ENHANCED OPTICAL NONLINEARITIES OF SUPERLATTICES WITHIN THE KRONIG-PENNEY MODEL INCORPORATING INHERENT BULK NONLINEARITIES

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Third order nonlinear optical susceptibilities $\chi^{(3)}$ of GaAs/Ga_{1-x}AlAs superlattices have been predicted which are two orders of magnitude larger than those of bulk GaAs. This enhancement is due to the band nonparabolicity arising from the additional periodicity of the superlattice. These predictions, based on a tight-binding model of the superlattice dispersion, are here extended to the more realistic Kronig-Penney (KP) model. Corrections to tight-binding are non-negligible; however, enhancements of $\chi^{(3)}$ are still large but reduced approximately 30%-50% over previous estimates. The KP model is also here applied to superlattices employing InSb as the quantum well material. Because of the smaller effective mass of InSb, and taking account of its bulk nonparabolicity, the minibands move to higher energy, enhancing the interwell overlap and increasing $\chi^{(3)}$ by about one order of magnitude over that of bulk InSb. The role of the barrier material in this case is important and is discussed. The interplay between the bulk nonparabolicity and that arising from the superlattice is also addressed.

Introduction

In two earlier communications,[1,2] large enhancements of the third order nonlinear optical susceptibility $\chi^{(3)}$ were predicted for GaAs/GaAlAs superlattices due to the band nonparabolicities introduced by the additional periodicity of the superlattice. These earlier predictions were based on a tight-binding model of the electron's energy dispersion in the direction perpendicular to the layers. In the present paper, the validity of the tight-binding (TB) approximation is investigated and corrections are found to it. Assuming the validity of effective mass theory within the layers and abrupt conduction band discontinuities, the electron's dispersion is given by the Kronig-Penney (KP) model. A Taylor series expansion of the KP dispersion relation is developed which gives TB in lowest order and correction terms in higher order. These are found for the energy E(k), and $\chi^{(3)} \sim v'''(k)$. (The velocity $v = E'(k)$ and effective mass $\sim E''(k)^{-1}$ can also be found by this procedure.) Corrections to $\chi^{(3)}$ depend, of course, on the superlattice parameters (well size, barrier width, conduction band discontinuity), are not negligible, and for the GaAs/$Ga_{1-x}Al_x$As superlattices, reduce $\chi^{(3)}$ typically by 30%-50%. Since the original enhancement of the GaAs/$Ga_{1-x}Al_x$As superlattice with respect to bulk GaAs was by a factor $\approx 10^2$, the net enhancement is still appreciable.

The KP model is also applied in the present paper to InSb-based superlattices. The motivation for this is twofold. First, the component of electric field in the plane of the layers will yield a nonlinear contribution, in view of the bulk nonparabolicity of InSb; this was not true for GaAs, whose lower conduction band is quite parabolic. Secondly, the smaller band effective mass m* of InSb as compared to GaAs (0.013 m_e vs. 0.068 m_e) causes the minibands to move to higher energies, enhancing the interwell overlap and, hence, $\chi^{(3)} \sim v'''$. However, for the recently investigated InSb/CdTe system, the conduction band discontinuity is ~ 450 meV (320 meV for GaAs/$Ga_{0.7}Al_{0.3}$As) and $m^*(CdTe) = 0.14\ m_e$. These two effects, especially the latter, result in a more rapid spatial decay of the electron envelope function in the CdTe barrier region, reducing the interwell overlap and largely cancelling the enhancement effect of the smaller m* of InSb. However, for superlattices in which the effective mass of the barrier material is comparable to that of InSb (InSb/HgCdTe, InSb$_{1-x}$Be$_x$/InSb, InSb/PbTe, InSbAs/InSb are possible superlattice combinations), the enhancement in $\chi^{(3)}$ is about an order of magnitude larger than for bulk InSb. In all these applications, the bulk nonparabolicity

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of InSb must be taken into account in the Kronig-Penney solution for the superlattice. This is shown in the present paper, and the effect of the bulk nonparabolicity on superlattice behavior is discussed.

Physical Model and Formulation

As pointed out previously, the enhancement in $\chi^{(3)}$ is due to mobile electrons in nonparabolic (dispersive) energy bands, 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 relevant at realistic doping levels), 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. In the case of the GaAs/$Ga_{1-x}Al_xAs$ superlattice, a large, third order nonlinear susceptibility $\chi^{(3)}$ is obtained which is comparable to that of bulk InSb,[3] and approximately two orders of magnitude larger than that of bulk GaAs. In performing this calculation, the electron's energy-momentum relation in the direction perpendicular to the layers was initially taken to be of tight-binding form (sinisordal approximation):

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

with 2t the (lowest) miniband width, d the superlattice period, and k the quasi-momentum perpendicular to the layers.

However, the tight-binding form is only an approximation valid for weakly interacting quantum wells, i.e., wide barriers and large conduction band discontinuities. 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 further assumed that the conduction band discontinuities are abrupt, then the electronic motion perpendicular to the layers is given by the Kronig-Penney (KP) model. In an earlier paper,[2] the KP model was used to estimate $\chi^{(3)}$, which is proportional to the third derivative of the electron's group velocity with respect to k or, equivalently, to the fourth derivative of the energy dispersion. These quantities are given as a power series expansion, the leading term of which is of tight-binding form, while higher order terms represent corrections to tight-binding. Specifically, the dispersion relation of the electron is written as a sum of the energy in the plane of the layers (momentum $\underline{K}$) and perpendicular to the layers (momentum k):

$$E(\underline{K},k) = \hbar^2K^2/2m^* + E(k) \qquad (1)$$

where a parabolic effective mass approximation is made for electron motion parallel to the layers, while E = E(k) is a solution of the one-dimensional Kronig-Penney problem

$$\cos(kd) = \cosh(2k,a)\cos(2k_2b) + (\varepsilon/2)\sinh(2k,a)\sin(2k_2b), \qquad (2)$$

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

$$k_1 + [2m^*_1 (V_0-E) / \hbar^2]^{1/2},$$
$$k_2 = [2m^*_2 E/\hbar^2]^{1/2},$$
$$\varepsilon = (k_1/k_2 - k_2/k_1), \qquad (3)$$

and V_0 is the conduction-band energy discontinuity.

The procedure used to solve Eq.(2) for E(k) and its higher derivatives is to expand its right-hand side f(E) in a power series about $E = E_0$ corresponding to the bottom of the band k = 0 (though the expansion can be done about any point in the band),

$$\cos(kd) = c_0 + c_1(E-E_0) + c_2/2! (E-E_0)^2 + .., \qquad (4)$$

where, in this case, $c_0 = 1$ and $c_k = (\partial^k f/\partial E_k) \mid_{E=E_0}$. The justification for this procedure is given in some detail in ref.(2). Solving for E(k) by the method of successive substitution (i.e., iteratively), the quantities of specific interest are

$$E = E_0 + \frac{1}{c_1}[\cos(kd)-1] - \frac{c_2}{c_1^2}[\cos kd-1]^2$$
$$E'''' = \frac{1}{c_1}\cos(kd) + \frac{c_2}{c_1^3}[\cos(kd)-4\cos(kd)] + E_c'''' \qquad (5)$$

where the prime denotes differentiation with respect to (kd). The correction term E_c'''' rigorously vanishes if the expansion is at k = 0, though not elsewhere. For details, see ref.(2). Thus,

$$E''''(k = 0) = \frac{1}{c_1}\left(1 - 3\frac{c_2}{c_1^2}\right). \qquad (6)$$

Before proceeding to our results, we indicate the generalization of Eq.(2) and Eq.(3) to the case in which the quantum well material possesses intrinsic bulk nonparabolicities. The modification is simply to replace k_2 of Eq.(3) by

$$k_2 = \frac{1}{\hbar}\sqrt{2m^*_2\left(\frac{E^2 + E\cdot E_g}{E_g}\right)} \qquad (7)$$

where E_g is the bulk bandgap of InSb (taken to be 160 meV). The parabolic case [Eq.(3)] may be recovered by taking $E_g \to \infty$. ($E_g >> E$), and this test was used to study the effect of the bulk nonparabolicity on the superlattice dispersion and $\chi^{(3)}$.

	d(Å)	L_1(Å)	L_2(Å)	$(-1/c_1)$	$1-3\frac{c_2}{c_1^2}$	$\left(-\frac{1}{c_1}\right)\left(1-3\frac{c_2}{c_1^2}\right)$	$\frac{\chi^{(3)}(SL)}{\chi^{(3)}(InSb)}$
$GaAs/Ga_{.3}Al_{.7}As$	75	50	25	14.8	0.3	4.4	0.19
InSb/CdTe	75	50	25	(99.4) 11.6	(0.03) 0.511	(3) 6	(0.13) 0.26
	55	50	5	(192) 57.5	(0.0076) 0.64	(1.46) 37	---- 0.45
'InSb/InSb'	75	50	25	(101.6) 54	(0.03) 0.906	(3) 49	(0.13) 2.18
	55	50	5	(194) 140	(7.5×10^{-4}) 3.49	(0.145) 489	---- 6
	60	50	10	102.5	2.08	213	4
	75	70	5	80.5	2.19	176.6	8

Entries for Nonlinear Susceptibility Ratios

Results and Discussion

Table 1 displays $(-1/c_1)$, the effective half-bandwidth in the KP model [cf. Eq.(5)], the KP correction factor $(1 - 3\ c_2/c_1^2)$, their product given by Eq.(6), and the susceptibility ratio $\chi^{(3)}(SL)\ /\ \chi^{(3)}(InSb)$. These are given for two superlattice configurations specified by L_1 = well width, and L_2 = barrier width ($d = L_1 + L_2$ is the superlattice period). Several other configurations were investigated, but the results given are sufficiently illustrative. For the $GaAs/Ga_{0.7}Al_{0.3}As$ superlattice, $m^*_1 = m^*_2 = 0.068\ m_e$, and the conduction band discontinuity (barrier height) is $V_0 = 320$ meV. The same results are shown for the InSb/CdTe superlattice, where $V_0 = 450$ meV[4], $m^*_1 = 0.14\ m_e$, and $m^*_2 = 0.013\ m_e$ (1 denotes the barrier region, 2 the well). Here, the bulk bandgap of InSb is taken to be $E_g = 160$ meV, while the entries in parentheses refer to the case $E_g \to \infty$, where the bulk nonparabolicity is removed [cf. Eq.(7)]. Finally, the same results are shown for a superlattice comprised of InSb and a second barrier/well material having the same effective mass $m^*_1 = m^*_2 = 0.013\ m_e$. Again, $V_0 = 450$ meV, $E_g = 160$ meV and the limit $E_g \to \infty$ is taken.

For the GaAs/GaAlAs superlattices, the 50 Å - 25 Å superlattice has a $\chi^{(3)}$ 0.19 that of bulk InSb. The $\chi^{(3)}$ of bulk InSb is ~ 10^2 larger than that of bulk GaAs[3], so an enhancement by a factor ~ 20 is realized over bulk GaAs for this case. It is seen that the role of KP correction factors, which represent departures from tight-binding theory, is crucial in this and the following two cases.

For the InSb/CdTe system, the susceptibility ratios are small and are not enhanced. The reason for this is the relatively large effec-

tive mass of the CdTe barrier region, 0.14 m_e, a factor of 10 larger than that of InSb. This results in a more rapid spatial decay of the electronic wavefunction in the barrier region, decreasing the overlap and $\chi^{(3)}$. For the two cases presented, it is seen that the bulk nonparabolicity acts to enhance $\chi^{(3)}$ of the superlattice, though this is not true in all cases examined. The interplay between the two nonparabolicities involves subtleties that are parameter-dependent and must be handled with caution. Further study of this feature is required.

Finally, the susceptibility enhancements are given for InSb superlattices in which the barrier material has a small effective mass comparable to that of InSb. Possible candidates are InSb/PbTe, InSb/$InSb_{1-x}Bi_x$, InSb/InAsSb, and InSb/InGaSb, the latter two being strained-layer-superlattices. Specifically, we consider the InSb/$InSb_{1-x}Bi_x$ superlattice where the addition of Bi results in a shrinkage[4] of the bulk bandgap E_g (from 160 meV for x = 0 to 125 meV for x = 0.03). This results in a decrease in m* since, according to the Kane band theory, $m^* \approx A E_g$. However, not knowing m* for the alloy, we take it to be the same as for InSb. Also, we take the conduction band offset to be the same as for the InSb-CdTe superlattice, 450 meV. From the table, it is seen that enhancements approaching one order of magnitude are obtained. Again, the bulk nonparabolicity enhances $\chi^{(3)}$ for the cases presented. Although the 5 Å and 10 Å barriers are perhaps unrealistically small (effective mass theory is questionable here), they have the unique feature that c_2 is negative. This means that KP corrections to TB act to enhance $\chi^{(3)}$, and this occurs only when the bulk nonparabolicity is included. We want to understand this interplay better, and to extend the calculations to other configurations and/or semiconductor combinations in a search for larger enhancements. It is pointed out that only the (semiclassical) contribution from the lowest subband to $\chi^{(3)}$ has been considered; intersubband transitions which would occur at higher frequencies would require extension of the theory.

References

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