Influence of indirect minima on electron concentration in GaAs-$Al_xGa_{1-x}As$ superlattices: A numerical study

Z. Ikonić
Faculty of Electrical Engineering, Bulevar Revolucije 73, Belgrade, Yugoslavia

V. Milanović
Faculty of Electrical Engineering, Bulevar Revolucije 73, Belgrade, Yugoslavia and High Post, Telegraph, Telephone School, Belgrade, Yugoslavia

D. Tjapkin
Faculty of Electrical Engineering, Bulevar Revolucije 73, Belgrade, Yugoslavia
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This paper treats the influence of indirect minima on electron concentration in GaAs-$Al_xGa_{1-x}As$ superlattices. It is shown that the population of the minizones arising from indirect minima significantly depends on superlattice parameters. For higher values of the mole fraction x and for thinner wells, the relative population of indirect-minima minizones is remarkable, and may even be equal to unity for $x=1$ and for very thin layers (~ 3 nm). This effect is of importance in the determination of superlattice parameters.

I. INTRODUCTION

The development of special technologies (e.g., molecular-beam epitaxy[1]) makes possible the realization of very thin semiconductor layers (in the range of tens to hundreds of angstroms, shorter than the electron mean free path but longer than the lattice spacing.[2] Thus, superlattice (SL) structures representing periodic sequences of thin semiconductor layers of alternating composition or type of doping are now being realized. The properties of semiconductor SL's have been intensively studied in recent years in relation to the possibility of making semiconductor lasers having low threshold current and temperature sensitivity, negative differential conductivity, and high low-temperature mobility (see, e.g., Ref. 3).

Determination of the relevant parameters of the SL's requires fairly good knowledge of their band structure. A considerable number of papers has been devoted to this problem. The SL that has been most thoroughly studied both theoretically and experimentally is certainly GaAs-$Al_xGa_{1-x}As$ (see, e.g., Ref. 4). In theoretical considerations of the band structure of this SL, due attention has been paid to the effective-mass difference and nonparabolicity in the host materials.[5,6]

However, most of the papers deal only with electrons belonging to the Γ minimum, having the lowest energy in GaAs and in $Al_xGa_{1-x}As$ with mole fraction x less than 0.45 (see, e.g., Ref. 7). Generally, however, the existence of a complex band structure of the host materials which have X and L minima along with the Γ minimum implies the necessity of accounting for their influence on the complete band structure of SL. As far as we know this problem has been treated in only two papers.[8,9] In Ref. 8 the proper method to deal with this problem was in principle given, but no exact calculations were presented. A more detailed analysis was given in Ref. 9, but we think it is inconsistent with Ref. 8 (see Sec. II for details).

In this paper we shall present a detailed theoretical and numerical analysis of the band structure of GaAs-$Al_xGa_{1-x}As$ SL's, within the effective-mass approximation, paying special attention to the influence of X and L minima.

II. THEORETICAL CONSIDERATIONS

We shall analyze the SL structure consisting of GaAs layers of thickness a and $Al_xGa_{1-x}As$ layers of thickness b (the SL period is $d=a+b$). The band structures of GaAs and $Al_xGa_{1-x}As$ are very similar, having minima at the same points in $\mathbf{k}$ space, differing only in the energies of these minima, which depend on mole fraction x ($0\leq x\leq 1$). There are three types of minima, Γ at $\mathbf{k}=\mathbf{0}$, X along the [100] direction, and L along the [111] direction, the last two being at the edge of Brillouin zone. We shall take the constant-energy surfaces to be spheres at Γ and ellipsoids at the X and L minima, for all energies of interest. Certainly, although the band structures, even in the vicinity of these minima, are more complex (see, e.g., Ref. 10), the influence of nonparabolicity is not too strong, at least for the energy states most carriers occupy.[11]

Within this approximation the motion of carriers belonging to any of these minima can be described by the effective-mass Schrödinger equation:

$$(\hat{T}+\hat{U})\psi=E\psi\ , \tag{1}$$

where ψ is the envelope wave function, E is the total energy of the carriers, $\hat{U}$ is the potential-energy operator, and $\hat{T}$ is the kinetic energy operator given by

$$\hat{T}=\tfrac{1}{2}(\hat{p}-\hat{p}_i)\underline{M}^{-1}(\hat{p}-\hat{p}_i)\ . \tag{2}$$

$\hat{p}$ being the momentum operator, $\hat{p}_i$ the position of the ith minimum, and $\underline{M}^{-1}$ is the reciprocal effective-mass tensor.

A number of papers has been devoted to the form of the T operator for the case of a position-dependent effective mass.[12,13] For an abrupt change of effective mass (the case we treat here) the two most used forms, Harrison's[14] (used here) and Leibler's,[15] are equivalent.

When applying (2) to SL's composed of materials having the same type of band structure (which is the case for the GaAs-$Al_xGa_{1-x}As$ SL's we consider here) one can treat the carriers belonging to different minima independently (as was discussed by Schik[8]), implying that each minimum forms its own set of minizones, their parameters being determined by the properties of this minimum in the host materials. This assumption relies on the fact that, as pointed out by Schik,[8] the electronic transition between any two different minima is possible only by scattering with a phonon having wave vector of the order of π/δ (δ is lattice period). As stated previously, the SL period is smaller than the electron mean free path, so electrons will pass a considerable number of SL periods without scattering (especially with phonons of order of π/δ), remaining all the time in one minimum [this enables one to use the Schrödinger equation (1) (without scattering) when treating thin-period quantum SL's].

There is also another mechanism of valley mixing in SL's arising from electron transmission and reflection at the well-barrier boundaries.[16] However, no more than 0.7% of the electrons undergo intervalley transitions at a single boundary (the exact fraction depends on mole fraction x and the electron wave vector). Hence, intervalley scattering is a second-order effect which will be neglected here.[17]

In Ref. 9 it was assumed that electrons in GaAs belong to the Γ minimum and those in $Al_xGa_{1-x}As$ ($x>0.45$) belong to the X minima; the corresponding wave functions were matched at the interface, which, according to the preceding discussion is not adequate.

With these assumptions, the SL-periodic part η of the envelope function for any particular minimum satisfies[18]

$$-\frac{\hbar^2}{2}\left|\frac{d}{dz}\left[\frac{1}{m_{zz}}\frac{d\eta}{dz}\right]\right|+U_{\text{eff}}\eta=E\eta\ , \tag{3}$$

where U_{eff} is the effective potential energy given by

$$U_{\text{eff}}=U(z)+\frac{\hbar^2}{2}\left|(k_x-k_{x0})^2\left[\frac{1}{m_{xx}}-\frac{m_{zz}}{m_{xz}^2}\right]+2(k_x-k_{x0})(k_y-k_{y0})\left[\frac{1}{m_{xy}}-\frac{m_{zz}}{m_{xz}m_{yz}}\right]+(k_y-k_{y0})^2\left[\frac{1}{m_{yy}}-\frac{m_{zz}}{m_{yz}^2}\right]\right| . \tag{4}$$

In (4), $U(z)$ is the energy dependence of the bottom of the corresponding minimum (which includes the space-charge potential, as well). The effective-mass tensor components depend on the choice of coordinate-axis orientation. These components and the longitudinal (m_l) and transverse (m_t) effective masses are related (see, e.g., Ref. 19):

$$\frac{1}{m_{xx}}=\frac{1}{m_l}\sin^2\theta+\frac{1}{m_t}\cos^2\theta,\quad \frac{1}{m_{yy}}=\frac{1}{m_t},\quad \frac{1}{m_{zz}}=\frac{1}{m_t}\sin^2\theta+\frac{1}{m_l}\cos^2\theta\ ,$$
$$\frac{1}{m_{xy}}=\frac{1}{m_{yx}}=0,\quad \frac{1}{m_{xz}}=\tfrac{1}{2}\sin(2\theta)\left[\frac{1}{m_l}-\frac{1}{m_t}\right]\ , \tag{5}$$

where θ is the angle between the z axis and the rotational axis of the ellipsoid.

Applying the periodic boundary conditions[20] one arrives at the dispersion relation (for symmetric SL's)

$$\cos(k_z d)=1+\frac{2y_2(d/2)y_1'(d/2)}{y_1(d/2)y_2'(d/2)-y_2(d/2)y_1'(d/2)}\ , \tag{6}$$

where k_z is the SL Bloch vector and $y_1(z)$ and $y_2(z)$ are the even and odd solutions, respectively, of (3).

After having determined the whole band structure (minizones and wave functions) of all minima, the electron concentration can be found from[18]

$$n(z)=\sum_{i,l}n_{i,l}(z)=\frac{d}{2\pi^3}\int_0^{\pi/d}dk_z\int_{-\infty}^{+\infty}\frac{|\eta|^2dk_xdk_y}{\exp|(E-E_F)/kT|+1}\ , \tag{7}$$

where summation is to be performed over all minizones (l) and minima (i). Wave-vector components in (7) are reduced to the position of ith minimum in $\mathbf{k}$ space.

III. NUMERICAL RESULTS AND DISCUSSION

We shall proceed with the numerical part of this paper for the GaAs-$Al_xGa_{1-x}As$ SL's grown in the [100] direction, which is almost exclusively used today. As already mentioned, both materials have similar band structure with minima at the Γ, X and L points in $\mathbf{k}$ space.

In order to determine the effective potential energy [see Eq. (3)], one has to find the discontinuities ΔE_g^i of the corresponding minima in the conduction band. Obviously, we have

$$\Delta E_c^i+\Delta E_v=\Delta E_g^i\ , \tag{8}$$

where ΔE_g^i is the band-gap discontinuity, while ΔE_v is the

valence-band discontinuity, which is unique since all gaps E_g^i are measured from the top of the valence band (in $\mathbf{k}=0$). The ΔE_g^i were taken from Ref. 7. It is widely accepted now that $\Delta E_v = Q_h \Delta E_g^\Gamma$ ($Q_h = 0.15$), which has been experimentally verified for mole fractions x between 0.19 and 0.27 (see Ref. 21). However, it was shown in Ref. 22 that $\Delta E_v = 0.15 E_g^\Gamma$ holds for all values of x. Recently, the value of $Q_h = 0.15$ has been questioned in Refs. 23 and 24, where it was found that the value of $Q_h = 0.43$ makes a better fit to experimental results. In our calculations we shall study both cases ($Q_h = 0.15$ and $Q_h = 0.43$), assuming that Q_h is mole-fraction independent. Even if a dependence of Q_h on x exists it is certainly very weak, so the results of this paper should not be significantly changed.

Having the aforementioned considerations in mind and using values ΔE_g^i from Ref. 7, we arrive at (ΔE_c^i in eV)

$$
\begin{aligned}
\Delta E_c^\Gamma &= \begin{cases} (1-Q_h)(1.247x) & \text{for } x < 0.45 , \\ (1-Q_h)[1.247x + 1.147(x-0.45)^2] & \text{for } x \geq 0.45 , \end{cases} \\
\Delta E_c^L &= 0.642 - \begin{cases} 1.247 Q_h x & \text{for } x < 0.45 , \\ [1.247x + 1.147(x-0.45)^2] Q_h & \text{for } x \geq 0.45 , \end{cases} \\
\Delta E_c^X &= 0.125x + 0.143x^2 - \begin{cases} 1.247 Q_h x & \text{for } x < 0.45 , \\ [1.247x + 1.147(x-0.45)^2] Q_h & \text{for } x \geq 0.45 . \end{cases}
\end{aligned}
\tag{9}
$$

The graphs of ΔE_c^i versus x ($i = \Gamma, X, L$) are given in Fig. 1. Inspection of Fig. 1 immediately reveals that the ΔE_c^i are remarkably dependent on x and Q_h, and also that the following very interesting cases exist: (i) for $Q_h = 0.15$ and $x = 0.4339$, $\Delta E_c^X = 0$, and (ii) for $Q_h = 0.43$ and $x = 0.886$, $\Delta E_c^L = 0$, implying that SL's with these parameters have no conduction-band discontinuities (for these minima only). Even in these cases however, the energy spectrum is bandlike, because of the effective-mass discontinuity, as predicted in Ref. 5. One can see from Fig. 1 that for the Γ minimum, the barriers are in $Al_xGa_{1-x}As$ and the wells in GaAs for all values of x and for both Q_h; for the L minimum the same is true for $Q_h = 0.15$, but for $Q_h = 0.43$ and $x > 0.886$, the barriers and wells reverse their positions; for the X minimum, the barriers are in GaAs and wells in $Al_xGa_{1-x}As$, except for $Q_h = 0.15$ and $x > 0.4339$. These conclusions are valid for the zero transverse component of the wave vector. For nonzero values, the positions of barriers and wells may interchange, depending on the effective-mass ratio and the sign of the conduction-band-edge discontinuity (see Ref. 5 for details).

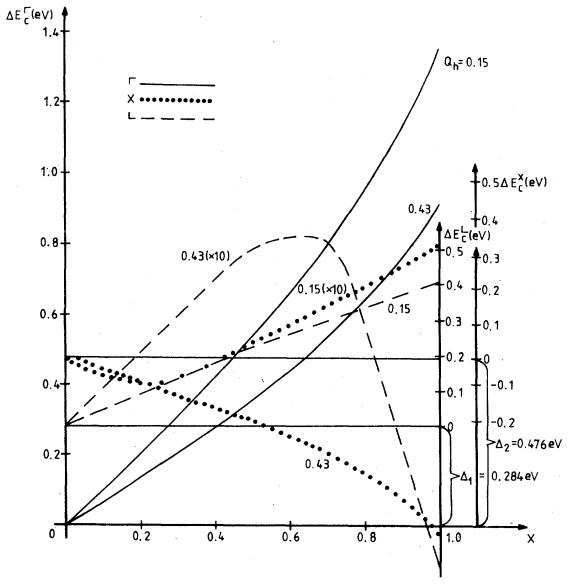

FIG. 1. Dependence of conduction-band-edge discontinuities on mole fraction x, for various minima and Q_h values. Solid, dashed, and dotted lines correspond to Γ, L, and X minima, respectively. Δ_1 and Δ_2 are energies of L- and X-minima bottoms in GaAs, measured from the bottom of the Γ minimum in GaAs. For sake of clarity $\Delta E_c^L(Q_h = 0.43)$ and $\Delta E_c^X(Q_h = 0.15)$ are magnified tenfold.

The input parameters in the band-structure calculation were the conduction-band-edge discontinuities given by (8) and the effective masses in the barriers and wells, also dependent on x. The effective mass for $Al_xGa_{1-x}As$ are obtained from a linear extrapolation as a function of mole fraction x from the GaAs masses to the AlAs masses, obtained from[25]

$$
\begin{aligned}
m^\Gamma &= \begin{cases} 0.067 m_e \\ 0.15 m_e , \end{cases} \\
m_t^L &= \begin{cases} 0.0754 m_e \\ 0.15 m_e , \end{cases} \qquad m_l^L = \begin{cases} 1.9 m_e \\ 1.32 m_e , \end{cases} \\
m_t^X &= \begin{cases} 0.23 m_e \\ 0.19 m_e \end{cases} \qquad m_l^X = \begin{cases} 1.3 m_e \\ 1.1 m_e , \end{cases}
\end{aligned}
\tag{10}
$$

where the upper values correspond to GaAs, and lower values to AlAs (m_e is the free-electron mass).

We shall treat rectangular potential energy versus z-coordinate dependence, arising from conduction-band-edge discontinuities only. This may be considered as a trial solution of the fully self-consistent treatment, which would take into account the potential of free carriers and impurities.

Since the barriers for certain values of the SL parameters may be very small (see Fig. 1) it is not possible to use the tight-binding approximation, therefore we must use the exact (numerical) solution of (3) to obtain quantitatively acceptable results.

The band structure was calculated numerically via (2.6). Results for a SL with $a=b=3$ nm and $x=0.5$ are depicted in Fig. 2. Obviously, there is one set of minizones each belonging to the Γ and L minima and two sets belonging to the X minimum, one corresponding to ellipsoids with rotational axis parallel to the z axis, and the other to those with axis perpendicular to the z axis. Certainly, this corresponds to SL's grown in the [100] direction; generally the number of sets of minizones belonging to X and L minima depends on the growth direction, while there is only one set of Γ minizones.

After having determined the SL band structure we were able to calculate the average electron concentration, given by

$$n_{\mathrm{av}}=\frac{1}{d}\int_0^d n(z)dz\ . \tag{11}$$

Of course the neutrality condition implies that n_{av} equals the average donor concentration in the SL, N_{av}.

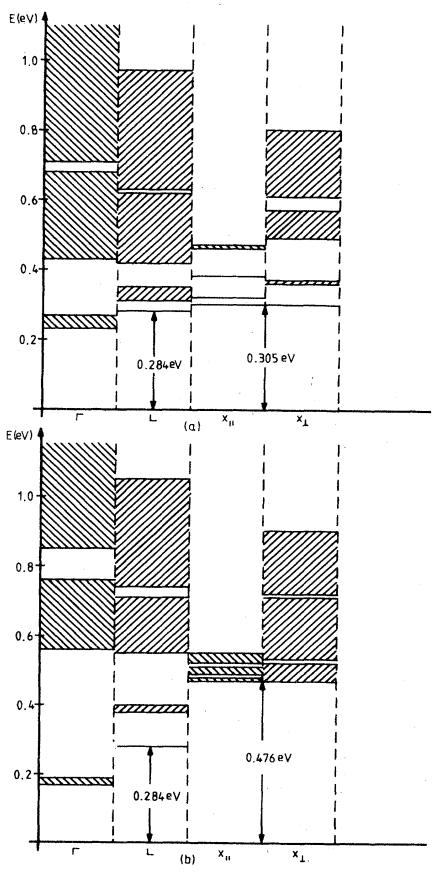

FIG. 2. Band structure of $Ga_{0.5}Al_{0.5}As$-GaAs SL ($a=b=3$ nm) for $k_x=k_y=0$ and (a) $Q_h=0.15$ or (b) $Q_h=0.43$. Shaded regions denote allowed minizones. All energies are measured from the bottom of Γ minimum in GaAs. $X_{||}$ and $X_{\perp}$ correspond to X minima with principal ellipsoid axes parallel and normal to z.

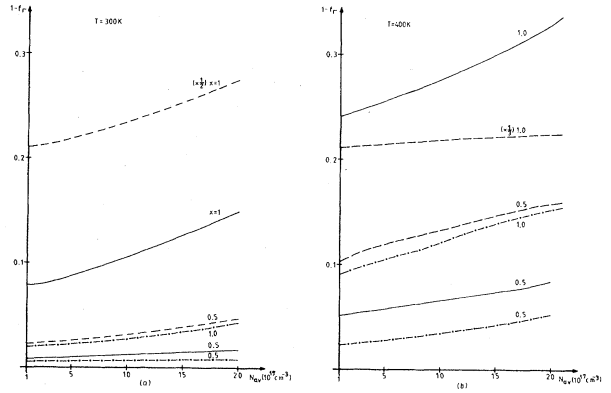

FIG. 3. Dependence of f_Γ on average impurity concentration N_{av} for various values of well thickness a (solid, dashed, and dotted lines correspond to $a=2.5$, 2.0 and 3.0 nm, respectively) and temperature T. Values for $x=1$ and $a=2$ nm from (a) and (b) should be multiplied by (a) 2 and (b) 3.

The purpose of this work was to determine the relative population of each minimum. The relative population of the Γ minimum, f_Γ ($f_\Gamma=n_{\mathrm{av}}^{\Gamma}/n_{\mathrm{av}}$) versus various parameters (a, x, n_{av}, and the temperature T) is depicted in Figs. 3, 4 ($Q_h=0.15$), and 5 ($Q_h=0.43$).

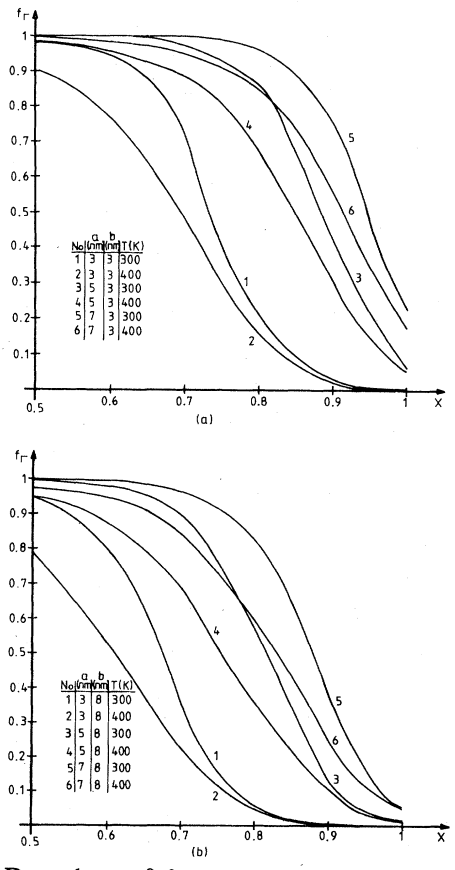

FIG. 4. Dependence of f_Γ on mole fraction x for $Q_h=0.15$, $N_{\mathrm{av}}=3\times10^{17}$ cm^{-3} and (a) $b=3$ nm and (b) $b=8$ nm.

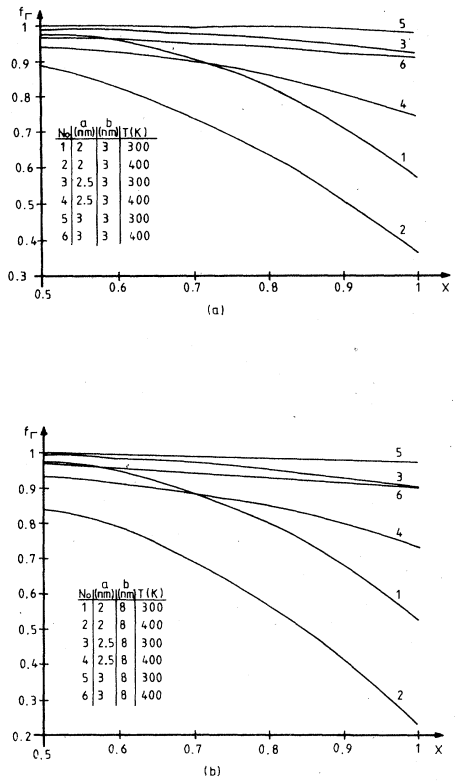

FIG. 5. Dependence of f_Γ on mole fraction x for $Q_h=0.43$, $N_{av}=3\times10^{17}$ cm^{-3} and (a) $b=3$ nm and (b) $b=8$ nm.

Inspection of Figs. 3 and 4 reveals that f_Γ decreases with increasing mole fraction x and increases with increasing GaAs layer thickness. For $x<0.5$, f_Γ is very close to unity for all values of a, n_{av}, and for all temperatures of interest. Also, for $a>5$ nm, f_Γ is near unity as well. Consequently, in these cases, disregarding the existence of X and L minizones is quite justifiable. However, SL's having large x, thin wells, and thicker barriers can have a considerable fraction of electrons on the indirect minima, as depicted in Figs. 3 and 4 (most of them being on the X minimum). These effects are due to the fact that (for $Q_h=0.15$) $\Delta E_c>\Delta E_c^x$, ΔE_c^L and the bottom of Γ minimum in GaAs is lowest in energy. Hence, for not too large values of x and thicker wells, Γ minizones will lie below the X and L minizones (due to the fact that Γ minimum in GaAs is lowest in energy). However, with increasing x and decreasing well thickness, the energies of the Γ minizones will increase faster than those of the X and L minizones (because $\Delta E_c>\Delta E_c^x,\Delta E_c^L$) and will approach them more and more. Therefore, the relative population of X and L minizones will grow. This effect is enhanced by the fact that effective masses in X and L minima are greater than those in the Γ minimum implying greater density of states in the former. One can also see from Fig. 3 that dependence of f_Γ on n_{av} is weak. f_Γ decreases with temperature because the increasing number of carriers populates higher (X and L) minizones.

For $Q_h=0.43$ the dependence of f_T on n_{av} is also weak, so it is not given here. The dependence of f_T on x, with a, b, and T as parameters is depicted in Fig. 5 for a typical value of $n_{av}=3\times10^{17}$ cm^{-3}. One can see that with increasing x, a decreasing number of electrons populates the Γ minizones, especially for thinner GaAs and thicker $Al_xGa_{1-x}As$ layers (e.g., for $a=$ 3nm and $x=1$, f_Γ is virtually zero). One can find the explanation of this effect by referring to Fig. 1: the bottoms of the X and L minima in $Al_xGa_{1-x}As$ approach the bottom of the Γ minimum in GaAs, as x tends to unity, and for $x=1$ these minima lie below Γ minimum. ΔE_c^x and ΔE_c^L are negative in this range (implying that the GaAs layers are barriers and the $Al_xGa_{1-x}As$ layers are wells) and are somewhat less in absolute value than ΔE_c. This in conjunction with the fact that effective masses in X and L minima are greater, implies that X and L minizones lie below Γ minizones; consequently the populations of the former is higher.

Since only direct-minimum electrons are active in laser action in SL's (stimulated emission via one-quantum transition a nonunity value of f_r should be taken into account when analyzing SL lasers. There are two interesting (in respect to the previous comment) papers reporting the laser action in optically pumped all-binary GaAs-AlAs SL's,[26,27] having $a=b=8$ nm and $a=b=5$ nm, respectively. If Q_h were 0.15, f_Γ would be close to unity in both cases, and if $Q_h=0.43$, f_Γ would be about 5% and 3%, respectively. However, this fact does rule out the $Q_h=0.43$ value as one could conclude at first glance, for the following reasons. Both lasers were pumped optically, and due to higher absorption in direct transitions than in indirect ones, the majority of electrons will be created in Γ minizones, giving a nonequilibrium electron distribution within the conduction band. This distribution will relax to the equilibrium one (with aforementioned values of f_Γ) with time constant equal to the intervalley scattering time (being ~1 ps in GaAs, Ref. 28, and probably having a similar value in the SL). The lifetime of Γ electrons (and holes) in SL when laser action takes place is equal to the photon lifetime in the resonator[29]

$$t_c=nl/c[\alpha l-\ln(R_1R_2)^{1/2}],$$

n and α being the index of refraction and the absorption coefficient of the active medium (SL), respectively $R_{1,2}$ the mirrors reflectivity, and l the resonator length. Taking $n=3.4$, $R_1=R_2=0.3$, $\alpha=10$ cm^{-1}, and $l=200$ μm we get $t_l=1.6$ ps. Therefore, a considerable fraction ($\sim30\%$) of the electrons will be swept out by stimulated recombination instead of relaxing to indirect minizones, and a low value of f_Γ in this case does not influence, too adversely, the laser operation. However, if carriers were injected in the SL from lower-refractive-index confining layers (indirect-gap semiconductor), electrons would populate mostly the indirect minizones and only a small fraction f_Γ would relax to the direct ones, implying a remarkable increase in threshold pumping power.

It is not a goal of this work to consider the influence of f_Γ on device operation, so we shall not proceed with this discussion here. We just note that experiments of this type (comparing optical versus electrical pumping thresholds), though not fundamental, may help in determining the value of Q_h.

IV. CONCLUSION

In this paper the influence of indirect minima on electron concentration in GaAs-$As_xGa_{1-x}As$ SL's was analyzed. In Sec. II, we presented theoretical considerations based on the Schrödinger equation for electrons belonging to various minima, within the approximation of parabolicity in host materials, proceeding to the numerical calculation of the band structure and electron concentration.

The main goal of this work was the determination of the relative population of minizones arising from direct or indirect minima. Since two values of Q_h (ratio of valence-band to energy-gap discontinuities) are currently used in the literature, the usual value 0.15 and recently proposed value 0.43, were studied in both cases. The numerical results indicate that the population of indirect minima, is negligible for x less than 0.5. For higher values of x it can have significant values, increasing with increasing mole fraction and decreasing with increasing GaAs layer thickness. For $x=1$, $Q_h=0.43$ and equal layer thicknesses of 3 nm, the population of Γ minizones is practically zero (Fig. 4). However, this effect is not pronounced for $Q_h=0.15$, f_Γ approaching ~ 0.25 for the thinnest GaAs layers of practical interest. Also, the population of the indirect minima increases with temperature, and weakly increases with average impurity concentration, at least for $N_{av}<2\times 10^{18}$ cm^{-3}.

Generally, indirect-minima electrons may play a significant role in SL's especially for higher values of the mole fraction and for thinner GaAs layers. Finally, it is shown that a nonunity value of f_Γ in a SL laser, in certain cases, implies a remarkable increase in threshold pumping power.

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