs. (9) and (10), are here assumed to be already normalized, i.e., $\int |\psi_i^{IST}(y)|^2 dy = 1$.

V   Numerical results and discussion

V.1   Second harmonic generation

To illustrate the above considerations, we have performed optimization calculations to design a QW structure for double resonance SHG of CO_2 laser radiation ($\hbar\omega = \Delta E = 116$ meV). Taking the lowest three levels to be relevant in this process, a well depth of at least 400 meV is necessary to accomodate these levels, and the compounds AC and BC should be chosen accordingly. The commonly used $Al_xGa_{1-x}As$ system meets this requirement, and has the additional advantages that it is strain-free, and technologically well understood, so it was chosen for further work. The conduction band , -valley discontinuity is $\Delta V = 750$ meV, and electron effective masses in the two compounds are $m_{GaAs} = 0.066\,m_0$ and $m_{AlAs} = 0.15\,m_0$ [14].

In QW's with , -valley related levels, it is only the z component (perpendicular to the QW layer) of the electric field that is active in intersubband transitions. Considering the process of SHG, the second-order nonlinear susceptibility, that relates the field at pump frequency and the polarization at harmonic frequency, is largest under double resonance conditions, i.e., $E_2 - E_1 = E_3 - E_2 = \hbar\omega$, and then its only relevant component amounts to

$$\chi_{zzz}^{2\omega} = \frac{e^3(\rho_{11} - \rho_{22})}{L_z\epsilon_0}\frac{\mu_{12}\mu_{23}\mu_{31}}{(\hbar, {}_2)^2}. \tag{28}$$

where $\mu_{ij} = \langle i|z|j\rangle$ are the transition matrix elements (dipole moments), L_z denotes the QW structure width, ρ_{ii} is the electron density in state i per unit well surface, $\hbar, {}_2$ is the linewidth (taken common for all transitions). By varying the QW profile (potential shape) one varies the dipole moments, while the linewidth is affected far less significantly. Therefore, maximizing $\chi_{zzz}^{2\omega}$ effectively amounts to maximizing the cyclic product of matrix elements, $\Pi^{(2)} = \mu_{12}\mu_{23}\mu_{31}$, in the numerator of (28).

To find the optimized asymmetric potential, with the largest $\Pi^{(2)}$, we start from a family of truncated parabolic potentials of the form

$$U(z) = \begin{cases} \frac{m^*}{2}\left(\frac{\Delta\mathcal{E}(N)}{\hbar}\right)^2 z^2, & |z| < \Delta \\ V, & |z| > \Delta \end{cases} \tag{29}$$

where $\Delta\mathcal{E}(N) = 84.75 + N \cdot 0.25$ [meV], and N is an integer incremented from 1 to 241. The effective mass in (29) is taken constant and equal to that in GaAs, and the potential is truncated at $V = 400$ meV (hence the well width Δ changes with incrementing N). Because of truncation this is not exactly a linear harmonic oscillator, and in no case will the levels be strictly equispaced. Furthermore, their energies will change as N (i.e., the well width) varies, so none of the potentials (29) meets the double resonance condition. Applying an isospectral transform like supersymmetry [17] will make them asymmetric, and all μ_{ij} would then be nonzero, but will not correct the level's energies. Deviation of these energies from their desired positions, that would provide double resonance at $\hbar\omega = 116$ meV (i.e., in the spirit of Sec. II, the necessary shifts $\epsilon_2 = E_2^{shift} - E_2$ and $\epsilon_3 = E_3^{shift} - E_3$), are given in Fig. 2 as they depend on the frequency $\Delta\mathcal{E}/\hbar$.

The initial potentials are then processed along the lines presented in Sec. II: The third level of $U(z)$ is shifted to be spaced by 232 meV from the first, i.e., $E_3^{shift} = E_3 + \epsilon_3 = E_1 + 232$ meV, and, by choosing the constants, the parity of its wave function is set opposite from that of $\psi_3(z)$. Next, the second level is shifted according to $E_2^{shift} = E_2 + \epsilon_2 = E_1 + 116$ meV. This procedure was repeated for all potentials in the family (29), and in each case the asymmetrization parameter α was varied while monitoring the value of the matrix elements product $\Pi^{(2)}$ (calculated by using Eqs. (9) and (10)). With the initial potentials chosen symmetric, it is enough to give only positive values to the parameter α, because here $\Pi^{(2)}(-\alpha) = -\Pi^{(2)}(\alpha)$, otherwise both positive and negative values should be explored. We should also note here that, in doing these calculations, one has to mind that the Wronskians in Eqs. (4-10) do not cross zero, in order to avoid singularities and physically unacceptable solutions. The results of this search are displayed in Fig. 3. The largest value of $\Pi^{(2)}$ is
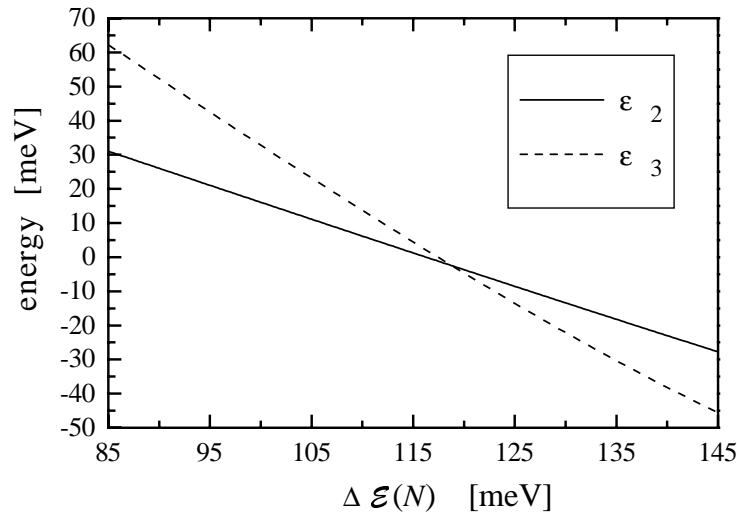
FIG. 2. The shifts $\epsilon_2 = E_2^{shift} - E_2$ and $\epsilon_3 = E_3^{shift} - E_3$, of levels 2 and 3, necessary to obtain equal spacings (double resonance), as they depend on the parabolic potential slope $\Delta\mathcal{E}$.

found with the values of parameters $N = 68$ (i.e., $\Delta\mathcal{E}(68) = 101.75$ meV), and $\alpha_{opt} = 0.18$. Here we have $\Pi^{(2)} = 4375.2$ Å^3, with individual dipole moments $\mu_{12} = 16.96$ Å, $\mu_{23} = 28.12$ Å, and $\mu_{31} = 9.17$ Å. The corresponding optimized potential $U_{IST}(z; \alpha_{opt})$ is given in Fig. 4. It may be interesting to note that the original potential, from which this optimized potential was derived, had its levels at $E_1 = 50.87$ meV, $E_2 = 152.54$ meV, and $E_3 = 253.45$ meV, therefore (with $\Delta E_{21} = 101.67$ meV and $\Delta E_{32} = 100.91$ meV) it was not matched for resonant SHG of $\hbar\omega = 116$ meV radiation, and its asymmetryzation via the isospectral supersymmetric transform alone would not help. However, the IST based levels shifts, accompanied by asymmetryzation, did the job.
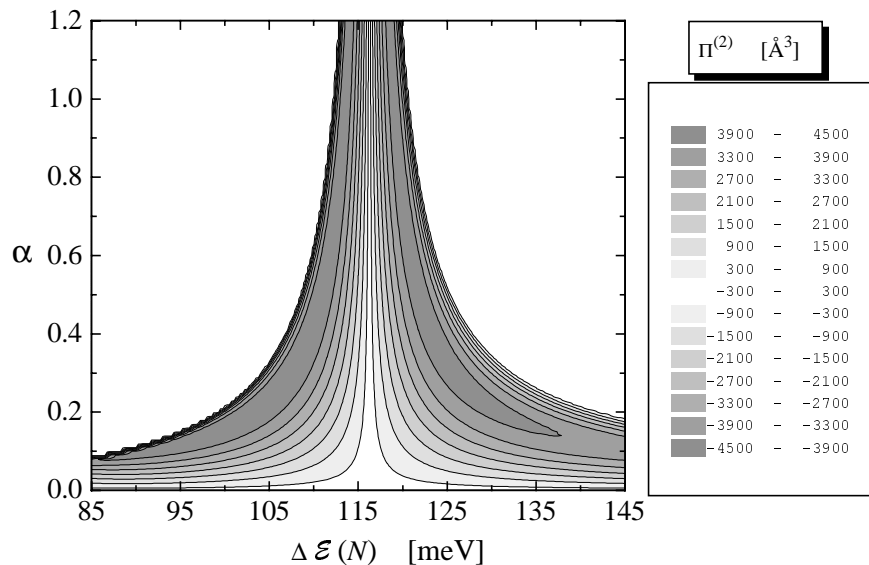

FIG. 3. Values of the matrix elements product $\Pi^{(2)} = \mu_{12}\mu_{23}\mu_{31}$ obtainable with various values of the QW design parameters $\Delta\mathcal{E}(N)$ and α.

The potential optimized so far, assuming a constant effective mass, is not directly realizable in graded ternary alloys, because the effective mass there cannot be kept constant. To make it realizable, we have to map the Hamiltonian with $U_{IST}(z; \alpha_{opt})$ and $m^* = const$ into the Hamiltonian with the

effective mass following the potential, as corresponds to the chosen ternary alloy system. This is performed along the lines described in Sec. IV, in particular Eqs. (24-26) and (18). Having obtained the optimized Hamiltonian realizable in graded $Al_xGa_{1-x}As$, it remains to check the product of matrix elements in this new (but realistic) system. Using Eq. (27) we find that the matrix elements $\mu_{ij} = \langle \underline{\psi}_i(y)|z(y)|\underline{\psi}_j(y)\rangle$ now amount to $\mu_{12} = 16.31$ Å, $\mu_{23} = 25.86$ Å, and $\mu_{31} = 9.27$ Å, so $\Pi^{(2)} = 3910$ Å^3, Ref. [16], has somewhat decreased from what was predicted above. This should have been expected, since the effective mass throughout the structure generally exceeds the constant value $m^* = 0.066$ initially taken, and $\mu \sim m^{-1/2}$, Ref. [2]. The realistic optimized potential, given in Fig. 4, has a negative undershoot, and hence has a somewhat larger span of values than the original $U(z)$. However, the negative undershoot itself makes no difficulties in realization, because the reference zero of the potential is irrelevant for physics, the whole picture may be shifted up or down at will. In realizing this potential by grading the $Al_xGa_{1-x}As$ alloy, via the relation $x(z) \cdot \Delta V = V(z) + const$, one may set $const = |min(V(z))|$, so that $x = 0$ at the lowest point of the well, and the full potential span offered by $Al_xGa_{1-x}As$ is taken advantage of.

Comparison of the maximal value of $\Pi^{(2)}$ found here against the best values obtained elsewhere in step-graded QW's (e.g. $\Pi^{(2)} = 2394$ Å^3, Ref. [2], or $\Pi^{(2)} = 2635$ Å^3, Ref. [18]) shows that $\Pi^{(2)}$ obtained in this work (though in a continuously graded QW) is better by $\sim 30\%$. We should also note that, in case of deeper wells, one would have to account for the bulk nonparabolicity. It cannot be directly included in the theory described above because an energy dependent potential would result, and this effect should better be accounted by slight numerical "repolishing" of the optimized potential obtained without it. Our previous experience with SUSYQM based QW optimization in respect to second harmonic generation [7] indicates that these have a rather mild influence on the best potential shape (slightly squeezing the output of idealized calculations in order to compensate for the nonparabolicity-increased effective mass).
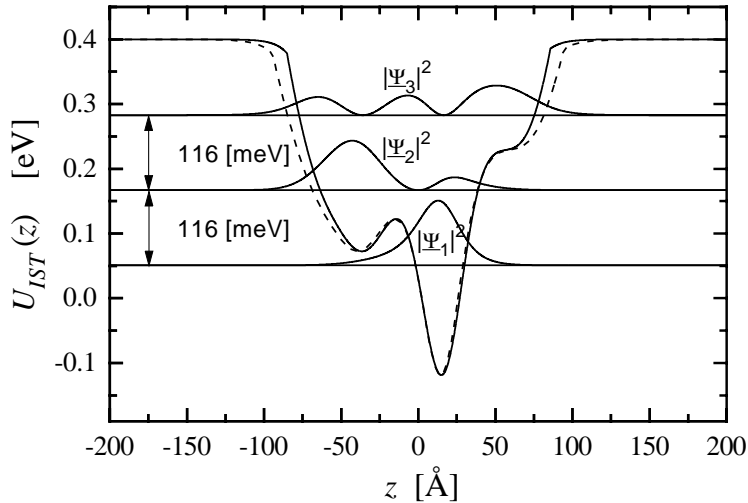

FIG. 4. The optimized potential $U_{IST}(z)$, corresponding to $\Delta\mathcal{E} = 101.75$ meV and $\alpha_{opt} = 0.18$, with constant effective mass, and the optimized realizable potential $V(z)$ with the variable effective mass following it, as corresponds to graded AlGaAs alloy.

It should be noted that the particular schedule of steps used above is just one among a few other possibilities. To achieve the required positions of the three levels, one may choose to fix the second or the third, and shift the other two, which would produce a potential different from the one obtained here. Furthermore, additional freedom may be gained by introducing more free constants when constructing the ψ_ϵ functions from the fundamental solutions. This should be done with great care, however, because e.g. introducing a parameter β in ψ_{ϵ_3} (analogous to α in ψ_{ϵ_2}) would result in a highly oscillatory term $(d^2/dz^2)\ln[W\{\psi_3, \psi_{\epsilon_3}\}]$, which would eventually yield a highly oscillatory, and

difficult to realize, final potential.

V.2 Optical rectification

The density matrix calculations [2, 19] for a two-level system shows that optical rectification coefficient $\chi_0^{(2)}$ becomes largest at resonance (when the spacing between the two relevant levels of the system equals the photon energy, i.e., $\hbar\omega = E_2 - E_1 = \hbar\omega_{21}$), and then amounts to

$$\chi_{0\,max}^{(2)} = 2\frac{e^3 T_1 T_2}{\epsilon_0 \hbar^2}(\rho_{11} - \rho_{22})\mu_{12}^2 \delta_{12} \qquad (30)$$

Here ρ_{ii}, $i = 1, 2$ denote electron densities on i-th quantized state per unit well surface, $\mu_{12} = \langle 1|z|2 \rangle$ and $\delta_{12} = \langle 1|z|1 \rangle - \langle 2|z|2 \rangle$ are the transition dipole moment and the difference of permanent dipole moments of the two states. Furthermore, T_1 and T_2 are the diagonal and off-diagonal relaxation times in the density matrix equation (the excited state lifetime and the dephasing time, respectively).

Concerning the possibilities of increasing $\chi_0^{(2)}$, the dephasing time T_2, arising from various scattering mechanisms, depends only slightly on the QW shape [20]. The same is true for the lifetime T_1. In fact, the effective T_1 may be increased by introducing additional, optically inactive, metastable states into the system, but $\chi_0^{(2)}$ is then increased at the expense of making the response slower [21]. The dipole moments μ_{12} and δ_{12}, on the other hand, very much depend on the QW shape, which may thus be employed for maximizing $\chi_0^{(2)}$. While varying the QW shape, it is essential to keep the levels spacing as specified, to match the resonance condition. Clearly, only asymmetric structures are useful for optical rectification, and may be realized by asymmetric composition grading in a stepwise constant or continuous manner.

In order to optimize the QW shape in respect to $\chi_0^{(2)}$, therefore, one first sets some initial potential $U(z)$, the states of which are likely not to be properly spaced. Then, a family of modified potentials (2) is generated, with the spacing between the two states relevant for $\chi_0^{(2)}$ corrected by making an appropriate shift ϵ. By varying α, one evaluates the matrix elements $\mu_{12} = \langle \psi_1^{IST}(z)|z|\psi_2^{IST}(z) \rangle$ and $\delta_{12} = \langle \psi_1^{IST}(z)|z|\psi_1^{IST}(z) \rangle - \langle \psi_2^{IST}(z)|z|\psi_2^{IST}(z) \rangle$, and finds the optimal value of the parameter $\alpha = \alpha_{opt}$ which maximizes the product of matrix elements $\Pi^{(0)} = \mu_{12}^2 \delta_{12}$ appearing in $\chi_0^{(2)}$. The corresponding potential $U_{IST}(z; \alpha_{opt})$ is the best potential shape that may be derived from the particular original potential $U(z)$ chosen (on which the optimized $\chi_{0\,max}^{(2)}$ will clearly depend). The search may be widened by starting with a number, or a family, of initial potentials.

To give a specific example, consider the optimized design of a QW structure for resonant optical rectification of CO_2 laser radiation ($\hbar\omega = \Delta E = 116$ meV). We start with a family of truncated parabolic potentials Eq. (29) where $\Delta\mathcal{E}(N) = 100 + N \times 0.5$ [meV], and N is an integer incremented from 0 to 145. The effective mass in (2) is taken constant and equal to that in GaAs, and the potential is truncated at $V = 190$ meV or $V = 200$ meV in two sets of calculations (note that the well width Δ changes with incrementing N). Levels energies will change as N (i.e., the well width) varies, so the potentials (2) generally do not meet the resonance condition, and shifting of one of the levels is first necessary, which we perform for the second level ($k = 2$ in the above expressions) to obtain $E_2^{IST} - E_1^{IST} = \hbar\omega$, with $E_2^{IST} = E_2 + \epsilon$ and $E_1^{IST} = E_1$. For each specific initial potential (i.e., the value of N in (2) we vary the parameter α and evaluate the dipole matrix elements, from the wave functions, in order to find the largest $\Pi^{(0)} = \mu_{12}^2 \delta_{12}$ corresponding to this N, and repeat this procedure for all N. It is worth noting that, due to the symmetry of the initial potentials, it is enough to scan only over positive values of α (because $\Pi^{(0)}(-\alpha) = -\Pi^{(0)}(\alpha)$), otherwise both positive and negative values should be explored. Furthermore, one has to mind that the Wronskians in Eq. (2) do not cross zero, in order to avoid singularities and physically unacceptable solutions.

Results of the search done for the truncation $V = 200$ meV are given in Fig. 5(a). We find that the largest value of $\Pi^{(0)}$ occurs for the values of parameters $N = 101$ (i.e., $\Delta\mathcal{E}(101) = 150.5$ meV), and $\alpha_{opt} = 0.48$. Here we have $\Pi_{max}^{(0)} = 15677.5$ Å^3, with individual dipole moments $\mu_{12} = 15.75$ Å,
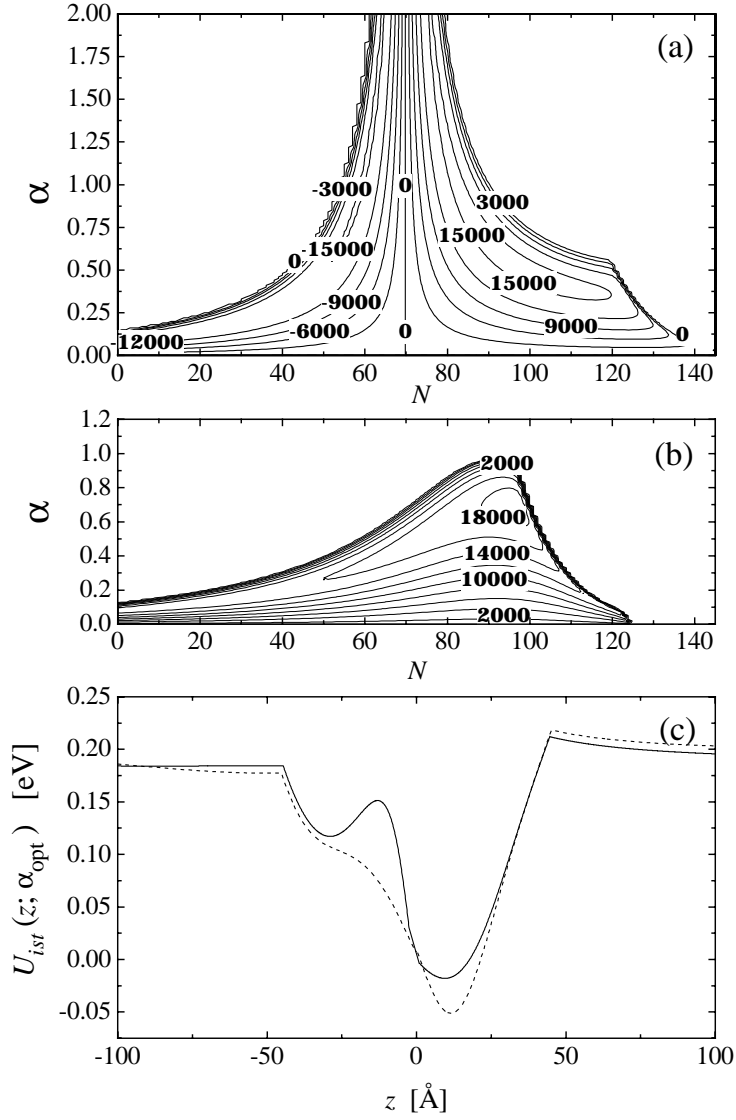
FIG. 5. (a) Values of the matrix elements product $\Pi^{(0)} = \mu_{12}^2 \delta_{12}$ obtainable with various values of the QW design parameters N and α, with the initial parabolic potential truncated at $V = 200$ meV, and $V = 190$ meV (b); (c) the optimized potentials $U_{IST}(z)$ for $N = 101$ and $\alpha_{opt} = 0.48$ coresponding to $V = 200$ meV (dashed line) and $N = 97$ and $\alpha_{opt} = 0.70$ coresponding to $V = 190$ meV (solid line).

and $\delta_{12} = 63.20$ Å. The initial potential had its levels at $E_1 = 73.01$ meV and $E_2 = 192.65$ meV, so a shift of $\epsilon = -3.64$ meV of the second state was performed to bring the spacing $\Delta E_{21} = 119.64$ meV to the desired 116 meV. In another search (Fig. 5(b)), with the truncation $V = 190$ meV, we find $\Pi_{max}^{(0)} = 18770.6$ Å^3, Ref. [12], with $\mu_{12} = 13.51$ Å, and $\delta_{12} = 102.84$ Å, which is obtained for $\Delta \mathcal{E}(97) = 148.5$ meV and $\alpha_{opt} = 0.70$. The corresponding initial potential in this case had $E_1 = 71.75$ meV and $E_2 = 185.71$ meV, and required a shift $\epsilon = 2.04$ meV of the second level to correct the spacing. The optimized potentials $U_{IST}(z; \alpha_{opt})$ are given on Fig. 5(c).

While the optimized potentials may be realized by grading the AlGaAs alloy, in real structures it may be necessary to include corrections due to the position (i.e., the alloy composition) dependent mass and the nonparabolicity. However, with rather low energies of states in the above examples, these corrections are here expected to be very small indeed. Comparison of $\Pi_{max}^{(0)}$ found here against the values obtained elsewhere [2, 22] in step-graded QW's (also based on AlGaAs, within the constant mass approximation, to make a fair comparison), shows that our $\Pi_{max}^{(0)}$ is better by up to 40%, and also

eff. mass model	$\mu_{12}\mu_{23}\mu_{31}$ [Å^3]			$\mu_{12}^2(\mu_{11}-\mu_{22})$ [Å^3]		
	const.	pd.	np.	const.	pd.	np.
Ref. [4]		390.7			1887.6	
Ref. [5]		3256.2				
Ref. [2]	2394			12267		
Ref. [18]			2634.7			8557.2
Ref. [24]		1223.4				
Ref. [22]					12424	
Ref. [25]	3003					
SUSYQM	4279.7	3836.2	3297.6	15322		10660.2
IST	4375.2	3910		18771		

TABLE 1. *The publicated values of dipol matrix element products relevant for SHG and OR. Also in table are lsted values obtained by supersymmetric quantum mechanics and inverse spectral theory. All results are related to GaAs/AlGaAs QW's with diferent effective mass models: const.-$m^* = 0.066m_0$, pd. - position dependant effective mass, and np. - with nonparabolicity of subbands included.*

somewhat better that the value we have previously obtained using the SUSYQM based optimization ($\Pi^{(0)} = 15322$ Å^3) [8]. In this latter we have started with the potential (2) truncated at $V = 200$ meV, its parameters already fitted to provide the correct levels spacing, and then varied it isospectrally in order to find the best potential shape (however, the potential truncated at $V = 190$ meV could not be handled by this approach). The similarity between the SUSYQM and IST results in the $V = 200$ meV case might have been physically expected from the rather small value of the shift ϵ which was here necessary to correct the levels spacing. Indeed, the SUSUQM is in fact contained in IST, as we show above.

Finaly, the dipol matrix element products relevant for the second harmonic generation and optical rectification published until now, are summarized in Table 1.

VI   Conclusion

A procedure for the design of quantum well structures optimized in respect to the intersubband double resonant second harmonic generation and resonant optical rectification was proposed and discussed. Starting with an arbitrary potential, with levels energies not properly positioned for this application, this procedure allows one to shift the relevant levels to the desired positions, by using the inverse spectral theory, and vary the potential shape, controlled by free parameters, in order to find the shape that maximizes the matrix elements relevant for second order nonlinearity. Furthermore, the procedure is devised to handle the case of position dependent effective mass, and is thus applicable to realistic QW structures. Its applicability was demonstrated by designing a graded AlGaAs ternary alloy quantum well optimized in the second order nonlinearity at 10.6 μm. Starting with a truncated parabolic potential, the final optimized QW potential was obtained, with the second order susceptibility significantly ($\sim$30%) exceeding values previously obtained in the literature, and is slightly better than obtained in Refs. [7, 8].

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