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Corrections to enhanced optical nonlinearity of superlattices

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the active layer, typically $2\ \mu\text{m}$, is obtained. The solution of $\text{K}_3\text{Fe}(\text{CN})_6 + \text{KOH} + \text{H}_2\text{O}$ (1:10:100 by weight, at $25\ ^\circ\text{C}$) was used in this process and its etching rate for the composition of quaternary layer lasing at $1.3\ \mu\text{m}$ is shown in Fig. 2. The final etching was carried out using $\text{HCl} + \text{H}_2\text{O}$ again. Since the quaternary active layer is not etched by HCl , it works as the inner etching mask for the InP layer. Therefore, the cladding layers above and below the active layer are etched showing reversed-mesa and normal-mesa shapes, respectively. As shown in Fig. 1(d), the sidewalls of the upper cladding layer are $\{211\}$ *A* surfaces, and the lower layer exposes vertical $\{011\}$ surfaces followed by $\{111\}$ *B* slopes. The etching is continued until the cladding layer is etched up to the oxide mask; this can be observed above the oxide mask with a microscope. Then, the wafer was immersed in buffered HF in order to remove the oxide film with residual InP triangle parts outside the *n*-cladding layer: when the wafer is immersed until the thickness of oxide film is reduced to half of the initial value, the bridging parts of the film are etched off completely.

After the etching process mentioned above, the burying layers, namely, *n*- InP blocking ($\sim 1\text{-}\mu\text{m}$ thickness, $n \sim 5 \times 10^{17}\ \text{cm}^{-3}$) and *p*- InP confining ($2\text{--}4\ \mu\text{m}$,

$p \sim 5 \times 10^{17}\ \text{cm}^{-3}$) layers were grown on the etched wafer with the oxide film by the second cycle LPE; the total growth time was shorter than 5 min at $600\ ^\circ\text{C}$. Thus the active layer is surrounded completely by InP layers with smaller refractive indices resulting in the index guiding. Figure 3 shows a scanning electron micrograph of the cross section of BH LD. Metals of Au-Zn and Au-Ge-Ni were deposited on the wafer as *p* and *n* electrodes, respectively, and the laser dice with a cavity length of $250\ \mu\text{m}$ was mounted on the diamond heat sink via Au-Sn solder after the cleavage.

A typical example of the output power and the far-field patterns parallel to the junction plane of the present BH LD with an active layer width of $2\ \mu\text{m}$ obtained under cw operation is depicted in Fig. 4. By the results shown in Fig. 4, the fundamental transverse mode operation is confirmed. It is also verified by measurements of dL/dI versus injected currents. The lowest value of threshold current of 15 mA with the active layer width of $0.8\ \mu\text{m}$ has been achieved.

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Corrections to enhanced optical nonlinearity of superlattices

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In recent publications, a large enhancement of the third order nonlinear optical susceptibility was predicted for GaAs-GaAlAs superlattices, as a result of the band nonparabolicities introduced by the additional periodicity of the superlattice. These predictions, based on the tight binding model, are here extended to the more realistic Kronig-Penney model. Results show that corrections to tight binding are non-negligible; however, enhancements of $\chi^{(3)}$ are still large, but reduced by approximately 30%–50% over previous estimates.

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In two earlier publications,^{1,2} predictions were made of enhanced third order nonlinear optical susceptibilities of GaAs-GaAlAs superlattices. The enhancement is due to mobile electrons in nonparabolic (dispersive) energy bands,^{3,4} which in turn arise from the additional periodicity of the superlattice. This converts the bulk band structure into a series of minibands each extending over only a small fraction of the original Brillouin zone. In the lowest miniband (the only one ordinarily relevant at realistic doping concentrations), the nonparabolicity results in the Bloch velocity of the electron being a nonlinear function of its momentum, and it is this feature, in the presence of impressed laser fields, which causes the optical mixing and a large nonvanishing third order susceptibility $\chi^{(3)}$ comparable to that of bulk InSb and two orders of magnitude larger than that of bulk GaAs . In performing this calculation, the electron's energy-momentum relation in the direction perpendicular

(\perp) to the layers, was taken to be of the standard tight-binding form⁵ (sinusoidal approximation).

$$E(k) = t [1 - \cos(kd)], \quad (1)$$

where t is the transfer integral (half the bandwidth in the direction perpendicular to the layer) due to the overlap of wave functions of adjacent quantum wells, and d is the superlattice period. While this approximation is commonly made in studying superlattice phenomena,⁵ its validity has not been investigated in general, and for the nonlinear susceptibility in particular. If it is assumed that the layers are sufficiently wide for the (bulk) effective mass approximation to be valid within the layers, and if it is also assumed that the conduction-band discontinuities are abrupt, then the electron's motion perpendicular to the layers is given by that of the Kronig-Penney⁶ model. In this short letter we extend our previous calculation^{1,2} of $\chi^{(3)}$ to the more realistic

Kronig–Penney model to describe the superlattice band structure. We investigate the electron’s dispersion in this model as it approximates the tight-binding form of Eq. (1) and the magnitude and form of the correction terms thereto. As would be intuitively expected, the tight-binding approximation obtains in the limit of weakly interacting wells—wide barriers and large conduction-band discontinuities, V_0 . For the well widths, barrier widths, and values of V_0 in the present study, we find that the corrections to tight binding are not negligible and reduce the magnitude of the predicted susceptibility enhancement ratio by the order 30%–50%; however, the net enhancement nevertheless is still appreciable. The correction is larger than what would apply to the Bloch velocity or effective mass, since $\chi^{(3)}$ is proportional to the third derivative of the velocity, or the fourth derivative of the dispersion relation, as will be shown below.

The theory to calculate $\chi^{(3)}$ proceeds analogously to that presented in Ref. 1. We consider a typical GaAs–GaAlAs superlattice doped with electrons to a carrier density n . As shown in Ref. 1, $\chi^{(3)}$ is proportional to the third derivative of the electron’s group velocity with respect to k or equivalently to the fourth derivative of its energy dispersion relation. The difference here from our previous theory is that the energy dispersion is taken to be given by the Kronig–Penney model rather than by the tight-binding approximation.

The electron’s motion in the superlattice is assumed separable, i.e., the dispersion relation can be written as a sum of energy in the layers (momentum K) and perpendicular to the layers (momentum k):

$$E(K, k) = \hbar^2 K^2 / 2m^* + E(k). \quad (2)$$

Here, with the assumptions stated previously, a parabolic effective mass approximation is made for electron motion parallel to the layers, while perpendicular to the layers $E = E(k)$ is the solution of the one-dimensional Kronig–Penney problem⁶:

$$\cos(kd) = \cosh(2k_1 a) \cos(2k_2 b) + (\epsilon/2) \sinh(2k_1 a) \sin(2k_2 b), \quad (3)$$

where $L_2 = 2a$ is the barrier width, $L_1 = 2b$ the well width, ($d = 2a + 2b$),

$$k_1 = [2m^*(V_0 - E)/\hbar^2]^{1/2}, \quad k_2 = [2m^*E/\hbar^2]^{1/2}, \\ \epsilon = (k_1/k_2 - k_2/k_1),$$

and V_0 is the barrier height (conduction-band energy discontinuity).

Equation (3) presents a rather complicated, implicit functional relation for energy versus momentum making a direct calculation of the fourth derivative difficult. However, the right-hand side of Eq. (3) is a function of E , and so we may write

$$\cos(kd) = f(E). \quad (4)$$

Then expanding $f(E)$ about $E = E_0$ corresponding to the bottom of the band ($k = 0$), we have

$$\cos(kd) = c_0 + c_1(E - E_0) + (c_2/2!)(E - E_0)^2 + \dots, \quad (5)$$

where $c_0 = 1$ and $c_k = (\partial^k f / \partial E^k)|_{E=E_0}$. Neglecting second order corrections and higher ($c_k = 0, k \geq 2$), the tight-binding form may be recovered

$$E(k) = t [1 - \cos(kd)], \quad (6)$$

with $t = -1/c_1$. Using the full Taylor series, we obtain

$$E = E_0 + [\cos(kd) - c_0] / [c_1 + (c_2/2!)(E - E_0) + \dots]. \quad (7)$$

This is solved by the method of successive substitutions. The entire right-hand side of (7) is substituted for E in Eq. (7). To third order, our iterative solution becomes

$$E = E_0 + (1/c_1)[\cos(kd) - c_0] - (c_2/c_1^3)[\cos(kd) - c_0]^2 \\ \times [(c_3 c_1 - 3c_2^2)/6c_1^5][\cos(kd) - c_0]^3 + \dots \quad (8)$$

We apply this when E_0 corresponds to $k = 0$ (bottom of miniband). In this case, $c_0 = 1$.

The preceding formula is potentially more valuable than numerically evaluating the fourth derivative for two reasons. First, the functional dependence of $\chi^{(3)}$ gives greater physical intuition than numerical solutions. Second, taking a fourth derivative (fourth difference) numerically requires very high precision in the computer representation of the numbers for accurate answers.

In the remaining analysis, we are concerned with the separate problem of finding a criterion for validity of the tight-binding, or first order approximation, Eq. (6). However, the same technique could be used to find a validity criterion for the second order approximation of Eq. (8). Equation (3) may be looked at as the scalar product,

$$\cos(kd) = \mathbf{x} \cdot \mathbf{y},$$

$$\text{with } \mathbf{x} = [\cos(2k_2 b), \sin(2k_2 b)] \text{ and } \mathbf{y} = [\cosh(2k_1 a), \\ (\epsilon/2)\sinh(2k_1 a)]. \quad (9)$$

Note that $|x| = 1$ and $|y| > 1$ in (9) for all possible values of a, b, V_0 and E . If $k_1 a \gg 1$ (decay length in barrier \ll barrier width), then in fact $|y| \gg 1$. Since $|\cos(kd)| \leq 1$, \mathbf{x} and \mathbf{y} must be nearly orthogonal. Since \mathbf{x} and \mathbf{y} rotate in opposite directions with increasing E , it follows that Eq. (3) can be satisfied only for a small range of E , over the entire miniband. Thus, one would expect a small variation of $f(E)$ over the entire band, and so $f(E)$ can be replaced by the zeroth and first order terms of its Taylor series expansion. To summarize, replacing the right-hand side of (3) with the zeroth and first order terms of its Taylor series is often justified when $k_1 a \gg 1$. Under these conditions, x and y are nearly perpendicular.

Ultimately, replacing $f(E)$ by the first two terms of its Taylor series can be justified only by comparing the magnitudes of successive terms. Nevertheless, the preceding argument can be made more precise. While the mathematical details are left out, the more precise argument leads to the conclusion that for $\pi \hbar \ll 2(m^* V_0)^{1/2} b$, the tight-binding criterion will be valid when

$$[b^2 m^* V_0 / (\pi \hbar^2)] \sinh^2 [a(8m^* V_0)^{1/2} / \hbar] \gg 1. \quad (10)$$

In what follows, we use the second order approximation from (8). All coefficients of $[\cos(kd) - c_0]^n$, for $n \geq 3$, are set to zero. To this order, we have

$$E = E_0 + (1/c_1)[\cos(kd) - 1] \\ \times [1 - (c_2/2c_1^2)[\cos(kd) - 1]], \quad (11a)$$

$$E' = -(1/c_1)\sin(kd) + (c_2/c_1^3) \\ \times [-\sin(kd) + (1/2)\sin(2kd)], \quad (11b)$$

$$E'' = -(1/c_1)\cos(kd) + (c_2/c_1^3)[- \cos(kd) + \cos(2kd)], \quad (11c)$$

TABLE I. Coefficients of correction terms.

d	$L_1 = 2b$ (Å)	$L_2 = 2a$ (Å)	V_0 (meV)	Comparison with tight binding [Eq. (11e)]		Energy dispersion equation coefficients [Eq. (8)]		
				c_2/c_1^2	$(1 - 3c_2/c_1^2)$	$1/c_1$	$-c_2/2c_1^3$	$(c_3c_1 - 3c_2)/6c_1^5$
100	50	50	320	0.09	0.72	- 3.89	0.18	- 0.013
75	50	25	320	0.24	0.29	- 15.13	1.78	- 0.34
90	50	40	320	0.14	0.57	- 6.87	0.49	- 0.06
110	60	50	320	0.07	0.78	- 2.61	0.10	- 0.006
100	50	5	320	0.33	0.01	- 37.29	6.13	- 1.61

$$E''' = (1/c_1)\sin(kd) + (c_2/c_1^3)[\sin(kd) - 2\sin(2kd)], \quad (11d)$$

$$E'''' = (1/c_1)\cos(kd) + (c_2/c_1^3)[\cos(kd) - 4\cos(2kd)], \quad (11e)$$

where the primes on the left-hand side denote differentiation with respect to kd . These expressions are listed for reference, in case other properties (e.g., group velocity or effective mass) should be investigated by this procedure. It is seen that successive differentiations increase the magnitude of the correction term, as noted previously.

It is natural to ask whether the second order term is sufficient, or whether further terms would be needed. While the validity criterion previously discussed might be used in a rough way, we have included the third and fourth order correction terms corresponding to Eq. (11e).

$$\begin{aligned} E'''' = & (1/c_1)\cos(kd) + (c_2/c_1^3)[\cos(kd) - 4\cos(2kd)] \\ & - [(c_1c_3 - 3c_2^2)/8c_1^5] \\ & \times [5\cos(kd) - 32\cos(2kd) + 27\cos(3kd)] \\ & + [(c_1^2c_4 - 10c_1c_2c_3 + 15c_2^3)/24c_1^7] \\ & \times [7\cos(kd) - 56\cos(2kd) + 81\cos(3kd) \\ & - 32\cos(4kd)]. \end{aligned} \quad (12)$$

Note that at the bottom of the band, $kd = 0$, the third and fourth order correction terms are identically zero. Examination of Eq. (8) shows this to be true for all higher order terms, since they correspond to fourth derivatives of $[\cos(kd) - c_0]^n$, $n \geq 3$. Hence, at the bottom of the band, our new second order approximation to E'''' , and therefore $\chi^{(3)}$, is exact, while the previously used tight-binding approximation is not.

Table I shows the coefficient, c_2/c_1^2 , for two of the superlattice parameters studied earlier. Also, since our earlier study showed that only the bottom of the lowest miniband was occupied at realistic doping levels, we also show the magnitude of the correction factor in Eq. (11e) at $k = 0$, $(1 - 3c_2/c_1^2)$. As is clear from Ref. 1, the calculated $\chi^{(3)}$ is proportional to this term. Several representative cases are shown in the table: a 50 Å-50 Å superlattice with $V_0 = 320$ meV has a small interwell overlap ($t \approx 4.3$ meV) and shows

only a 30% reduction in $\chi^{(3)}$ from its initially predicted two orders of magnitude enhancement; however, when the barrier width is reduced to 25 Å increasing the coupling ($t \approx 21.5$ meV), the correction is 60%. The other cases shown exhibit corrections of similar magnitude. Also listed are the coefficients of the first, second, and third order correction terms in Eq. (8) for the energy dispersion relation. It is seen that the second order correction term c_2/c_1^2 is adequate for all cases examined.

Also shown in the table is a superlattice with a 50-Å well width, but only a 5-Å barrier width and $V_0 = 320$ meV. This case is an extrapolation of the Kronig-Penney model to a very thin (monatomic) barrier, even though the effective mass approximation in the barrier region could not be valid here. In this case, the interwell overlap is enhanced ($t \approx 80$ meV) and departures from tight binding are considerable. The entry in the last column shows $\chi^{(3)}$ to be nearly zero for this case. This is as expected, since the band is relatively wide and nearly parabolic at its minimum. However, if the tight-binding¹ (lowest order) approximation were extrapolated to this case, the last column would be one, and it would incorrectly predict an enhanced $\chi^{(3)}$, since here $\chi^{(3)} \approx td$.⁴

Clearly, more cases and superlattices with different semiconductor combinations need to be investigated. With regard to superlattice fabricated from narrow band-gap semiconductors which exhibit bulk band nonparabolicities (e.g., InSb, InAs), we are presently incorporating this feature into the Kronig-Penney procedure. Finally, as thinner layers are considered, the limits of validity of the present effective mass approximation and consequently, use of the Kronig-Penney model, must be assessed.

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