

# Intersubband Nonlinear Optical Properties in Semiconductor Quantum Wells Optimized by Supersymmetric Quantum Mechanics

Stanko Tomić <sup>†</sup>, Vitomir Milanović and Zoran Ikonić

<sup>†</sup> *Laboratory of Physics (010), VINČA Institute of Nuclear Sciences,  
P.O. Box 522, 11001 Belgrade, Yugoslavia  
Faculty of Electrical Engineering, University of Belgrade,  
Bulevar Revolucije 73, 11000 Belgrade, Yugoslavia*

A systematic procedure is proposed for the design of ternary alloys based quantum well structures optimized for resonant second order nonlinear optical effects. The method relies on the supersymmetric quantum mechanics as derived here for the case of position-dependent effective mass. Starting from a symmetric, truncated quasi-parabolic potential, itself lacking any second-order nonlinearity, we generate a family of asymmetric potentials, fully isospectral with the starting potential, and choose the one which maximizes the product of transition matrix elements relevant for the second order nonlinearity. Realization of the optimized potential (in an approximate manner) by grading a ternary alloy (e.g.  $Al_xGa_{1-x}As$ ) with position dependent effective mass, is then described. The best value of nonlinear susceptibilities obtained exceeds those reported in the literature.

## I Introduction

There is currently a considerable research interest in optical phenomena based on intersubband transitions in semiconductor quantum wells (QW). The transition dipole matrix elements are of the order of QW width, i.e., a few nanometers, grossly exceeding those of atoms and molecules. Due to generally large values of dipole transition matrix elements and the possibility of achieving the resonance conditions, both the linear and, even more so, nonlinear optical processes in these structures are very intense. Considering the second-order nonlinear phenomena, the most prominent examples of which are the second harmonic generation (SHG) and optical rectification, these are significant only in noncentrosymmetric media [1], implying that one should design asymmetric QW structures for these types of nonlinearities. Their resonant enhancement is obtained by making the levels spacing (about) equal to the pump photon energy. The commonly used method to realize asymmetric QW's is to employ the alloy composition grading in a continuous, eg. Ref. [2], or suitable stepwise-constant,

eg. Ref. [3], manner. Refinement of these structures resulted in an increase of  $\chi^{(2)}$  from the initial estimate of 10–100 to 1900 times larger than that of bulk GaAs [3]. An alternative route is to bias the initially symmetric structure by an external electric field [4, 5]. Designing asymmetric QW's which provide the resonance conditions and at the same time offer large values of relevant matrix elements has largely relied on qualitative considerations, physical intuition, and trial-and-error method. In brief, the structure parameters are varied, using the results for the idealized model as a guideline, so as to keep the levels spacing at the desired value, and checks whether the product of relevant matrix elements is large than obtained previously. Keeping the levels in the real structure equidistant clearly requires a nontrivial numerical effort.

We have recently proposed [6] a systematic procedure for the design of optimized asymmetric QW's for resonant SHG. It used the supersymmetric quantum mechanics (SUSYQM) approach, enabling one to generate a family of smooth asymmetric potentials isospectral to some original potential. This family depended, in the simplest case, on a single parameter  $\lambda$  which was varied to get largest product of relevant matrix elements, while the levels equidistance was automatically guaranteed throughout this variation. The optimized nonlinearity obtained that way was better than has been previously reported in the literature, but there was a drawback from the technological viewpoint in that the effective mass had to be kept constant throughout the structure, and hence graded quaternary alloys would have to be used for its fabrication.

In this paper we extend the method to make it applicable to design of optimized structures based on graded ternary alloys, where the effective mass is necessarily position dependent, just as is the potential. Using the technologically well studied, strain-free material like  $Al_xGa_{1-x}As$  for fabrication the optimized QW's is highly desirable, and is here demonstrated to be possible.

## II Theoretical considerations

### II.1 Nonlinear optical susceptibilities

Conversion of the pump field to the second harmonic may be described by second-order susceptibility,  $\chi_{\omega_3}^{(2)}$ , which relates the polarization at  $\omega_3 = \omega_1 \pm \omega_2$  to the radiation electric field  $\mathbf{E} = \mathbf{E}_1 e^{i\omega_1 t} + \mathbf{E}_2 e^{i\omega_2 t} + c.c.$ , squared:

$$\mathbf{P}(t) = \varepsilon_0 \chi^{(1)} \mathbf{E} + \varepsilon_0 \chi^{(2)} \mathbf{E}^2 + \dots \quad (1)$$

The system evolution may be found from the Liouville-von Neumann equation:

$$\frac{\partial \rho_{ij}(t)}{\partial t} = -\frac{i}{\hbar} [\hat{H}, \rho(t)]_{ij} - \gamma_{ij} (\rho(t) - \rho^{(0)})_{ij} \quad (2)$$

where  $\rho_{ij}^{(0)} = \rho_{ii} \delta_{ij}$  are the thermic equilibrium values of the density matrix elements, and  $\gamma_{ij}$  the relaxation rates. The full Hamiltonian  $\hat{H}$  is the sum of unperturbed  $\hat{H}_0$  and interaction  $\hat{H}^{(1)} = -\hat{\mu} \mathbf{E}(t)$  parts, where  $\hat{\mu}$  denotes the dipole moment operator. The electronic polarization of the  $n$ th order is given by

$$P_\gamma^{(n)} = \frac{1}{S} \text{Tr}(\rho^{(n)} \mu_\gamma) \quad (3)$$

where  $S$  is the area of interaction. In the special case  $\omega_1 = \omega_2 = \omega$ ,  $\omega_3 = 2\omega$ , the accurate-to-first-order solution (2) for a system with three (important) states gives the expression for the second-order susceptibility

$$\chi_{2\omega}^{(2)} = \frac{e^3}{\varepsilon_0 \hbar^2 V} \frac{(\mu_\alpha)_{12} (\mu_\beta)_{23} (\mu_\gamma)_{31}}{2\omega - \omega_{31} - i, 13} \left[ \frac{(\rho_{22} - \rho_{11})}{\omega - \omega_{21} - i, 12} - \frac{(\rho_{33} - \rho_{22})}{\omega - \omega_{32} - i, 23} \right] \quad (4)$$

where  $\omega_{ij} = (E_i - E_j)/\hbar$  transition frequency between states  $i$  and  $j$  and  $\mu_{ij} = \langle i | z | j \rangle$  are the transition dipole moments (all fields are taken to be polarized along the  $z$  direction, i.e.,  $\alpha = \beta = \gamma = z$ , the only

case of importance for intersubband transitions in  $n$ -doped direct semiconductor based QW's,  $z$  being perpendicular to the QW plane). Furthermore,  $V = SL_z$ , where  $L_z$  denotes the QW width. To obtain the maximum value of  $\chi_{2\omega}^{(2)}$  one should meet the double resonance conditions, i.e.,  $\omega_{21} = \omega_{32} = \omega$ . The second-order susceptibility then becomes

$$\chi_{2\omega \max}^{(2)} = \frac{e^3(\rho_{11} - \rho_{22})\mu_{12}\mu_{23}\mu_{31}}{L_z \epsilon_0 (\hbar, \gamma_2)^2} \quad (5)$$

where  $\rho_{11}$  denotes the electron density per unit well surface,  $\gamma_2$  the off-diagonal relaxation rate (assumed common for all transitions), and the population of states 2 and 3 is ignored, which is justified in situations not much departing from thermal equilibrium. This is the expression we want to maximize via appropriate tailoring of the potential, and hence the matrix elements  $\mu_{ij}$ .

Density matrix considerations [3, 7] of a two-level system in the presence of close-to-resonance light (ie. under conditions when just two levels are important) give the expression for optical rectification coefficient  $\chi_0^{(2)}$ , which takes the largest value at exact resonance,  $\hbar\omega = E_2 - E_1 = \hbar\omega_{21}$ , and then amounts to

$$\chi_{0 \max}^{(2)} = 2 \frac{e^3 T_1 T_2}{\epsilon_0 \hbar^2} (\rho_{11} - \rho_{22}) \mu_{12}^2 \delta_{12} \quad (6)$$

where  $e$  is the electron charge,  $\rho_{11}$  and  $\rho_{22}$  are the electron surface densities of the lower  $|1\rangle$  and the upper  $|2\rangle$  state,  $\mu_{12} = |\langle 1|z|2\rangle|$  is the transition dipole moment and  $\delta_{12} = |\langle 1|z|1\rangle - \langle 2|z|2\rangle|$  is the difference of permanent dipole moments, ie. the mean electron displacement of the transition. Furthermore,  $T_2$  is the off-diagonal relaxation time in the Liouville equation, related to the linewidth, and  $T_1$  is the diagonal relaxation time, ie. the excited state lifetime.

The value of  $\chi_{0 \max}^{(2)}$  clearly increases with increasing  $T_1$  and  $T_2$ . The first of these may be significantly increased by introducing a third, optically inactive metastable state, its wave function being spatially displaced from that of the ground state [8], the role of which is the long-life charge storage of excited electrons. By increasing  $\chi_{0 \max}^{(2)}$  this way, however, one sacrifices the operating speed of the device. On the other hand  $T_2$ , which depends on various scattering mechanisms, is eventually limited from above by electron-phonon scattering and cannot be significantly enhanced by "band structure engineering" [9]. Finally, there remains the product of matrix elements  $\mu_{12}^2 \delta_{12}$  which may be varied by suitable profiling of the QW potential, in course of which one should not perturb the resonance condition  $\omega = \omega_{21}$ . Indeed, finding

It should be noted that under strong pump conditions  $\chi^{(2)}$  alone may not be enough to describe the conversion process, since e.g. the pump power itself and the diagonal relaxation time also play significant roles. However, the target function to be optimized cannot then simply be defined analytically, and, while this case-sensitive optimization may also be performed by the method described below, here we restrict to low or intermediate pump powers, i.e., to expressions (5) and (6).

## II.2 Supersymmetric quantum mechanics with the position-dependent effective mass

In this section we consider the extension of the supersymmetric transition to construct isospectral Hamiltonians of the position-dependent effective mass [10](a comprehensive review of the constant mass supersymmetric quantum mechanics is given in Ref. [11]). The 1D Schrödinger equation, describing the electron motion within the effective mass approximation reads

$$\hat{H}_0 \psi_0^{(i)}(z) = -\frac{\hbar^2}{2} \frac{d}{dz} \left( \frac{1}{m(z)} \frac{d\psi_0^{(i)}(z)}{dz} \right) + U_0(z) \psi_0^{(i)}(z) = E_0^{(i)} \psi_0^{(i)}(z) \quad (7)$$

where  $U_0(z)$  and  $m(z)$  denote the position dependent potential and the effective mass, and  $\psi_0^{(i)}$  and  $E_0^{(i)}$  are the eigenfunction and energy of  $i$ -th state ( $i=1,2,3,\dots$ ). The Hamiltonian  $\hat{H}_0$  can be factorized

as

$$\hat{H}_0 = \hat{A}^+ \hat{A}^- + \varepsilon \quad (8)$$

where  $\hat{A}^+$  and  $\hat{A}^-$  are mutually adjoint. The factorization energy  $\varepsilon$  may generally take any arbitrary value (i.e., may or may not coincide with an eigenenergy  $E_0^{(i)}$ ). The operators  $\hat{A}^+$  and  $\hat{A}^-$  may be written as

$$\hat{A}^+ = \frac{d}{dz} \frac{\hbar}{\sqrt{2m(z)}} + W(z) \quad \text{and} \quad \hat{A}^- = -\frac{\hbar}{\sqrt{2m(z)}} \frac{d}{dz} + W(z) \quad (9)$$

where  $W(z)$  is the superpotential. Choosing  $\varepsilon$  to coincide with one of eigenenergies  $E_0^{(k)}$  results in the Riccati equation

$$\frac{d}{dz} \left( \frac{\hbar}{\sqrt{2m(z)}} W(z) \right) + W^2(z) = \frac{1}{\psi_0^{(k)}(z)} \frac{d}{dz} \left( \frac{\hbar^2}{2m(z)} \frac{d\psi_0^{(k)}(z)}{dz} \right) \quad (10)$$

the solution of which is the superpotential

$$W(z) = \frac{\hbar}{\sqrt{2m(z)}} \frac{d}{dz} \left[ \ln \psi_0^{(k)}(z) \right] \quad (11)$$

Next we construct a new Hamiltonian

$$\hat{H}_1 = -\frac{\hbar^2}{2} \frac{d}{dz} \left( \frac{1}{m(z)} \frac{d}{dz} \right) + U_1(z) = \hat{A}^- \hat{A}^+ + \varepsilon \quad (12)$$

with  $\varepsilon = E_0^{(k)}$ , as above. Multiplying the equation  $\hat{A}^+ \hat{A}^- \psi_0^{(i)} = (E_0^{(i)} - E_0^{(k)}) \psi_0^{(i)}$  by  $\hat{A}^-$  from the left we find that  $\hat{H}_1$  has the identical eigenspectrum as  $\hat{H}_0$ , except for the eigenstate  $E_0^{(k)}$  of  $\hat{H}_0$ , and the eigenfunctions  $\psi_1^{(i)}$  of  $\hat{H}_1$  may be expressed through  $\psi_0^{(i)}$  as

$$\psi_1^{(i)}(z) = \frac{1}{\sqrt{E_0^{(i)} - E_0^{(k)}}} \hat{A}^- \psi_0^{(i)}(z) \quad i \neq k \quad (13)$$

It follows from Eq. (13) that the function  $\psi_1^{(k)}$  cannot be normalized, since  $E_0^{(k)}$  is not an eigenenergy of  $\hat{H}_1$ , while all other  $\psi_1^{(i)}$  can and are therefore physically acceptable. All the states of  $\hat{H}_0$  (with the exception of the factorization state  $E_0^{(k)}$ ) thus have their  $\hat{H}_1$  counterparts, and the two Hamiltonians are "almost" isospectral. Due to the same factorization energy, the potential in the new Hamiltonian is given by

$$U_1(z) - U_0(z) = \hat{A}^- \hat{A}^+ - \hat{A}^+ \hat{A}^- = [\hat{A}^-, \hat{A}^+] \quad (14)$$

i.e.,

$$U_1(z) = U_0(z) - 2 \frac{\hbar}{\sqrt{2m(z)}} \frac{dW(z)}{dz} - \frac{\hbar^2}{2} \frac{1}{\sqrt{m(z)}} \frac{d^2}{dz^2} \frac{1}{\sqrt{m(z)}} \quad (15)$$

To find the supersymmetric partner potential  $U_{ss}(z)$  which will make a Hamiltonian fully isospectral to  $\hat{H}_0$  (with  $U_0(z)$ ), we start from the potential  $U_1(z)$  with identical  $\varepsilon = E_0^{(i)}$ . The eigenfunction  $\psi_1^{(k)}$ , Eq. (13), corresponding to  $E_0^{(k)}$  cannot be normalized, but the general solution of the Schrödinger equation with the potential  $U_1(z)$ , at  $E = E_0^{(k)}$  is given by

$$\psi_1^{(k)}(z) = \frac{\sqrt{2m(z)}}{\hbar} \frac{1}{\psi_0^{(k)}(z)} [\lambda + I(z)] \quad I(z) = \int_{-\infty}^z \psi_0^{(k)}(t)^2 dt \quad (16)$$

where  $\lambda$  is an integration constant. Using the procedure described above we find the proper, nonnormalizable eigenfunction corresponding to  $E_0^{(k)}$  as

$$\psi_{ss}^{(k)}(z) = \frac{\sqrt{2m(z)}}{\hbar} \frac{1}{\psi_1^{(k)}(z)} = \frac{\psi_0^{(k)}(z)}{\lambda + I(z)} \quad (17)$$

The normalizability of  $\psi_{ss}^{(k)}$  is provided by suitable choice of the parameter  $\lambda$ : any value outside the interval  $(-1,0)$  is acceptable. This is the eigenfunction corresponding to the eigenvalue  $E_0^{(k)}$  of the Hamiltonian  $\hat{H}_{ss}$ , with the potential

$$U_{ss}(z) = U_1(z) - 2 \frac{\hbar}{\sqrt{2m(z)}} \frac{dW_1(z)}{dz} - \frac{\hbar^2}{2} \frac{1}{\sqrt{m(z)}} \frac{d^2}{dz^2} \frac{1}{\sqrt{m(z)}} \quad (18)$$

with the superpotential  $W_1(z)$  given by

$$W_1(z) = \frac{\hbar}{\sqrt{2m(z)}} \frac{d}{dz} \left[ \ln \psi_1^{(k)}(z) \right] = - \frac{\hbar}{\sqrt{2m(z)}} \frac{d}{dz} \left[ \ln \left( \frac{\hbar}{\sqrt{2m(z)}} \frac{\psi_0^{(k)}(z)}{\lambda + I(z)} \right) \right] \quad (19)$$

For states other than  $k$  the eigenfunctions of  $\hat{H}_{ss}$  read

$$\psi_{ss}^{(i)}(z) = \frac{1}{E_0^{(i)} - E_0^{(k)}} \hat{A}_1^- \left[ \hat{A}^- \psi_0^{(i)}(z) \right] \quad i \neq k \quad (20)$$

where  $\hat{A}_1^-$  has the same form as  $\hat{A}^-$  Eq. (9), but with  $W(z) \rightarrow W_1(z)$ . Thus, the Hamiltonian  $\hat{H}_{ss}$ , the kinetic energy part of which is unchanged from the original  $\hat{H}_0$ , Eq. (7), and the potential given by (18), is fully isospectral to the original Hamiltonian  $\hat{H}_0$ . It is important to note that  $\hat{H}_{ss}(\lambda)$  is generally asymmetric with respect to the coordinate  $z$ , i.e., its eigenfunctions do not have definite parity even if  $\hat{H}_0$  itself is symmetric. The system described by  $\hat{H}_{ss}$  therefore allows for SHG. Furthermore, while  $W(z)$ ,  $U_1(z)$  and  $\psi_1^{(i)}(z)$  may have singularities,  $\hat{H}_{ss}$  is strictly regular, and important feature for realization of  $\hat{H}_{ss}$  in semiconductor QW. Finally,  $\hat{H}_{ss}$  is continuously variable through the choice of the scalar parameter  $\lambda$ , but it also depends on the choice of the factorization energy  $\varepsilon = E_0^{(k)}$ . Should one choose some other value,  $\varepsilon = E_0^{(j)}$ , all the expressions remain valid upon substitution  $\psi_0^{(k)}(z) \rightarrow \psi_0^{(j)}(z)$ .

### III Supersymmetric optimization of quantum wells based upon ternary alloys

As noted in the Introduction, we aim at maximizing the second-order susceptibility at double resonance, i.e., the product of matrix elements  $\Pi^{(2)} = \mu_{12}\mu_{23}\mu_{31}$ . These clearly depend on the potential profile, which should be varied to obtain  $\Pi^{(2)}$  as large as possible while keeping the levels spacing constant. We perform this variation in a systematic manner, by varying a scalar parameter  $\lambda$ , which guarantees that all eigenenergies will be preserved while the potential, and hence the eigenfunctions and the dipole matrix elements, experience gross changes. In any less systematic procedure keeping the levels equidistant is certainly a more laborious part than calculating the matrix elements, and this is completely alleviated by using the SUSYQM method.

As the first step, we find an (original) potential such that its lowest three states are equispaced. Here we have chosen the truncated quasiparabolic potential

$$U_0(z) = \begin{cases} a_1 z^2 + a_2 z^4 & , |z| < \Delta \\ V & , |z| > \Delta \end{cases} \quad (21)$$

with  $a_1$  and  $a_2$  fitted so that levels spacing are equal to a desired value (the pump photon energy), with the truncation point  $\Delta$  and the well depth  $V$  chosen so that it corresponds to a realistic QW structure. In finding  $a_1$  and  $a_2$ , via the numerical solution of the Schrödinger equation, we take account of the requirement that (21) is realized by grading the mole fraction  $x = x(z)$  of a ternary alloy  $Al_xGa_{1-x}As$ . The potential and the effective mass have both depend on  $x$ , i.e.,  $U_0(z) = \Delta U x(z)$  and  $m(z) = m_{AC} x(z) + m_{BC} [1 - x(z)]$ , wherefrom

$$m(z) = \frac{m_{AC} - m_{BC}}{\Delta U} U_0(z) + m_{BC} \quad (22)$$

with  $m_{AC}$  ( $m_{BC}$ ) denoting the effective masses in  $AC$  ( $BC$ ) compounds, and  $\Delta U$  is the difference of the conduction band edges.

Being symmetric, the potential (21) lacks any second-order susceptibility, but it is just a starting point for the SUSYQM-based search for the best potential shape. In accordance with the expressions given in Sec.II, the supersymmetric partner potential to  $U_0(z)$ , fully isospectral to it, reads

$$U_{ss}(z) = U_0(z) - \frac{\hbar^2}{\sqrt{m(z)}} \frac{d}{dz} \left[ \frac{1}{\sqrt{m(z)}} \frac{d}{dz} [\ln(\lambda + I(z))] \right] = U_{ss}(z; \lambda) \quad (23)$$

where  $m(z)$  is defined by Eq. (22). The normalized wave functions, corresponding to  $U_{ss}(z)$  are

$$\psi_{ss}^{(i)}(z) = -\psi_0^{(i)}(z) + \frac{\phi(z) \mathcal{M}_{\phi i}}{\lambda + I(z)} = \psi_{ss}^{(i)}(z; \lambda) \quad i \neq k \quad (24)$$

$$\psi_{ss}^{(k)}(z) = \frac{\sqrt{\lambda(\lambda + 1)}}{\lambda + I(z)} \psi_0^{(k)}(z) \quad (25)$$

where  $\phi(z)$  is any specified wave function  $\psi_0^{(k)}(z)$  of the original potential  $U_0(z)$ , and

$$I(z) = \int_{-\infty}^z \phi^2(t) dt \quad (26)$$

$$\mathcal{M}_{\phi i} = \int_{-\infty}^z \phi(t) \psi_0^{(i)}(t) dt \quad (27)$$

Since we have chosen the original potential to be symmetric, with the wave functions either even or odd, the integral  $I(z)$ , Eq. (26), satisfies  $I(z) = 1 - I(-z)$  and the supersymmetric potential has the property

$$U_{ss}(z; \lambda) = U_{ss}(-z; -(\lambda + 1)) \quad (28)$$

It follows that in this case all physically different  $U_{ss}(z)$  are obtained by giving only positive values to  $\lambda$  (those with  $\lambda < -1$  are just reversed).

As displayed in Fig.1, various values of  $\lambda$  produce significantly different potential shapes, which are prominently asymmetric. The same holds true for the wave functions, and consequently the transition dipole moment

$$\mu_{ij} = \int_{-\infty}^{+\infty} \psi_{ss}^{(i)}(z) z \psi_{ss}^{(j)}(z) dz \quad (29)$$

and the difference of permanent moments

$$\delta_{ij} = \int_{-\infty}^{+\infty} \psi_{ss}^{(i)}(z) z \psi_{ss}^{(i)}(z) dz - \int_{-\infty}^{+\infty} \psi_{ss}^{(j)}(z) z \psi_{ss}^{(j)}(z) dz \quad (30)$$

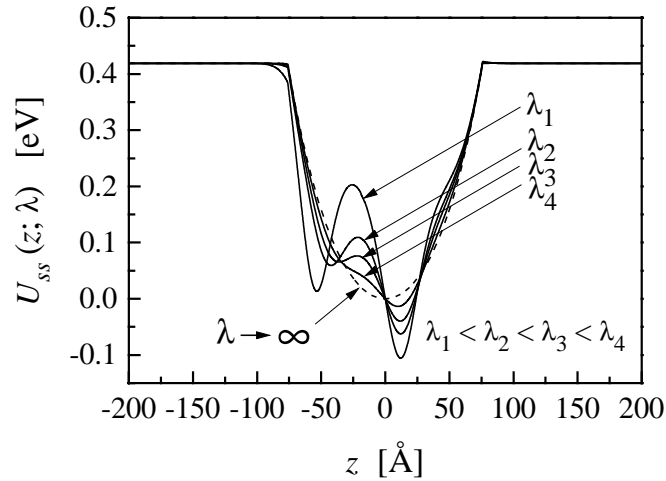


FIG. 1. The supersymmetric partner potentials  $U_{ss}(z)$  (solid lines) evaluated for a few positive values of the  $\lambda$  parameter, isospectral to the original potential  $U(z)$  (dashed line).

will both vary with  $\lambda$  in the range  $(0, +\infty)$ , and there remains to find the optimum value  $\lambda_{opt}$  which maximizes matrix element products  $\mu_{12}\mu_{23}\mu_{31}$  or  $\mu_{12}^2\delta_{12}$ . The best potential shape is then given by Eq. (23) with  $\lambda = \lambda_{opt}$ .

At this point we should note, however, that only the potential got transformed by SUSYQM method, while the effective mass retained the original symmetric form (22). Such a Hamiltonian cannot be realized by grading a ternary alloy. Enforcing the effective mass to follow the optimized potential in the manner prescribed by Eq. (22) with  $U_0(z) \rightarrow U_{ss}(z)$  would result in the loss of levels equidistance. Therefore, we have to perform some "retailoring" of the optimized potential, with the effective mass following it, in order to restore levels equidistance. The amount of retailoring may be expected to be rather small, because in real structures the variable mass does not have as large influence on levels energies as does the potential. Another effect requiring the optimized potential to be retailored is the nonparabolicity. It cannot be quit simple included in the SUSYQM, so we have to ignore it initially, and then correct the final result. In effect, having found the optimized supersymmetric potential  $U_{ss}(z)$  we set the Schrödinger equation

$$-\frac{\hbar^2}{2} \frac{d}{dz} \left( \frac{1}{m(z, E^{(i)})} \frac{d\psi_{ss}^{(i)}(z)}{dz} \right) + U_{ss}(z)\psi_{ss}^{(i)}(z) = E^{(i)}\psi_{ss}^{(i)}(z) \quad (31)$$

where the effective mass varies in the ternary-alloy-like manner and is energy dependent (Kane model of nonparabolicity), i.e.,

$$m(z, E^{(i)}) = \left( \frac{m_{AC} - m_{BC}}{\Delta U} U_{ss}(z) + m_{BC} \right) \left[ 1 + \frac{E^{(i)} - U_{ss}(z)}{E_g(z)} \right] \quad (32)$$

with the position dependent band gap given by  $E_g(z) = E_{gAC}x(z) + E_{gBC}[1 - x(z)]$ . The eigenenergies of (31), found numerically, deviate from being strictly equidistant. We correct for this by simple coordinate scaling, i.e., define the final potential  $U_{ss}^*(z') = U_{ss}(z)$  with  $z' = \alpha z + \beta|z|$ , and the effective mass following it, analogous to Eq. (32), where the symmetric and asymmetric dilatation coefficients  $\alpha$  and  $\beta$  are determined by numerical "experimenting". Since this last step of the procedure is not exactly analytical it is desirable that  $(\alpha, \beta)$  be very close to  $(1, 0)$ : it is then reasonable to assume that  $U_{ss}^*(z)$ , realizable by graded ternary  $A_xB_{1-x}C$  alloy, will be (almost) optimal, as well. At this point we may note that suitable values of  $\alpha$  and  $\beta$  might also be used to correct the optimized potential if we have started from the constant-mass truncated parabolic original. However, since the optimized potential and the effective mass following it are globally more similar to variable-mass original we

have started from, the degree of final correction is expected to be smaller than in the constant-mass case.

## IV Numerical results and discussion

### IV.1 Second harmonic generation

For numerical illustration of the method we have chosen to design a QW structure optimized for double resonance SHG of CO<sub>2</sub> laser radiation,  $\hbar\omega = 116$  meV. This implies that the QW depth should be at least  $\sim 400$  meV or so, to be able to support three bound states, hence the compounds *AC* and *BC* should be chosen accordingly. The commonly used  $Al_xGa_{1-x}As$  alloy is capable of providing such band offset, and has the additional advantage of being well understood technologically, and is strain free, so it was adopted for this design. The conduction band edge effective masses are  $m_{GaAs} = 0.066$  and  $m_{AlAs} = 0.15$  (in free electron mass units), and the  $\Gamma$ -valley conduction band offset is taken to be [12]  $\Delta U = 750$  meV. Eq. (22) then gives

$$m(z) = U_0(z)/8.9286 + 0.066 \quad (33)$$

where  $U_0(z)$  is in eV units. The band gap vs. the mole fraction dependence is described by  $E_g = 1.420 + 1.082x + 0.438x^2$ .

Choosing the constants in Eq. (21) as  $a_1 = 5.63 \times 10^{-5}$  eVÅ<sup>-2</sup>,  $a_2 = 2.81 \times 10^{-9}$  eVÅ<sup>-4</sup> and  $\Delta = 76$  Å, (so  $V = 418.9$  meV), we made the original symmetric truncated quasiparabolic potential  $U_0(z)$  and the corresponding effective mass  $m(z)$ , Eq. (22). It accommodates three bound states with energies  $E_1 = 55.1$  meV,  $E_2 = 171.7$  meV and  $E_3 = 288.3$  meV above the QW bottom, i.e.,  $\Delta E_{21} = \Delta E_{32} = 116.6$  meV.

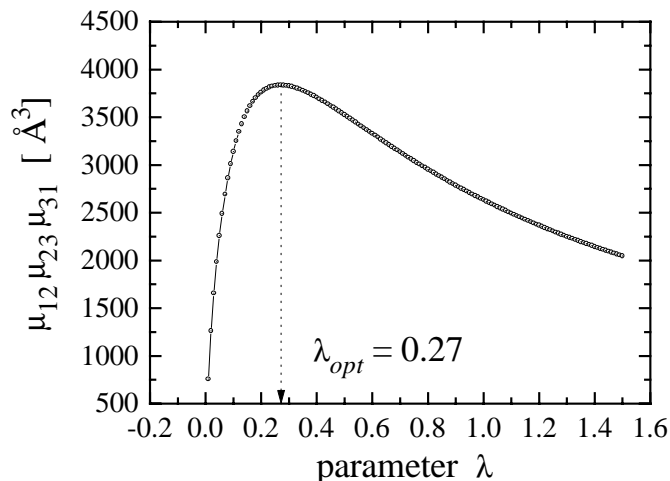


FIG. 2. The dependance of the dipole matrix elements product on the parameter  $\lambda$  in asymmetric QW with  $U_{ss}(z)$ , Eq. (23), obtained with the choice  $\phi(z) = \psi_0^{(2)}(z)$  in the SUSYQM theory.

This original potential was then SUSYQM-transformed, as described above, and varied in order to obtain the largest product of matrix elements  $\Pi^{(2)}$ . Among a couple of choices for the factorization state that we have tried, the best results were obtained by choosing the first excited state, i.e.,  $\phi(z) = \psi_0^{(2)}(z)$ , and only these results are described below. By varying the parameter  $\lambda$  we find that the largest value of  $\Pi_{max}^{(2)} = 3837.3$  Å<sup>3</sup> occurs for  $\lambda_{opt} = 0.27$  (Fig.2), the individual matrix elements being  $\mu_{12} = 17.27$  Å,  $\mu_{23} = 25.51$  Å,  $\mu_{31} = 8.71$  Å. However, this QW retains the original  $m(z)$  dependence, preventing its realization by ternary alloys, and also the nonparabolicity ignored. Solving



the nonparabolic Schrödinger equation (31) with the presently obtained potential gives the levels energies  $E_1 = 54.3$  meV,  $E_2 = 168.3$  meV and  $E_3 = 276.6$  meV, so the levels spacing  $\Delta E_{21} = 114.0$  meV and  $\Delta E_{32} = 108.4$  meV deviates slightly from what was desired. Finally, we make the coordinate scaling  $z' = \alpha z + \beta|z|$ , and find that  $\alpha = 1.065$  and  $\beta = 0.011$  restore the levels equidistance, going  $E_1 = 60.1$  meV,  $E_2 = 176.0$  meV and  $E_3 = 292.1$  meV, hence  $\Delta E_{21} = 115.9$  meV and  $\Delta E_{32} = 116.1$  meV. In this final QW, where the effective mass follows the potential and the nonparabolicity is accounted [10], for the matrix elements amount to  $\mu_{12} = 16.27 \text{ \AA}$ ,  $\mu_{23} = 22.87 \text{ \AA}$ , and  $\mu_{31} = 8.86 \text{ \AA}$ , i.e.,  $\Pi^{(2)} = 3296.8 \text{ \AA}^3$ . The drop of  $\Pi^{(2)}$  from the larger value found above is inevitable, and is mainly caused by nonparabolicity [6]: the well had to be contracted in order to compensate for the increase of the effective mass at higher energies, Eq. (31). The final optimized potential  $U_{ss}^*(z)$ , the "temporary"  $U_{ss}(z)$  and the original  $U_0(z)$  are displayed in Fig.3, together with three wave functions in the final case. The fact that  $\alpha$  is found close 1 and  $\beta$  close to 0 indicates that  $U_{ss}^*(z)$  is not much distorted from  $U_{ss}(z)$ , as is indeed clear from Fig.3, its "degree of asymmetry" is about preserved, so we expect that  $U_{ss}^*(z)$  is at least very close to being really optimal.

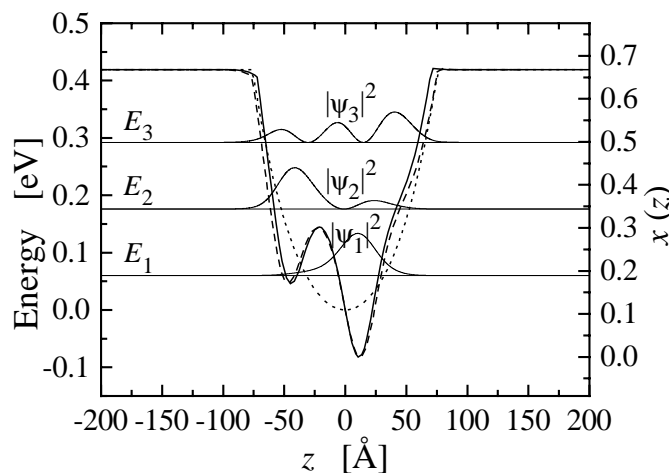


FIG. 3. The original  $U_0(z)$  (dotted line), SUSYQM-optimized  $U_{ss}(z)$  (dashed line) and finally polished  $U_{ss}^*(z)$  (solid line) potential profiles, maximizing the resonant second-order susceptibility. The solid line also gives the Al mole fraction grading function  $x(z)$  necessary to realize the optimal  $U_{ss}^*(z)$  in  $Al_xGa_{1-x}As$  alloy (values to be read on the right hand axis). The wavefunction moduli squared are also displayed.

The calculated potential  $U_{ss}^*(z)$  and the corresponding effective mass  $m(z)$  can be realized by appropriate grading of  $Al_xGa_{1-x}As$ , using the relation

$$\Delta Ux(z) = U_{ss}^*(z) + const. \quad (34)$$

where the *const.* account for the fact that the reference point for measuring the potential is generally arbitrary, and in this case may be chosen with some freedom, as convenient. As displayed in Fig.3 the optimized potential has a negative undershoot, and therefore has a larger span than the original  $U_0(z)$  which starts from zero. It is thus wise to set  $const. = |\min(U_{ss}^*(z))|$ , i.e., 81.88 meV in this case (Fig.3), in order to take advantage of the full band offset offered by  $Al_xGa_{1-x}As$ . The mole fraction  $x(z)$ , i.e., the grading function for the optimized QW is also given in Fig.3 (right hand axis).

To make a fair comparison of the present design against those reported in the literature we quote values of  $\Pi^{(2)}$  obtained previously in QW's based also on  $Al_xGa_{1-x}As$  and for the some pump photon energy (note that  $\Pi^{(2)}$  scales as  $\Pi^{(2)} \sim m^{-3/2}$ , assuming  $m(z)$  is constant, and using alloys with lower effective mass would increase  $\Pi^{(2)}$  aside from any optimization). In an asymmetric step QW the value  $\Pi^{(2)} = 2394 \text{ \AA}^3$  was obtained [3] with the effective mass taken constant, and in a similar, slightly redesigned structure  $\Pi^{(2)} = 2635 \text{ \AA}^3$ , this time with the nonparabolicity included [13]. In

simple rectangular QW based by a strong field  $K = 3.7 \times 10^6$  V/m a rather large  $\Pi^{(2)} = 3256 \text{ \AA}^3$  was calculated [5], but with the nonparabolicity ignored, it would drop well below  $3000 \text{ \AA}^3$  in a real system. Therefore, the optimized value of  $\Pi^{(2)} = 3296.8 \text{ \AA}^3$  found in this work considerably exceeds (by 20% or more) the previously reported, which is a technically significant increase.

Also, we want to mention that using constant effective mass  $m(z) = 0.066m_0$  and fitting  $a_1 = 5.43 \times 10^{-5} \text{ eV\AA}^{-2}$ ,  $a_2 = 1.09 \times 10^{-9} \text{ eV\AA}^{-4}$  and  $\Delta = 82.4 \text{ \AA}$ , ( $V = 418.9 \text{ meV}$ ) in Eq. (22) it accommodates three eigenenergies  $E_1 = 56.8 \text{ meV}$ ,  $E_2 = 171.9 \text{ meV}$  and  $E_3 = 288.0 \text{ meV}$ , i.e.,  $\Delta E_{21} = 115.1 \text{ meV}$  and  $\Delta E_{32} = 116.1 \text{ meV}$ . In that case SUSYQM transformation provide maximal value of  $\Pi_{max}^{(2)} = 4279.7 \text{ \AA}^3$  occurs for  $\lambda_{opt} = 0.26$  and  $\mu_{12} = 17.68 \text{ \AA}$ ,  $\mu_{23} = 26.73 \text{ \AA}$ ,  $\mu_{31} = 9.06 \text{ \AA}$ .

Second-order optical susceptibility, Eq. (5), is derived with assumption that sheet density of states 2 and 3, in the thermal equilibrium, can be ignored in comparison with  $\rho_{11}$ . In according with expression for electron concentration given by [5]:

$$n = \frac{m^* kT}{\pi \hbar^2} \sum_{i=1}^3 \ln \left[ 1 + \exp \left( \frac{E_F - E^{(i)}}{kT} \right) \right] \quad (35)$$

in the case of final asymmetrical QW and  $n = 10^{11} \text{ cm}^{-2}$ , we calculated Fermi level, on the room temperature  $T = 300 \text{ K}$ , at  $E_F = 10.8 \text{ meV}$ . We also obtained sheet densities:  $\rho_{11} = 9.88 \times 10^{10} \text{ cm}^{-2}$ ,  $\rho_{22} = 1.19 \times 10^9 \text{ cm}^{-2}$ , and  $\rho_{33} = 1.34 \times 10^7 \text{ cm}^{-2}$ . Ignoring the sheet density of higher levels, as we mentioned above, is completely acceptable in respect that  $\rho_{11}/\rho_{22} = 82.6$  and  $\rho_{11}/\rho_{33} = 7.36 \times 10^3$ . Much more better results are in the case of  $T = 77 \text{ K}$ , when  $E_F = 58 \text{ meV}$ ,  $\rho_{11} = 9.99 \times 10^{10} \text{ cm}^{-2}$ ,  $\rho_{22} = 3.45 \times 10^3 \text{ cm}^{-2}$ , and  $\rho_{33} = 8.12 \times 10^{-5} \text{ cm}^{-2}$ , i.e.,  $\rho_{11}/\rho_{22} = 2.9 \times 10^7$  and  $\rho_{11}/\rho_{33} = 1.23 \times 10^{15}$ . This fact also demonstrate useful of  $Al_xGa_{1-x}As$  ternary alloy for manufacturing appropriate microstructure.

Finally, we should note that using the ground state of the original potential,  $\phi(z) = \psi_0^{(1)}(z)$ , in constructing the isospectral  $U_{ss}(z)$  gave results which are only slightly worse (by a few percent) than those described above, although the corresponding QW was less deep. This might be advantageous at higher pump photon energies, but in the case we have studied here the  $\phi(z) = \psi_0^{(2)}(z)$  derived QW also turned out to be realizable by  $Al_xGa_{1-x}As$ . On the other hand, choosing  $\phi(z) = \psi_0^{(3)}$  results in  $\Pi^{(2)}$  four times lower than that obtained with  $\phi(z) = \psi_0^{(2)}$ .

## IV.2 Optical rectification

We attempt to design a QW structure optimized for optical rectification of  $10.6 \text{ \mu m}$  radiation ( $\hbar\omega = 116 \text{ meV}$ ). Starting from the truncated parabolic potential Eq. (21) with  $a_2 = 0$ , and a constant effective mass, we assume the final QW design to be realized by grading the semiconductor alloy  $Al_xGa_{1-x}As$ , advantages of which are that it is technologically well understood and is strain-free, so we set the effective mass equal to its value in GaAs, i.e.  $m^* = 0.066$  in free electron mass units. The value of the parameter  $a_1$  may be first estimated from the linear harmonic oscillator expression  $a_1 = \frac{m^*}{2}(\Delta E_{12}/\hbar)^2$ , and the potential truncation value  $V$  chosen so that it safely accomodates two bound states. We choose  $V = 0.2 \text{ eV}$ , which is well below the maximum band offset offered by  $Al_xGa_{1-x}As$ , i.e.  $\Delta U = 0.75 \text{ eV}$  (Ref. [12]), and then find by numerical experimenting that the value  $a_1 = 7.89 \cdot 10^{-5} \text{ eV\AA}^{-2}$  gives levels energies  $E_1 = 66.1 \text{ meV}$  and  $E_2 = 182.1 \text{ meV}$ , their spacing being as desired  $\hbar\omega_{21} = E_2 - E_1 = 116 \text{ meV}$ . The corresponding half-width of the truncated well is  $\Delta = 50.3 \text{ \AA}$ . In this calculation, as well as in the rest of this paper, it was assumed that the electron surface density is low enough (ie. not much more than  $10^{11} \text{ cm}^{-2}$ ) so that the self-consistent effects could be ignored.

Next we perform the procedure described by Eqs. (23-25) for a range of  $\lambda$  values, searching for the largest  $\chi_0^{(2)} \sim \mu_{12}^2 \delta_{12}$ . The best result was obtained with the choice  $\theta(z) = \psi_2(z)$  which gives  $\mu_{12} = 16.65 \text{ \AA}$  and  $\delta_{12} = 55.27 \text{ \AA}$ , i.e.  $\mu_{12}^2 \delta_{12} = 15322 \text{ \AA}^3$  at  $\lambda_{opt} = 0.26$ , Fig.4 (the corresponding value found with the choice  $\phi(z) = \psi_1(z)$  was  $\sim 5\%$  lower)[14]. To make a comparison against results

obtained elsewhere, we quote the values of  $\mu_{12}^2\delta_{12} = 12267 \text{ \AA}^3$  (Ref. [3]), and  $12118 \text{ \AA}^3$  (Ref. [15]), which were also calculated within the constant effective mass model in  $Al_xGa_{1-x}As$  based QW. Both of these are behind our result by  $\sim 25\%$ .

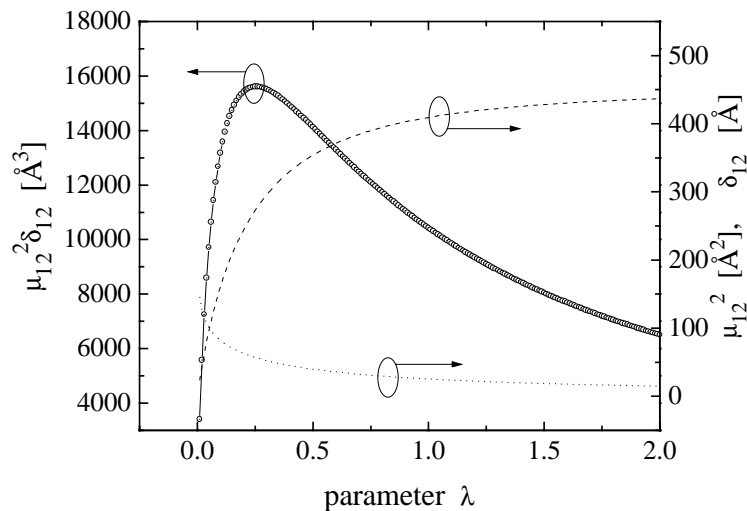


FIG. 4. Values of  $\mu_{12}^2$  and  $\delta_{12}$  (dotted and dashed lines, respectively), and of the product  $\mu_{12}^2\delta_{12}$  relevant for optical rectification (solid line) as they depend on  $\lambda$  in the system described by  $U_{ss}(z)$  and the constant effective mass in the text.

The final step of the design is to account for the position dependence of the effective mass and the nonparabolicity, as exist in real graded QW structures. Adopting the energy dependent effective mass model of nonparabolicity, actual level energies and wave functions are to be found by solving the Schrödinger equation of the form Eq. (31) where the expression for the effective mass Eq. (32) accounts for both the position dependence of its band-edge value (via the position dependent Al mole fraction  $x(z)$  in the  $Al_xGa_{1-x}As$  alloy) and the position dependent nonparabolicity (via the position dependent band gap  $E_g(z)$ ). Taking the same constants as in the case of second-harmonic generation, we find by numerical integration of Eq. (31) that the QW with optimized potential as above would in fact have energies of the two levels  $E_1 = 68.4$  meV and  $E_2 = 180.3$  meV, ie. their spacing  $\Delta E_{12} = 111.9$  meV deviates somewhat from the target value of 116 meV. We correct for this by introducing a coordinate scaling  $z' = \alpha z + \beta|z|$ , ie. define a (hopefully slightly) reshaped potential  $U_{ss}^*(z') = U_{ss}(z)$ . By numerical experimenting we find that  $\alpha = 1.065$  and  $\beta = -0.085$  restores the levels spacing to acceptable  $\Delta E_{12} = 184.9 - 69.4 = 115.5$  meV. The fact that  $\alpha$  is close to 1 and  $\beta$  to zero means that this reshaped potential  $U_{ss}^*(z)$  is quite similar to optimized  $U_{ss}(z)$ , ie. is still (at least very close to being) optimal under realistic conditions. Yet, calculating the matrix elements Eqs.(29-30) we find  $\mu_{12} = 13.25 \text{ \AA}$  and  $\delta_{12} = 60.72 \text{ \AA}$ , so the product  $\mu_{12}^2\delta_{12}$  has now dropped to  $10660.2 \text{ \AA}^3$ . This is lower than values given in Refs. [3] and [15], but their results would also be lower than stated, had the nonparabolicity been taken into account. Decreasing the matrix elements upon taking the nonparabolicity stems from the fact that QW has to be "squeezed" to compensate for an increased effective mass, if levels spacing is to be kept constant, hence all the dipole matrix elements decrease [6]. There is no remedy to it within a given semiconductor system (however, using a material with smaller effective mass would help). We may also note that the above coordinate scaling technique may be used to correct for the self-consistency effects, in cases of larger doping.

The final optimized potential, displayed in Fig.5, may be realized by a suitable grading of  $Al_xGa_{1-x}As$  alloy. To find the grading function  $x(z)$  we note that according to Eq.(34):

$$x(z) = \frac{U_{ss}^*(z) + |U_{min}|}{\Delta U}. \quad (36)$$

To take advantage of the full potential span offered by the  $Al_xGa_{1-x}As$  alloy, if this aspect becomes

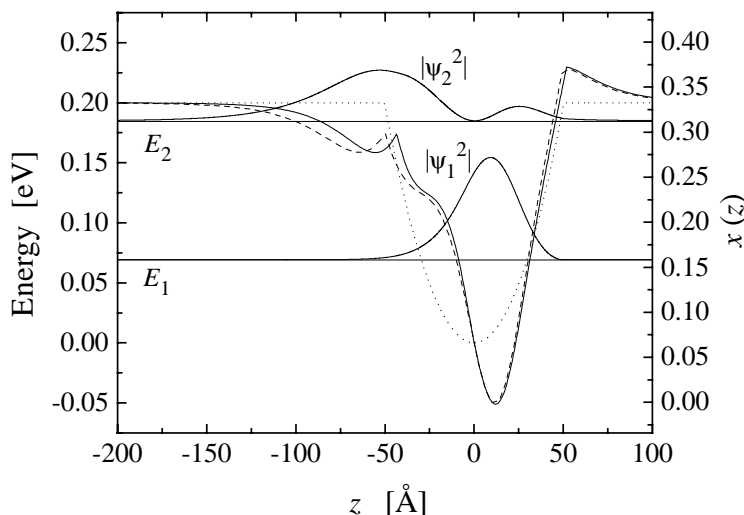


FIG. 5. The original truncated parabolic  $U(z)$  (dotted line), the constant mass SUSYQM optimized  $U_{ss}(z)$  (dashed line), and finally reshaped  $U_{ss}^*(z)$  (solid line) potential providing the largest optical rectification coefficient. The composition grading function  $x(z)$  necessary to realize these potentials in  $Al_xGa_{1-x}As$  may be read on the right hand axis. Also given are the wave functions moduli squared corresponding to  $U_{ss}^*(z)$  (thin solid lines).

critical, one should set  $const = |U_{min}|$ , where  $U_{min}$  is the minimum value of the potential in Fig.5 (ie.  $-49.5$  meV). The grading  $x(z)$  obtained that way is also displayed in Fig.5. We should note that using the ground state of the original potential,  $\phi(z) = \psi_0^{(1)}(z)$ , in constructing the isospectral  $U_{ss}(z)$  gave results which are worse (by  $\sim 5\%$ .) than those described above, although the corresponding QW was less deep.

## V Conclusion

A systematic procedure was described for the design of optimized, ternary alloys based QW's for double resonant second-harmonic generation and optical rectification. It relies on using the SUSYQM theory with variable effective mass, enabling one to vary the potential, and hence the wave functions and dipole matrix elements, while the function is implemented through variation of a single scalar parameter, so as to maximize the product of matrix elements relevant for the second-order susceptibilities. Additionally, correction to the optimized potential, to account for nonparaboly, is introduced. The predicted optimized nonlinear susceptibilities in QW's realized by graded  $Al_xGa_{1-x}As$  alloy significantly exceeds the values previously reported in the literature.

## References

- [1] J. F. Ward, Rev. Mod. Phys. **37** 1 (1965).
- [2] M. K. Gurnick and T. A. DeTemple, IEEE J. Quantum Electron. **QE-19** 791 (1983).
- [3] E. Rosencher and Ph. Bois, Phys. Rev. B **44** 11315 (1991).
- [4] M. M. Fejer, S. J. B. Yoo, R. L. Byer, A. Harwit and J. S. Harris, Phys. Rev. Lett. **62** 1041 (1989).
- [5] Z. Ikončić and V. Milanović, and D. Tjapkin, IEEE J. Quantum Electron. **QE-25**, 54 (1989)
- [6] V. Milanović and Z. Ikončić, IEEE J. Quantum Electron. **QE-32** 1316 (1996).
- [7] D. Ahn and S. L. Chuang, IEEE J. Quantum Electron. **QE-23** 2196 (1987).

- 
- [8] E. Rosencher, P. Bois, B. Vinter, J. Nagle, and D. Kaplan, *Appl. Phys. Lett.* **56** 1822 (1990).
  - [9] A. Selmeier, H. J. Hübner, G. Abstreiter, G. Weimann, and W. Schlapp, *Phys. Rev. Lett.* **59** 1345 (1987).
  - [10] S. Tomić, V. Milanović, and Z. Ikonić, *Phys. Rev. B* **56** 1033 (1997).
  - [11] F. Cooper, A. Khare, and U. Sukhatme, *Phys. Reports* **251** 267 (1995).
  - [12] G. Bastard, *Wave Mechanics Applied to Semiconductor Heterostructures*, Les Editions de Physique, Les Ules, 1990.
  - [13] S. J. B. Yoo, M. M. Fejer, R. L. Byer, and J. S. Haris Jr., *Appl. Phys. Lett.* **58**, 1724 (1991)
  - [14] S. Tomić, V. Milanović, and Z. Ikonić, *Optics Comm.* (in Press).
  - [15] M. Zaluzny, *J. Appl. Phys.* **74** 4716 (1993).

(Received February 1997.)