Two methods of quantum well profile optimization for maximal nonlinear optical susceptibilities

J. Radovanović
Institute of Physics, Pregrevica 118, 11080 Zemun-Belgrade, Yugoslavia

G. Todorović
Faculty of Civil Engineering, University of Belgrade, Bulevar Revolucije 73, 11120 Belgrade, Yugoslavia

V. Milanović*
Faculty of Electrical Engineering, University of Belgrade, Bulevar Revolucije 73, 11120 Belgrade, Yugoslavia

Z. Ikonić
School of Electronic and Electrical Engineering, University of Leeds, Leeds LS2 9JT, United Kingdom

D. Indjin
Faculty of Electrical Engineering, University of Belgrade, Bulevar Revolucije 73, 11120 Belgrade, Yugoslavia
(Received 24 April 2000; revised manuscript received 12 October 2000; published 2 March 2001)

Two different approaches for the optimization of the quantum well profile are proposed and discussed. One is the multiparameter procedure, based on the inverse spectral theory (IST) and supersymmetric quantum mechanics (SUSYQM), which is an extension of the single-parameter procedure devised earlier for this purpose. Another approach combines the simulated annealing and variational calculus. The two approaches are compared on the example of optimizing the well profile to get maximal resonant second-order susceptibility at 10.6 μm (116 meV). Within the multiparameter IST/SUSYQM procedures, we find that the two-parameter procedure delivers significantly better results than the single-parameter procedure, while introducing more parameters does not result in any further improvement. However, even better results (by about 20%) were obtained with the variational procedure, which, though more time consuming, is free from any unnecessary constraints and may thus lead to global optimization.

DOI: 10.1103/PhysRevB.63.115327 PACS number(s): 73.21.−b, 78.66.−w

I. INTRODUCTION

Intersubband optical transitions in quantum well (QW) structures are continuously attracting research attention in the last two decades. Due to considerable values of dipole transition matrix elements, the QW's have quite remarkable linear and nonlinear optical properties. Various effects in QW's may be enhanced by suitably tailoring their electronic structure (''band structure engineering''). A particular effect may be grossly enhanced by achieving the resonance conditions, i.e., appropriate spacings between the most relevant states, and also by tailoring the wave functions so that the (combinations of) matrix elements relevant for this particular effect are maximized.

A few techniques for systematic optimization of the QW profile so as to maximize a particular effect have been proposed. Within the class of continuously graded QW's, these rely on the inverse spectral theory (IST) and the supersymmetric quantum mechanics[1] (SUSYQM), which enable one to manipulate quantum states (shifting, deletion, insertion) and also perform the spectrum-preserving (isospectral) variation of the potential profile controlled by one or more scalar parameters introduced thereby, thus changing the wave functions.[2–4] Analogous techniques, simpler but also very effective, have been developed for step-graded QW profile optimization.[4,5] In our previous papers on the optimization of continuously graded QW's,[2–4] we have employed the variation of a single free parameter introduced by one SUSYQM transform. While results better than those reported elsewhere in the literature have been consistently achieved, it is clear that introducing additional free parameters in these procedures might allow a larger variation of the potential shape, and therefore generally better final results. Certainly, introducing more parameters make the optimization procedures more complicated, and in first part of this paper we address the problem of the effectiveness of such multiparameter SUSYQM optimization of the QW profile. To be specific, we start with the initial potential of the Pöschl-Teller-type, occasionally employed in the research of semiconductor QW's,[6,7] and attempt to maximize the double resonance second-order susceptibility relevant for the second-harmonic generation (SHG).

The SUSYQM based optimization is always constrained implicitly rather than explicitly: it is the starting potential that ultimately determines what may be achieved. While introducing more parameters does lead to a larger freedom, the variation of the potential shape is still too restricted in that the isospectral transform preserves not only the relevant part of the energy spectrum as is required, but (unnecessarily) preserves the whole energy spectrum. It may well be possible, therefore, that the QW profile obtained that way is not globally optimal (which does not imply that it has to be significantly worse than the globally optimal potential, however).

In the second part we make use of the variational method (i.e., the optimal control theory, in more modern terminol-

ogy) as a tool of QW profile optimization. This approach has been employed in recent years mostly in the time domain, e.g., to optimize trajectory of a driven quantum system, the exception being Ref. 8 where the potential was optimized for the tunneling transmission properties. Here we consider this method as a technique for free variation of the QW profile in order to find the globally optimal profile, maximizing the nonlinear susceptibility. As will be discussed in more details later, absent the analytic solution to this formulation, one should provide a good starting point to an iterative procedure devised to head towards the optimal QW profile. This starting point is provided by a stochastic optimization routine, in particular the simulated annealing method. In contrast to the SUSYQM based method which has some implicit unnecessary constraints, the variational approach has none, and may thus be expected to deliver a global optimum.

II. THEORETICAL CONSIDERATIONS

A. The second-order susceptibility

Considering the *n*-doped semiconductor QW's and the incident photon energies well below the band gap, the polarization response is predominantly determined by intersubband transitions between the quantized states within the conduction band. The second-order susceptibility describing the polarization at twice the incident photon frequency, is[5]

$$\chi^{(2)}_{zzz}=\frac{e^3}{L_z\epsilon_0}\sum_i\sum_j\frac{1}{2\hbar\omega-\Delta E_{ji}-i\hbar\Gamma_{ji}}\times\sum_l M_{ij}M_{jl}M_{li}\left[\frac{\rho_{ii}-\rho_{ll}}{\hbar\omega-\Delta E_{li}-i\hbar\Gamma_{li}}-\frac{\rho_{ll}-\rho_{jj}}{\hbar\omega-\Delta E_{jl}-i\hbar\Gamma_{jl}}\right], \tag{1}$$

where $M_{ij}=\langle\psi_i|z|\psi_j\rangle$ denotes the dipole transition matrix element, ΔE_{ij} the $i\rightarrow j$ transition energy, ρ_{ii} is the surface-electron density in ith state, Γ_{ij} the off-diagonal relaxation rate, and L_z the structure length. Taking the ground ($i=1$) state density $\rho_{11}\gg\rho_{ii}$ for all $i>1$, usually fulfilled in real structures, Eq. (1) becomes

$$\chi^{(2)}_{zzz}=\frac{e^3\rho_{11}}{L_z\epsilon_0}\frac{M_{12}M_{23}M_{31}}{(2\hbar\omega-\Delta E_{31}-i\hbar\Gamma_{31})(\hbar\omega-\Delta E_{21}-i\hbar\Gamma_{21})}. \tag{2}$$

Clearly, the $\chi^{(2)}_{zzz}$ will take the maximal value at double resonance, i.e., if $\hbar\omega=\Delta E_{21}=\Delta E_{31}/2$, and assuming all the relaxation rates are the same, Eq. (1) reduces to

$$\chi^{(2)}_{zzz}=\frac{e^3\rho_{11}}{L_z\epsilon_0\hbar^2\Gamma^2}M_{12}M_{23}M_{31}. \tag{3}$$

To obtain the largest $\chi^{(2)}_{zzz}$, therefore, one should maximize the product of dipole matrix elements $\Pi^{(2)}=M_{12}M_{23}M_{31}$, which depend on the QW profile via the corresponding wave functions. This will be the target function we want to maximize. Clearly, we have here assumed that the off-diagonal relaxation rate(s), i.e. the transition linewidths are essentially independent on the QW profile, which is probably justified to a good approximation.

B. Multiparameter supersymmetric optimization

In this section we consider the optimization of the second-order nonlinear susceptibility of a QW by using the SUSYQM and IST. This approach, but in the single-parameter formulation, has been used previously and here we extend it to introduce more free parameters, which should make the method capable of exploring a larger portion of the QW potential space and therefore to eventually give better results. The general expressions for the IST and SUSYQM transforms are given in Ref. 1, or our papers on the QW profile optimization.[2–4] Here we will mainly give the formulas applying for the initial potential of Pöschl-Teller form, since it allows for a number of results to be obtained in analytically (for the same reason we assume the electron effective mass m^* to be position independent). The initial potential reads

$$U(z)=-\frac{U_0}{\cosh^2(\alpha z)}, \tag{4}$$

and its bound states are known analytically[9]

$$E_i=-\frac{\alpha^2}{\beta}(s+1-i)^2,\quad i=1,2,3,\ldots, \tag{5}$$

where

$$s=\frac{\sqrt{1+4U_0\beta/\alpha^2}-1}{2} \tag{6}$$

and $\beta=2m^*/\hbar^2$. The parameter s determines the number of bound (normalizable) states $N_B(N_B=\mathrm{Int}[s])$ supported by this potential.

These relations are very helpful for setting up the initial potential. For instance, to get the potential with three bound states ($s=3$) and with the spacing $\Delta E_{32}=E_3-E_2=116$ meV between the two excited states, we find $U_0=4\Delta E_{32}$ and $\alpha^2=\beta\Delta E_{32}/3$. The corresponding normalized wave functions are (for $s=3$)

$$\psi_1(z)=\frac{\sqrt{15\alpha}}{4\cosh^3(\alpha z)},$$

$$\psi_2(z)=\frac{\sqrt{15\alpha}\sinh(\alpha z)}{2\cosh^3(\alpha z)}, \tag{7}$$

$$\psi_3(z)=\frac{\sqrt{3\alpha}[1-4\sinh^2(\alpha z)]}{4\cosh^3(\alpha z)}.$$

The ground state energy is $E_1=-3U_0/4$, so it is not properly spaced from state 2 to have double resonance for the SHG. However, by shifting the ground state by $\epsilon=U_0/6$, which is accomplished by the IST, we will obtain three states spaced by the required amount ΔE_{32}. The potential $U_{IST}(z)$

that will support such a ground state and not affect the energy of any other state, is given by[3]

$$U_{IST}(z)=U(z)-\frac{2}{\beta}\frac{d^2}{dz^2}[\ln W(z)], \tag{8}$$

where $W(z)$ is the Wronskian

$$W(z)=\psi_\epsilon\frac{d\psi_1}{dz}-\psi_1\frac{d\psi_\epsilon}{dz} \tag{9}$$

of the wave function ψ_1 and a solution ψ_ϵ of the Schrödinger equation

$$\frac{d^2\psi}{dz^2}+\beta[E_1+\epsilon-U(z)]\psi=0 \tag{10}$$

at an energy that is not an eigenenergy of the initial potential.

In case of Pöschl-Teller potential this solution may be written in terms of hypergeometric functions as

$$\psi_\epsilon(z)=\frac{\tanh^2(\alpha z)}{\cosh^2(\alpha z)}{}_2F_1\left[\frac{\sqrt{7}-2}{2},\frac{-\sqrt{7}-2}{2};\frac{3}{2},-\sinh^2(\alpha z)\right], \tag{11}$$

which ensures that $U_{IST}(z)$ and its ground state are even functions. The normalized wave functions of $U_{IST}(z)$ are given by[3]

$$\psi_{i\,IST}(z)=\frac{1}{\sqrt{1-\dfrac{\epsilon}{E_i-E_j}}}\times\left[\psi_i(z)-\beta\epsilon\frac{\psi_\epsilon(z)}{W(z)}\int_{-\infty}^{z}\psi_i(t)\psi_j(t)dt\right],\quad i\neq j \tag{12}$$

and

$$\psi_{j\,IST}(z)=\frac{C\,\psi_j(z)}{W(z)},\quad i=j,\quad i,j\in\{1,2,3\}, \tag{13}$$

where the constant C in Eq. (13) has to be found numerically. These wave functions may be further expressed fully analytically. Introducing the notation

$$S_1(z)={}_2F_1\left[\frac{\sqrt{7}-2}{2},\frac{-\sqrt{7}-2}{2};\frac{3}{2},-\sinh^2(\alpha z)\right],$$

$$S_2(z)={}_2F_1\left[\frac{\sqrt{7}}{2},\frac{-\sqrt{7}}{2};\frac{5}{2},-\sinh^2(\alpha z)\right], \tag{14}$$

the Wronskian has the form

$$W(z)=\frac{-\alpha\sqrt{15\alpha}}{4\cosh^5(\alpha z)}[S_1(z)+\sinh^2(\alpha z)S_2(z)] \tag{15}$$

and the normalized wave functions (12) and (13) read

$$\psi_{1\,IST}(z)=\sqrt{\frac{128\,522\alpha}{194\,307}}\cosh^2(\alpha z)\times[S_1(z)+\sinh^2(\alpha z)S_2(z)], \tag{16}$$

$$\psi_{2\,IST}(z)=\frac{\sqrt{5}\psi_1(z)}{\sqrt{3}}\left[1-\frac{2}{5}\frac{S_1(z)}{S_1(z)+\sinh^2(\alpha z)S_2(z)}\right], \tag{17}$$

$$\psi_{3\,IST}(z)=\frac{2\psi_2(z)}{\sqrt{3}}\left[1+\frac{2\sinh^2(\alpha z)}{1-4\sinh^2(\alpha z)}\times\frac{S_1(z)}{S_1(z)+\sinh^2(\alpha z)S_2(z)}\right]. \tag{18}$$

To make a family of potentials $U_{1\,SS}(z,\lambda_1)$ isospectral to $U_{IST}(z)$, we use the SUSYQM method and choose to delete and restore the ground state, so the transformed potential reads

$$U_{1\,SS}(z,\lambda_1)=U_{IST}(z)-\frac{2}{\beta}\frac{d^2}{dz^2}\left[\ln\left(\lambda_1+\int_{-\infty}^{z}\psi_{1\,IST}^2(t)dt\right)\right] \tag{19}$$

and the normalized wave functions

$$\psi_{i\,S}(z,\lambda_1)=\psi_{i\,IST}(z)-\frac{\psi_{1\,IST}(z)\displaystyle\int_{-\infty}^{z}\psi_{i\,IST}(t)\psi_{1\,IST}(t)dt}{\lambda_1+\displaystyle\int_{-\infty}^{z}\psi_{1\,IST}^2(t)dt},\quad i=2,3 \tag{20}$$

$$\psi_{1\,S}(z,\lambda_1)=\frac{\sqrt{\lambda_1(\lambda_1+1)}\psi_{1\,IST}(z)}{\lambda_1+\displaystyle\int_{-\infty}^{z}\psi_{1\,IST}^2(t)dt}. \tag{21}$$

Now, by deleting and restoring the bound state E_2 of the potential $U_{1\,SS}(z,\lambda_1)$ we derive the two-parameter potential $U_{2\,SS}(z,\lambda_1,\lambda_2)$

$$U_{2\,SS}(z,\lambda_1,\lambda_2)=U_{1\,SS}(z,\lambda_1)-\frac{2}{\beta}\frac{d^2}{dz^2}\left[\ln\left(\lambda_2+\int_{-\infty}^{z}\psi_{2\,S}^2(t)dt\right)\right] \tag{22}$$

and the corresponding wave functions

$$\psi_{i\,SS}(z,\lambda_1,\lambda_2)=\psi_{i\,S}(z,\lambda_1)-\frac{\psi_{2\,S}(z,\lambda_1)\int_{-\infty}^{z}\psi_{i\,S}(t,\lambda_1)\psi_{2\,S}(t,\lambda_1)dt}{\lambda_2+\int_{-\infty}^{z}\psi_{2\,S}^2(t)dt},$$

$$i=1,3, \tag{23}$$

$$\psi_{2\,SS}(z,\lambda_1,\lambda_2)=\frac{\sqrt{\lambda_2(\lambda_2+1)}\psi_{2\,S}(z,\lambda_1)}{\lambda_2+\int_{-\infty}^{z}\psi_{2\,S}^2(t,\lambda_1)dt}. \tag{24}$$

Similar to the conventional one-parameter SUSYQM, the parameters λ_1 and λ_2 take arbitrary values outside the segment $(-1,0)$, so the wave functions (23) and (24) have no singularities.

The product of matrix elements in Eq. (3) now depends on λ_1 and λ_2 as does each matrix element in it, i.e.,

$$M_{ij}(\lambda_1,\lambda_2)=\int_{-\infty}^{+\infty}\psi_{i\,SS}z\psi_{j\,SS}dz \tag{25}$$

and the maximum of $\Pi^{(2)}$ may thus be searched for in the (λ_1,λ_2) plane.

C. Optimization by variational calculus

In the variational calculus a function is varied either completely freely or subject to some number of clearly defined constraints. Therefore, effectively operating with an infinite number of degrees of freedom, it enables the global optimum to be reached. Here we use this approach to vary the QW potential $U(z)$, which ultimately determines all the properties of the QW system. It is first necessary to define the target [i.e., the functional of $U(z)$], which should be maximized. If the second-order susceptibility $\chi^{(2)}$ is of interest, this functional has the form

$$J=\frac{\Pi_{eff}^{(2)}}{C}=\frac{M_{12}M_{23}M_{31}}{\{(\hbar\Delta\omega_{21})^n+\Theta^n\}\{(\hbar\Delta\omega_{32})^n+\Theta^n\}}, \tag{26}$$

where $\Delta\omega_{21}=\omega_{21}-\omega=(E_{21}/\hbar)-\omega$ and $\Delta\omega_{32}=\omega_{32}-\omega$ are the detunings of the corresponding state spacings from the incident photon frequency. The quantity in the denominator, $C=\{(\hbar\Delta\omega_{21})^n+\Theta^n\}\{(\hbar\Delta\omega_{32})^n+\Theta^n\}$, clearly favors achieving resonant conditions. Normally, $n=2$ and Θ is the transition linewidth. More generally though, n may be any even integer and Θ a nonzero constant, which may be suitably chosen so that J is strongly driven towards double resonance in course of optimization, while remaining finite at the exact resonance (the precise form of the denominator becomes irrelevant at the maximum itself). Note that this denominator was not needed in the SUSYQM based optimization because the system was initially chosen to satisfy the double resonance conditions and was then varied isospectrally. Within the variational approach the potential is freely varied, however, which makes it necessary to keep the resonance denominator.

The potential $U(z)$ is allowed to be arbitrary in the ''physically interesting'' optimization segment $[-z_L,z_R]$, and is constant outside. Furthermore, on the boundaries of this segment we assume $\Psi_i(-z_L)=\Psi_i(z_R)=0$, and $\Psi_i'(-z_L)=\Psi_i'(z_R)=0$. The matrix elements are evaluated in this segment only, i.e., $M_{ij}=\int_{-z_L}^{z_R}\Psi_i z\Psi_j dz$. The entries in the target functional J all depend on the potential $U(z)$ since the wave functions Ψ_i and energies E_i solve the same Schrödinger equation

$$-\frac{\hbar^2}{2}\frac{d}{dz}\left(\frac{1}{m^*(z)}\frac{d\Psi_i}{dz}\right)+U(z)\Psi_i=E_i\Psi_i, \quad i=1,2,3. \tag{27}$$

In contrast to the SUSYQM procedure described above, here we allow the effective mass to be position dependent, i.e., $m^*(z)=m(z)m_0$, where m_0 is the free-electron mass. This is because the problem turns out not to be analytically solvable anyway, even if m^* was constant, and allowing for the position dependent effectives mass makes the description more realistic, while introducing no serious complications in the model or in the procedure for solving it.

The maximization of Eq. (26) is a constrained maximization problem, the constraints being Eqs. (27). This is solved by introducing Lagrange multiplier functions $\lambda_i(z)$, e.g., Ref. 8, which recasts the problem into the unconstrained optimization of the new functional

$$J^*=J-\sum_{i=1}^{3}\int_{-z_L}^{z_R}\lambda_i(z)\left[\frac{\Psi_i''}{m}-\frac{m'}{m^2}\Psi_i'+b_i\Psi_i\right]dz=J-\sum_{i=1}^{3}J_i, \tag{28}$$

where $b_i(z)=(2m_0/\hbar^2)[E_i-U(z)]=q[E_i-U(z)]$ and the constant $q=2m_0/\hbar^2$ amounts to $0.2625\ \mathrm{eV}^{-1}\text{Å}^{-2}$. The conditions for the extremum of the functional J^*, Eq. (28), are found by equating its first-order variation δJ^* to zero, i.e.,

$$\delta J^*=\delta\left(K_1M_{12}+K_2M_{23}+K_3M_{31}-\sum_{i=1}^{3}J_i\right)=\delta\int_{-z_L}^{z_R}\Phi(z)dz=0, \tag{29}$$

where

$$K_1=(M_{23}^oM_{31}^o)/C,\qquad K_2=(M_{12}^oM_{31}^o)/C,\qquad K_3=(M_{12}^oM_{23}^o)/C. \tag{30}$$

The quantities M_{12}^o, M_{23}^o, and M_{31}^o are taken as constants when finding the variation because they are evaluated with fixed functions Ψ_i^o and $U^o(z)$ that optimize the functional J^*. The unknown functions y_i [in the conventional notation, here these are the three Ψ_i's and $U(z)$], which give an extremum to Eq. (28) are to be obtained by solving the system of Euler-Lagrange equations

$$\frac{\partial \Phi}{\partial y_i} - \frac{d}{dz}\left(\frac{\Phi}{\partial y_i'}\right) + \frac{d^2}{dz^2}\left(\frac{\partial \Phi}{\partial y_i''}\right) = 0, \tag{31}$$

where the function Φ, from Eqs. (28) and (29), may be written as

$$\Phi = K_1 \Psi_1 z \Psi_2 + K_2 \Psi_2 z \Psi_3 + K_3 \Psi_1 z \Psi_3 - \sum_{i=1}^{3} \lambda_i \left[\frac{\Psi_i''}{m} - \frac{m'}{m^2} \Psi_i' + b_i \Psi_i \right]. \tag{32}$$

The variation of J^* over λ_i's is always zero, by way of setting up Eq. (28). In the particular problem considered here, the system (31) becomes

$$\lambda_1'' - \frac{m'}{m}\lambda_1' + m b_1 \lambda_1 = mz(K_1 \Psi_2 + K_3 \Psi_3) = F_1(z),$$

$$\lambda_2'' - \frac{m'}{m}\lambda_2' + m b_2 \lambda_2 = mz(K_1 \Psi_1 + K_2 \Psi_3) = F_2(z),$$

$$\lambda_3'' - \frac{m'}{m}\lambda_3' + m b_3 \lambda_3 = mz(K_2 \Psi_2 + K_3 \Psi_1) = F_3(z),$$

$$G = q(\lambda_1 \Psi_1 + \lambda_2 \Psi_2 + \lambda_3 \Psi_3) - \frac{1}{mU}(\lambda_1' \Psi_1' + \lambda_2' \Psi_2' + \lambda_3' \Psi_3') = 0, \tag{33}$$

where account is taken of the fact that in ternary alloys of the type $A_x B_{1-x} C$, usually used for making graded QW's, the potential and the effective mass are not independent variables, but are related via $U(z) = [\Delta E_c / \Delta m] m(z) \equiv \theta m(z)$, where ΔE_c is the conduction band offset between the materials AC and BC and $\Delta m = m_{AC} - m_{BC}$ is the difference of effective masses in the two. The solutions for the Lagrange multiplier functions may be written as

$$\lambda_i = \left[C_{1i} - \int_{z_0}^{z} \frac{y_{2i}(z) F_i(z)}{W(z)} dz \right] y_{1i}(z) + \left[C_{2i} + \int_{z_0}^{z} \frac{y_{1i}(z) F_i(z)}{W(z)} dz \right] y_{2i}(z), \tag{34}$$

where y_{1i} and y_{2i} are the solutions of the homogeneous differential equations in λ_i's of the system (33), i.e., for $F_i(z) \equiv 0$, and $W(z)$ is the Wronskian. Given the fact that these homogeneous equations have the form of the Schrödinger equation, the two linearly independent solutions may be taken as $y_{1i}(z) = \Psi_i(z)$ and $y_{2i}(z) = \Psi_i(z)\{\tilde{C} + \int_{z_0}^{z} [m(z)/\Psi_i^2(z)] dz\}$, where $\tilde{C}$ is a constant and the Wronskian is simply $W(z) = m(z)$. The constants C_{1i} and C_{2i} are found from the requirement that the boundary conditions for the correction of the potential are satisfied, i.e., $G(-z_L) = G(z_R) = 0$ and $G'(-z_L) = G'(z_R) = 0$. Finding the optimal QW profile thus reduces to evaluating the potential function $U(z)$ that will make the quantity G equal to zero, while simultaneously satisfying all the equations from the systems (33) and (27). This system of coupled equations has no analytic solution, but may be solved iteratively according to the algorithm we describe next.

(1) We first choose an initial potential $U^{(i=0)}(z)$ on the segment $[-z_L, z_R]$ assumed constant outside it.

(2) The Schrödinger equation (27) is solved for the current form of the potential and the wave functions $\Psi_j^{(i)}$, observing the boundary conditions $\Psi_j^{(i)}(-z_L) = \Psi_j^{(i)}(z_R) = 0$ and $\Psi_j'^{(i)}(-z_L) = \Psi_j'^{(i)}(z_R) = 0$, are determined [here the superscript (i) is the iterations counter and the subscript j is the bound-state index].

(3) By substituting the functions Ψ_i into the system (33) and taking into account the boundary conditions for the Lagrange multiplier functions λ_i [i.e., $G(-z_L) = G(z_R) = 0$ and $G'(-z_L) = G'(z_R) = 0$], one finds the variation over the potential $G^{(i)}$.

(4) The steepest descent method is used to correct the potential

$$U^{(i+1)}(z) = U^{(i)}(z) - \alpha^{(i)} G^{(i)}, \tag{35}$$

where $\alpha^{(i)}$ is a constant chosen to provide the largest value of the functional J^* in the current iteration, i.e., such that $J^*(U - \alpha \partial J^*/\partial U) = J^*(U - \alpha G)$ is maximized.

(5) With the new potential $U^{(i+1)}$ one returns to step (2) and iterates until the satisfactory convergence is achieved, i.e., $J[U^{(i+1)}(z)] - J[U^{(i)}(z)] < \gamma$ (where the termination criterium γ is a small positive number).

The results of this iterative procedure should not depend on the choice of the initial potential. However, executing it for a few different initial potentials soon reveals that the procedure is easily trapped in the nearest local extremum as a consequence of using the gradient type formula in step (4). To avoid such an unfavorable outcome, one should obviously start with a potential that is sufficiently close to the final, globally optimal potential. The problems with local extrema are probably not specific to the target functional used here (that for the second-order susceptibility). Indeed, the initial potential and the one optimized for its transmission properties in Ref. 8 are rather close to each other.

In this work the preparation of the initial QW profile was performed by the simulated annealing (SA) method. This is an iterative stochastic optimization technique, nowadays in widespread use, with readily available algorithms. While always accepting a step (random change of values of variables) that improves the target function, it also sometimes accepts steps degrading the target function and thus avoids being trapped in local extrema. This is essential for the problem we consider, which appears to be of highly multivalley type. The SA is here applied to multilayer QW's (i.e., stepwise constant potentials) with some reasonably small number of layers, of the order of ten. Each layer brings in two independent variables—its width and potential height, and an additional free variable is the height of ''outer'' barriers. The variables may vary within some limits imposed physically or technologically. We have used the SA algorithm[10] that can handle box-type constraints. The state energies, wave functions, and

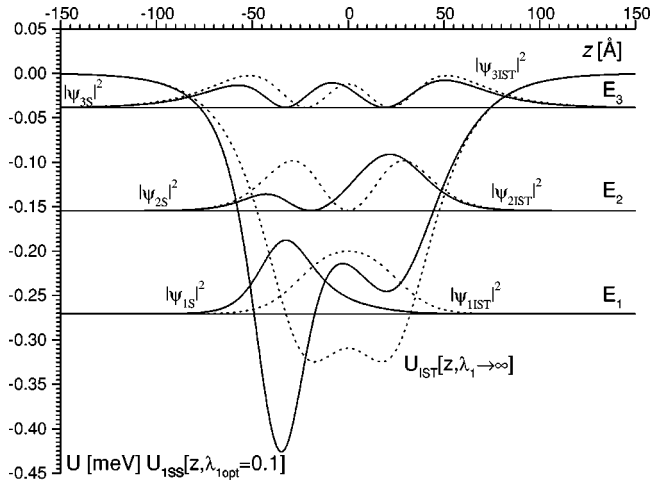

FIG. 1. The single-parameter optimized potential $U_{1\,SS}(z,\lambda_1)$ with the corresponding wave functions $\psi_{i\,S}$ (solid lines) and the initial symmetric $U_{IS}(z)$ with its wave functions $\psi_{i\,IS}(z)$ (dotted lines).

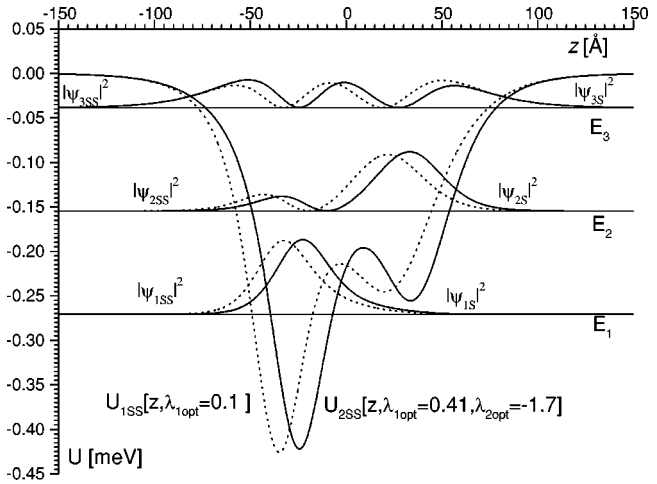

FIG. 2. The two-parameter optimized potential $U_{2\,SS}(z,\lambda_1,\lambda_2)$ with the corresponding wave functions $\psi_{i\,SS}(z,\lambda_1,\lambda_2)$ (solid lines) and the single-parameter optimized potential $U_{1\,SS}(z,\lambda_1)$ with the corresponding wave functions $\psi_{i\,S}(z,\lambda_1)$ (dotted lines).

matrix elements, i.e., the target function (26) for this type of structures may be conveniently evaluated using the transfer matrix method, and SA will vary the discrete set of layers parameters until it finds the globally optimal structure with prescribed total number of layers.

III. NUMERICAL RESULTS AND DISCUSSION

A. Multiparameter supersymmetric optimization

Here we consider a GaAs based QW ($m^*=0.067$) with three bound states. The Pöschl-Teller potential parameters are taken to be $\alpha=0.266$ nm^{-1} and $U_0=0.464$ eV, so that the spacing between the second and third state amounts to 116 meV. The state energies in this well are $E_{11}=-348$ meV, $E_2=-154.7$ meV, and $E_3=-38.7$ meV. In order to set the spacing between the lowest-two states to 116 meV, the potential $U_{IST}(z)$ was introduced so as to shift the ground state by $\epsilon=U_0/6$, i.e., 77.3 meV, its new position being $E_1=E_{11}+\epsilon$. To find its actual shape we first solve Eq. (10) and find the function (11), then find the Wronskian (9), and then the potential $U_{IST}(z)$ itself [Eq. (8)], and the corresponding wave functions (16), (17), and (18). Since $U_{IST}(z)$ is even, the matrix element $M_{13}=0$ and hence $\chi^{(2)}=0$ for this well despite the double resonance. However, this potential is then asymmetrized by SUSYQM transform [Eq. (19)], i.e., by deleting and then restoring the bound state at E_1. In this procedure we get a single-parameter-dependent potential $U_{1\,SS}(z,\lambda_1)$, which is fully isospectral with $U_{IS}(z)$ and coincides with it for $\lambda_1\to\pm\infty$. For any finite λ_1, however, $U_{1\,SS}$ is asymmetric, and $\Pi^{(2)}$ will generally be nonzero acquiring a maximum at some optimal value of λ_1. With the initial potential being symmetric, λ_1 may be varied over positive values only, because $U_{1\,SS}(z,\lambda_1)=U_{1\,SS}[z,-(1+\lambda_1)]$, and we find that the maximum occurs at $\lambda_1=0.1$ and amounts to $\Pi^{(2)}(\lambda_1)=3472$ Å^3, the individual matrix elements being $M_{12}=17$ Å, $M_{13}=7.65$ Å, and $M_{23}=26.7$ Å. In Fig. 1 we give the shape of $U_{1\,SS}(z,\lambda_1=0.1)$, together with the symmetric $U_{IS}(z)$ and the wave functions in both systems. The shift of wave functions, related to the potential asymmetrization, is clearly visible. The maximum of the matrix elements product $\Pi^{(2)}$ occurs due to opposite trends in M_{13} on one side and in M_{12} and M_{23} on the other, as λ_1 varies.

Next, by deleting and then restoring the bound state at E_2, we get the two-parameter potential $U_{2\,SS}(z,\lambda_1,\lambda_2)$ isospectral to $U_{1\,SS}(z,\lambda_1)$ and $U_{IS}(z)$. By varying both parameters we now find the maximum of $\Pi^{(2)}(\lambda_1=0.41,\ \lambda_2=-1.7)=3844$ Å^3 (note that λ_2 was given both negative and positive values because the initial potential in the second transform was no longer symmetric). This maximal $\Pi^{(2)}$ exceeds by approximately 10% the value obtained in the single-parameter optimization, with the individual matrix elements now being $M_{12}=16.5$ Å, $M_{13}=8.41$ Å, and $M_{23}=27.7$ Å. In Fig. 2 we give the optimized single-parameter and two-parameter potentials. The resulting dependence of $\Pi^{(2)}$ on λ_1 together with the corresponding dependence in the single-parameter case is given in Fig. 3.

We have also introduced the third free parameter λ_3 via deleting and restoring the bound state at E_2. However, in contrast to a practically significant increase of $\Pi^{(2)}$ when introducing the second free parameter, introducing the third one did not result in any further improvement of the maximal value of $\Pi^{(2)}$. Such behavior can be explained by tracing the influence of individual parameters on the shapes of transformed potentials. This may be inferred from Figs. 1, 2, and 4, and of course, by inspecting a number of intermediate cases. Take the parameter λ_1, introduced when deleting and restoring E_2, to tend from large positive values towards zero (the situation when λ_1 tends from large negative values to -1 is completely analogous, essentially just reversed in space). From the initial potential in this particular transform there ''separates'' a part that becomes increasingly confining as λ_2 gets smaller, but with the tendency of saturation (i.e.,

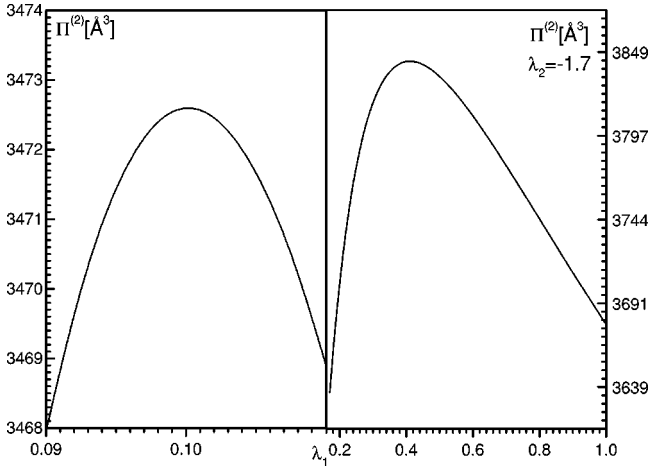

FIG. 3. The matrix elements product $\Pi^{(2)}$ vs λ_1 dependence in one- or two-parameter cases (left and right, respectively).

shape stabilization) as the wave function at E_1 becomes mostly confined by this part of the potential. An increased localization of this wave function generally decreases its overlap with other wave functions and may thus decrease the dipole matrix elements M_{2j}, but this is more than compensated by the beneficial influence of asymmetrization on the product of matrix elements, so there appears an optimum. The story repeats when introducing the second parameter λ_2, there still appears an optimum but the optimal pair (λ_1,λ_2) is now more remote from their limiting small values than was the case with the single parameter λ_1. That is, there still is some benefit from changing the shapes of wave functions, but the system avoids strong localization that would diminish $\Pi^{(2)}$. When introducing the third parameter λ_3, which tends to mostly affect (localize) the state at E_3, Fig. 4, it turns out that any amount of localization, induced when λ_3 becomes finite, actually degrades $\Pi^{(2)}$ to an extent that cannot be ''repaired'' by changes in the wave function shapes. We

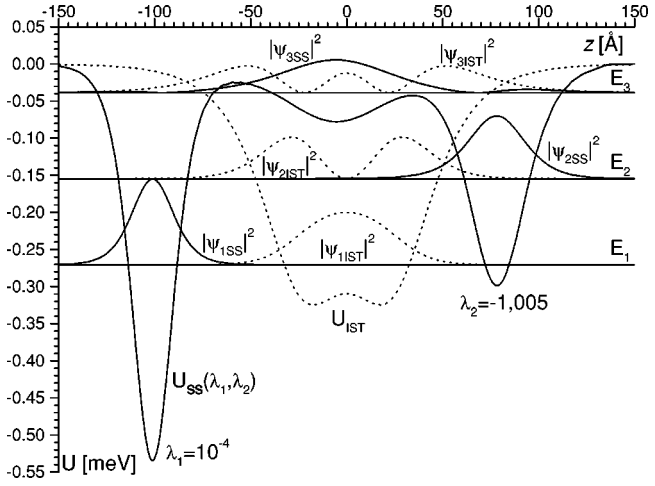

FIG. 4. The two-parameter potential U_{SS} obtained by the SUSYQM transform of U_{IS} via single deletion and restoring of bound states at E_1 and E_2, once each with the parameters $\lambda_1 = 10^{-4}$ and $\lambda_2 = -1.005$.

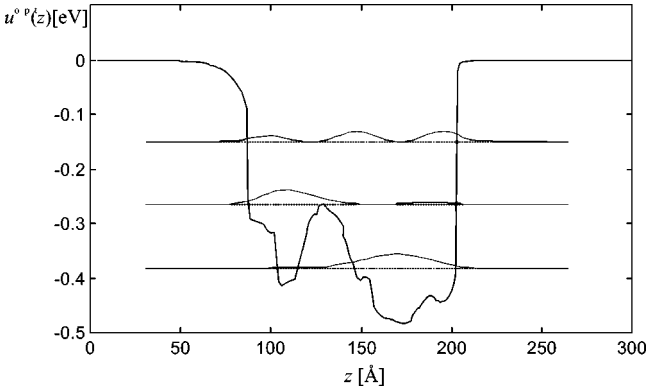

FIG. 5. The variationally optimized potential, providing the largest value of $\chi^{(2)}$ (started with a four-layer stepwise-constant SA preoptimization). Also displayed are the relevant bound state wave functions.

have also checked that changing the order of factorization states does not make the last introduced parameter useful.

The alternative way of introducing more free parameters by repeatedly deleting and restoring the same state also did not give any improvement in maximal $\Pi^{(2)}$, the overall effect of parameters obtained that way being the same as that of a single parameter.

We find, therefore, that for the shape optimization of a QW with three bound states, all three being relevant for SHG, it suffices to generate the two-parameter family of isospectral potentials. Somewhat better results might be expected in a QW having more bound states (but only three of them being relevant for the resonant SHG). Introducing new parameters by factorization of the existing irrelevant (''upper'') states would then allow the relevant wave functions to change shape to some extent without becoming too localized. However, this approach would require deeper wells to support an increased number of bound states, which may be impractical or even impossible in a given alloy system. In the case considered here, the maximal $\Pi^{(2)}$ we obtained is by approximation 25% larger than the maximal value obtainable in a single-step asymmetric QW ($\Pi^{(2)}_{max} = 3090$ Å^3, Ref. 4). Certainly, having obtained the optimal QW profile from this two-parameter method, one may proceed to generate the more realistic $Al_xGa_{1-x}As$ QW profile, where the effective mass varies along with the potential.

B. Optimization by variational calculus

Within the variational approach we again consider an $Al_xGa_{1-x}As$ alloy based QW. In course of the $\chi^{(2)}$ optimization we have set $n=4$ and $\Theta = 1$ meV in the target functional (26). In the first phase we used the SA based preoptimization to obtain a good starting point for the variational method. This was done for a few structures with different number of fixed composition layers. The material dependence of parameters is now taken into account with[11] $E_{g_{\mathrm{GaAs}}} = 1.42$ eV, $E_{g_{\mathrm{AlAs}}} = 2.67$ eV, $m_{\mathrm{GaAs}} = 0.067\ m_0$, and $m_{\mathrm{AlAs}} = 0.15\ m_0$ and the conduction band offset between GaAs and AlAs, $\Delta E_c = 0.75$ eV and Vegard law was used for the alloys.

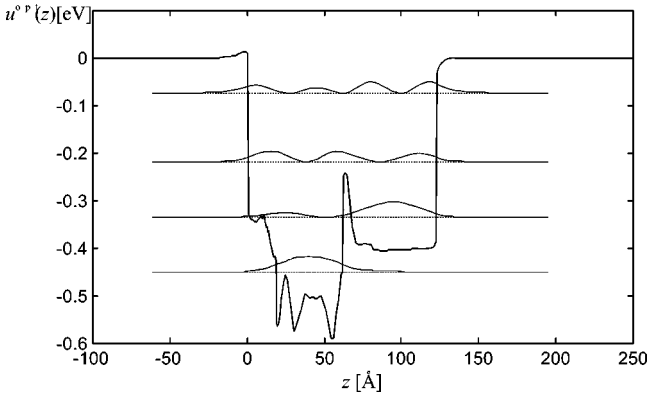

FIG. 6. Same as Fig. 5, but starting with an eight-layer stepwise-constant SA preoptimization.

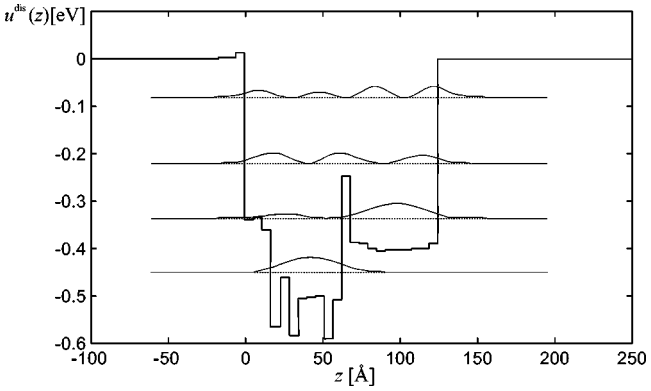

FIG. 7. The discretized, step-graded version of the optimal potential from Fig. 6 with two crystalline monolayers wide steps.

In one run we started with a structure with just four ''inner'' layers, i.e., with nine free parameters (the width and the composition for each inner layer and the composition of the outer barriers, the latter taken to be the same on either side). The minimal width of any one layer was limited to 5 Å and the maximal value of Al mole fraction to $x_{max}=0.6$. The QW optimized this way has three bound states with energies $E_1=71.5$ meV, $E_2=187.5$ meV, and $E_3=303.5$ meV and the matrix elements amount to $M_{12}=-16.7$ Å, $M_{23}=-25.7$ Å, and $M_{31}=9.1$ Å, their product being $|\Pi^{(2)}|=3906$ Å^3, clearly making this QW a good starting point for the variational procedure. After 142 iterations it has satisfactorily converged to a smooth potential $u^{opt}(z)$ (and the accompanying effective mass $m^{opt}(z)=[u^{opt}(z)+V_0]/\theta$ with $V_0=1.078$ eV and $\theta=9.036$ eV), displayed in Fig. 5. The bound state energies were now $E_1=-381.7$ meV, $E_2=-265.7$ meV, and $E_3=-149.7$ meV (with the outer barriers potential taken as reference zero), while the matrix elements product amount to $|\Pi^{(2)}|=4328$ Å^3 (individually, $M_{12}=-15.8$ Å, $M_{23}=24.9$ Å, and $M_{31}=-11.0$ Å). One can note a significant difference between the initial (SA preoptimized) and the final (variationally optimized) potentials, as a consequence of a rather coarse initial potential.

In another run we started with the structure having eight inner layers, i.e., the SA preoptimization was performed in the 17-parameter space and the search space was further extended by limiting x_{max} to 0.73. While this has drastically increased the computation time, the optimized QW found in this step was considerably better, having the product of matrix elements $|\Pi^{(2)}|=4114$ Å^3 (individually, $M_{12}=-16.9$ Å, $M_{23}=-25.9$ Å, and $M_{31}=-9.4$ Å) and the bound state energies $E_1=76$ meV, $E_2=192$ meV, $E_3=308$ meV, and $E_4=449.2$ meV (it has four bound states but the fourth one is not relevant for the present target). With this better initial potential the variational procedure needed just 57 iterations for convergence. The final optimized potential is displayed in Fig. 6 and the accompanying effective mass is evaluated as $m^{opt}(z)=[u^{opt}(z)+1.194\ \text{eV}]/\theta$. This design has $|\Pi^{(2)}|=4685$ Å^3 (individually, $M_{12}=-16.9$ Å, $M_{23}=-26.4$ Å, and $M_{31}=-10.5$ Å) and bound states at $E_1=-449.5$ meV, $E_2=-333.5$ meV, $E_3=-217.5$ meV, and $E_4=-73.1$ meV.

Certainly, the strictly smooth profile cannot be realized, the best approximation to it could be the profile discretized into one crystalline monolayer thin layers of fixed composition. The practicalities of fabrication may require even coarser discretization. This will slightly degrade the results obtained, more so if coarser discretization is employed. However, if one is willing to accept, e.g., two-monolayer wide steps (2×2.83 Å), the degradation in performance is rather small: the structure displayed in Fig. 7 has $|\Pi^{(2)}|=4507$ Å^3 (individually, $M_{12}=-16.7$ Å, $M_{23}=-26.2$ Å, and $M_{31}=10.3$ Å) and bound states at $E_1=-451.4$ meV, $E_2=-335.7$ meV, $E_3=-220.2$ meV and $E_4=-81.5$ meV, all of them quite close to the corresponding values in case of smooth potential. Clearly, the discretized potential could be obtained by SA with no variational procedure, but this would have been very time consuming because of the large number of free variables (step widths and heights) involved. It is only the combined use of SA and the variational method that leads to efficient design of optimal QW profile.

By comparing the results obtained here and those stated elsewhere, e.g., $|\Pi^{(2)}|=3090$ Å^3 for asymmetric step QW,[4] $|\Pi^{(2)}|\approx3300$ Å^3 obtained by SUSYQM,[12] or $|\Pi^{(2)}|=3910$ Å^3 obtained by IST and coordinate transform method,[13] we find the improvement by at least 15% over the previous results (all the data apply for the 116 meV, i.e., the CO_2 laser incident radiation).

IV. CONCLUSION

We have proposed and analyzed two methods of QW profile optimization to achieve maximal nonlinear susceptibilities. One method relies on multiparameter SUSYQM as a generalization of the previously developed single-parameter approach. It starts with a somewhat arbitrary potential, uses IST to bring its bound states to the required positions, and then the SUSYQM to vary the potential isospectrally while searching the free-parameters space. In particular, the two-parameter procedure gave $\approx10\%$ improvement of the results of single-parameter procedure, but the three-parameter procedure gave no further improvement.

The other method relies on the combined use of simulated annealing and the variational calculus and effectively performs a free variation of the QW profile, searching for the

global optimum. The results obtained with it are better by $\approx 20\%$ than the results obtained by the first method.

Both methods considered here improve the best achievements reported in the literature and both have their merits and disadvantages. The SUSYQM method may allow analytic work to a large extent and is faster, while the SA/variational method gives better final results though it is more demanding computationally.

*Email address: milanovic@kiklop.etf.bg.ac.yu

[1] F. Cooper, A. Khare, and U. Sukhatme, Phys. Rep. **251**, 267 (1995).
[2] S. Tomić, V. Milanović, and Z. Ikonić, Phys. Rev. B **56**, 1033 (1997).
[3] V. Milanović and Z. Ikonić, Solid State Commun. **104**, 445 (1997).
[4] D. Indjin, Z. Ikonić, V. Milanović, and J. Radovanović, IEEE J. Quantum Electron. **34**, 795 (1998).
[5] E. Rosencher and Ph. Bois, Phys. Rev. B **44**, 11 315 (1991).
[6] B. Y. Tang, Solid State Commun. **104**, 679 (1998).
[7] B. Y. Tang and N. Kiruischeva, Phys. Lett. A **229**, 49 (1997).
[8] P. Gross, V. Ramakrishna, E. Vilallonga, H. Rabitz, M. Littman, S. A. Lyon, and M. Shayegan, Phys. Rev. B **49**, 11 100 (1994).
[9] S. Flügge, *Practical Quantum Mechanics* (Springer-Verlag, Berlin, 1974).
[10] http://www.netlib.org/opt/simann.f
[11] G. Bastard, *Wave Mechanics Applied to Semiconductor Heterostructure* (Les Editions de Physique, Les Ulis, France, 1990).
[12] S. Tomić, V. Milanović, and Z. Ikonić, Opt. Commun. **143**, 214 (1997).
[13] S. Tomić, V. Milanović, and Z. Ikonić, J. Phys.: Condens. Matter **10**, 6523 (1998).