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# Optical interface phonon modes in graded quantum well structures: perturbation theory

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**Abstract.** The behaviour of the optical interface phonon in the graded quantum well (GQW) of $Al_xGa_{1-x}As$ with infinite $Al_{0.4}Ga_{0.6}As$ barrier layers is investigated based on the dielectric continuum model. The solutions of the optical interface phonon modes are obtained. The perturbation theory is developed for $kL \ll 1$. It is found that the properties of the optical interface phonon in the GQW for $kL \ll 1$ are quite different from those in the square quantum well (SQW), as are those of GaAs-type modes from those of AlAs-type ones.

## 1. Introduction

With the techniques of molecular-beam epitaxy (MBE) and metallorganic chemical vapour deposition (MOCVD), one can grow layered structures, such as that of $Al_xGa_{1-x}As$, with very precisely controlled thickness of layers and content $x$ of aluminum. The various superlattices and quantum well structures have been made artificially.

Since the first observation of a new transient electrical polarization phenomenon in sawtooth superlattices of $Al_xGa_{1-x}As$ [1], the graded-gap structures have attracted great attention [2–10] for their possible device applications. Various authors have investigated the properties of electron and exciton states in a graded quantum well (GQW) structure, in which the conduction- and valence-band edges vary linearly along the growth direction in the well layer. Their studies have shown that high device performance may be achieved in such a structure. But so far little attention has been paid to the phonon properties. It is important to specify the characteristics of the phonons in GQWs, especially that of the interactions of the phonons with electrons.

Based on both the data of [11] and the dielectric continuum model [12] which has given good representations of phonon modes [13–17], we study optical interface phonon in the graded quantum well $Al_xGa_{1-x}As$ with infinite $Al_{0.4}Ga_{0.6}As$ barrier layers. The rest of this paper is set out as follows. The equations of motion for the optical interface phonon modes in the GQW are given and its general solutions are obtained in section 2. Then its perturbation theory for $kL \ll 1$ is developed in section 3. Finally, in section 4, the numerical calculations for the perturbation solutions are carried out; the main characteristics of the phonons are discussed.

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## 2. Equations of motion for the phonon eigenmodes and their solutions

Let us consider an isolated GQW of $Al_xGa_{1-x}As$ between two infinite $Al_{0.4}Ga_{0.6}As$ barrier layers. The Al content $x$ of $Al_xGa_{1-x}As$ varies continuously from 0 to $x_0$ ($x_0 < 0.4$) along the growth direction that is taken as the $z$-axis,

$$x = (x_0/L)z \qquad (0 \leqslant z \leqslant L) \tag{1}$$

where $L$ is the width of the well region.

Within the framework of the dielectric continuum model [12], Maxwell equations lead to the coupled integral equation of motion for polarization eigenmodes [13]. Introducing the electric field component $E_y(\boldsymbol{k}, z)$ and the electric displacement component $D_z(\boldsymbol{k}, z)$, where $\boldsymbol{k}$ is the in-plane component of the phonon wavevector, and differentiating the coupled integral equation with respect to $z$, we obtained [7]

$$\begin{cases} \dfrac{\mathrm{d}E_y(z)}{\mathrm{d}z} = \dfrac{\mathrm{i}kD_z(z)}{\varepsilon(z)} \\ \dfrac{\mathrm{d}D_z(z)}{\mathrm{d}z} = -\mathrm{i}k\varepsilon(z)E_y(z) \end{cases} \tag{2}$$

with the dielectric function

$$\varepsilon(\omega) = \varepsilon_\infty \left( \frac{\omega_{Li}^2 - \omega^2}{\omega_{Ti}^2 - \omega^2} \right). \tag{3}$$

It is also a function of $x$ (i.e. $z$; cf (26)–(30) and (1)) in the well region, where the Lyddane–Sachs–Teller relation is assumed. In equation (3) $\omega_{Li}$ and $\omega_{Ti}$ are corresponding LO and TO phonon frequencies with $i = 1$ and $i = 2$ for GaAs and AlAs types of $Al_xGa_{1-x}As$, respectively. According to [11] the parameters of $Al_xGa_{1-x}As$ are based on an interpolation scheme between that of GaAs and AlAs, such as $\varepsilon_\infty$, $\omega_{Li}$ and $\omega_{Ti}$. The decoupled differential equations can be obtained from equations (2):

$$\begin{cases} \dfrac{\mathrm{d}^2E_y(z)}{\mathrm{d}z^2} + \dfrac{1}{\varepsilon(z)}\dfrac{\mathrm{d}\varepsilon(z)}{\mathrm{d}z}\dfrac{\mathrm{d}E_y(z)}{\mathrm{d}z} - k^2E_y(z) = 0 \\ \dfrac{\mathrm{d}^2D_z(z)}{\mathrm{d}z^2} - \dfrac{1}{\varepsilon(z)}\dfrac{\mathrm{d}\varepsilon(z)}{\mathrm{d}z}\dfrac{\mathrm{d}D_z(z)}{\mathrm{d}z} - k^2D_z(z) = 0. \end{cases} \tag{4}$$

In order to obtain the components of the eigenmode of the optical phonon in the GQW, the electric field $E_y(z)$ and the displacement $D_z(z)$, we have to solve equations (2) or equations (4) in all the regions. At first we solve equations (2) in the well region.

Equivalently we write (2) as follows:

$$\frac{\mathrm{d}}{\mathrm{d}z}\begin{pmatrix} E_y(z) \\ D_z(z) \end{pmatrix} = -\mathrm{i}S(z)\begin{pmatrix} E_y(z) \\ D_z(z) \end{pmatrix} \tag{5}$$

with

$$S(z) = k\begin{pmatrix} 0 & -1/\varepsilon \\ \varepsilon & 0 \end{pmatrix}. \tag{6}$$

Introducing $U(z, 0)$:

$$\begin{pmatrix} E_y(z) \\ D_z(z) \end{pmatrix} = U(z, 0)\begin{pmatrix} E_y(0) \\ D_z(z) \end{pmatrix} \tag{7}$$

where both $E_y(0)$ and $D_z(0)$ are the values of $E_y(z)$ and $D_z(z)$ at $z = 0$ respectively. It is evident that if $U(z, 0)$ is known, $E_y(z)$ and $D_z(z)$ are also known easily.

From (7) we have

$$\frac{\mathrm{d}U(z,0)}{\mathrm{d}z} = -\mathrm{i}S(z)U(z,0). \tag{8}$$

Equation (8) can be solved explicitly by using the Neumann–Liouville expansion [18] as

$$U(z,0) = \sum_{n=0}^{\infty} U^{(n)}(z,0) \tag{9}$$

where

$$U^{(0)}(z,0) = 1 \tag{10}$$

$$U^{(n)}(z,0) = (-\mathrm{i})^n \left( \prod_{l=1}^{n} \int_0^z \mathrm{d}z_l \theta(z_{l-1} - z_l) S(z_l) \right) \qquad n \neq 0. \tag{11}$$

$\theta(z_0 - z_1) \equiv 1$; $\theta(z) = 1$ for $z > 0$, and $\theta(z) = 0$ otherwise. Then (7) can be written as

$$\begin{pmatrix} E_y(z) \\ D_z(z) \end{pmatrix} = \sum_{n=0}^{\infty} U^{(n)}(z,0) \begin{pmatrix} E_y(0) \\ D_z(0) \end{pmatrix}. \tag{12}$$

In order to determine $E_y(0)$ and $D_z(0)$ in the solution (12), we have to solve equations (2) or (4) outside the GQW. We solve equations (4) and obtain the solutions as follows:

$$\begin{cases} E_y(z) = A \exp(kz) \\ \\ D_z(z) = -\mathrm{i}\varepsilon_{out} A \exp(kz) \end{cases} \qquad (z \leqslant 0) \tag{13}$$

$$\begin{cases} E_y(z) = B \exp(-kz) \\ \\ D_z(z) = -\mathrm{i}\varepsilon_{out} B \exp(-kz) \end{cases} \qquad (z \geqslant L) \tag{14}$$

where $\varepsilon_{out}$ is the dielectric function in $Al_{0.4}Ga_{0.6}As$ barrier layers. So we obtain

$$\begin{pmatrix} E_y(z) \\ D_z(z) \end{pmatrix} = E_y(0)U(z,0) \begin{pmatrix} 1 \\ -\mathrm{i}\varepsilon_{out} \end{pmatrix} \tag{15}$$

and the dispersion relation:

$$\varepsilon_{out}^2 U_{12}(L,0) - U_{21}(L,0) + \mathrm{i}\varepsilon_{out}[U_{22}(L,0) + U_{11}(L,0)] = 0 \tag{16}$$

representing the relation between $k$ and $\omega$, where $U_{ij}$ $(i, j = 1, 2)$ come from the matrix

$$U(z,0) = \begin{pmatrix} U_{11}(z,0) & U_{12}(z,0) \\ U_{21}(z,0) & U_{22}(z,0) \end{pmatrix}. \tag{17}$$

Notice that $U_{ij}$ are functions of $k$ and $\omega$, and $\varepsilon_{out}$ is a function of $\omega$.

## 3. Perturbation theory for $kL \ll 1$

In principle, our results (9), (15) and (16) can exactly hold for the problems discussed. However, this means that one needs to perform an infinite number of integrals, which, obviously, are impossible. Therefore, in practice, we have to make some approximations up to the required order. For example, we obtain the first order of perturbation from (9):

$$U(z,0) \approx 1 - \mathrm{i} \int_0^z \mathrm{d}z_1 S(z_1) \tag{18}$$

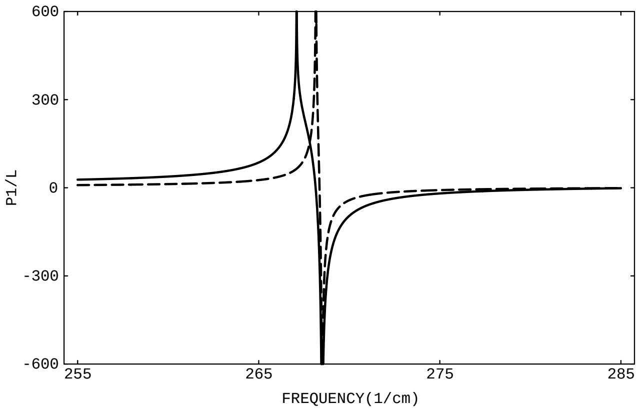


**Figure 1.** Function $P_1(\omega)/L$ versus the frequency $\omega$ with $z/L = 0.3$ (dashed curves) and $z/L = 1$ (solid curves), where $x_0$ is taken to be 0.2.

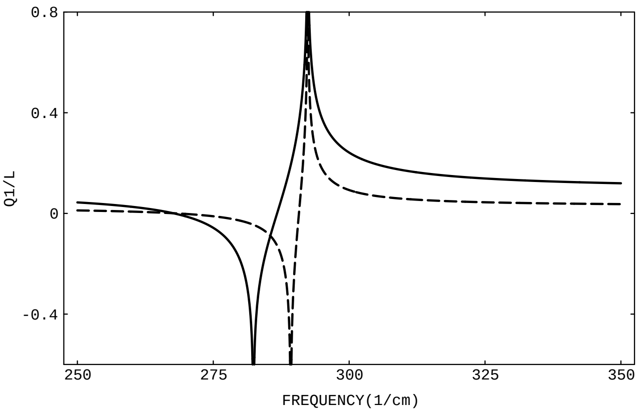


**Figure 2.** Function $Q_1(\omega)/L$ versus the frequency $\omega$ with $z/L = 0.3$ (dashed curves) and $z/L = 1$ (solid curves), where $x_0$ is taken to be 0.2.

for $kL \ll 1$. It can be written as follows:

$$U(z,0) \approx T \exp\left\{ -\mathrm{i} \int_0^z \mathrm{d}z_1 S(z_1) \right\} \tag{19}$$

where $T$ represents the $z$-ordered product. Here $kL \ll 1$, so we have

$$U(z,0) = \exp\left\{ -\mathrm{i}k \begin{pmatrix} 0 & -Q(z,\omega) \\ P(z,\omega) & 0 \end{pmatrix} \right\} \tag{20}$$

where

$$P(z,\omega) = \int_0^z \varepsilon(z_1,\omega)\,\mathrm{d}z_1 \tag{21}$$

$$Q(z,\omega) = \int_0^z \frac{\mathrm{d}z_1}{\varepsilon(z_1,\omega)}. \tag{22}$$

$P(z,\omega)$ and $Q(z,\omega)$ are $P_1(z,\omega)$ and $Q_1(z,\omega)$ or $P_2(z,\omega)$ and $Q_2(z,\omega)$ for the GaAs or AlAs type of $Al_xGa_{1-x}As$ (cf figures 1–4), respectively. $\varepsilon(z,\omega)$ is the dielectric function of

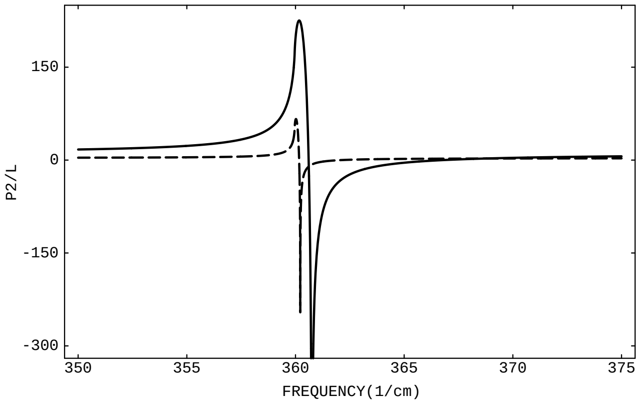


**Figure 3.** Function $P_2(\omega)/L$ versus the frequency $\omega$ with $z/L = 0.3$ (dashed curves) and $z/L = 1$ (solid curves), where $x_0$ is taken to be 0.2.

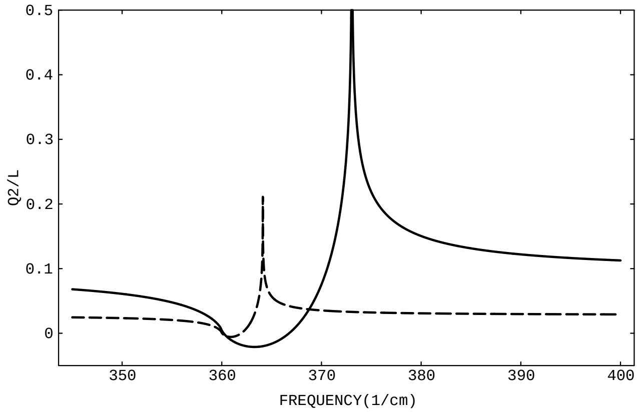


**Figure 4.** Function $Q_2(\omega)/L$ versus the frequency $\omega$ with $z/L = 0.3$ (dashed curves) and $z/L = 1$ (solid curves), where $x_0$ is taken to be 0.2.

$Al_xGa_{1-x}As$ in the well; it is determined by (3) and (26)–(30). From above we obtain

$$U(z,0) = \cosh[k\sqrt{P(z,\omega)Q(z,\omega)}] - \frac{\mathrm{i}}{\sqrt{P(z,\omega)Q(z,\omega)}}\begin{pmatrix} 0 & -Q(z,\omega) \\ P(z,\omega) & 0 \end{pmatrix} \times \sinh[k\sqrt{P(z,\omega)Q(z,\omega)}] \tag{23}$$

and finally the electric field and electric displacement components are obtained,

$$\begin{cases} E_y(z) = E_y(0)[\cosh(k\sqrt{P(z,\omega)Q(z,\omega)}) + \varepsilon_{out}(\omega)\sqrt{Q(z,\omega)/P(z,\omega)} \\ \quad \sinh(k\sqrt{P(z,\omega)Q(z,\omega)})] \\ D_z(z) = -\mathrm{i}E_y(0)[\varepsilon_{out}(\omega)\cosh(k\sqrt{P(z,\omega)Q(z,\omega)}) + \sqrt{P(z,\omega)/Q(z,\omega)} \\ \quad \sinh(k\sqrt{P(z,\omega)Q(z,\omega)})]. \end{cases} \tag{24}$$

The dispersion relation of the optical interface phonon in the GQW is

$$\tanh(k\sqrt{P(L,\omega)Q(L,\omega)}) = -\frac{2\varepsilon_{out}(\omega)\sqrt{Q(L,\omega)/P(L,\omega)}}{\varepsilon_{out}(\omega)^2[Q(L,\omega)/P(L,\omega)] + 1}. \tag{25}$$

## 4. Numerical calculations and discussion for the perturbation solutions

We calculate (24) and (25) numerically by using the optical dielectric constant [11]

$$\varepsilon_\infty(x) = 10.89 - 2.73x \tag{26}$$

and corresponding LO and TO phonon frequencies (in units of $\mathrm{cm}^{-1}$) for GaAs and AlAs types of $\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}$ [11]

$$\omega_{L1}(x) = 292.3736 - 52.8289x + 14.4372x^2 \tag{27}$$

$$\omega_{T1}(x) = 268.4998 - 5.1619x - 9.356\,06x^2 \tag{28}$$

and

$$\omega_{L2} = 359.9623 + 70.8149x - 26.7773x^2 \tag{29}$$

$$\omega_{T2} = 359.9623 + 4.4360x - 2.4196x^2 \tag{30}$$

respectively.

Calculations show that there are some properties of perturbation solutions of the optical interface phonon modes in the GQW which are different from those in the SQW, although there are some the same.

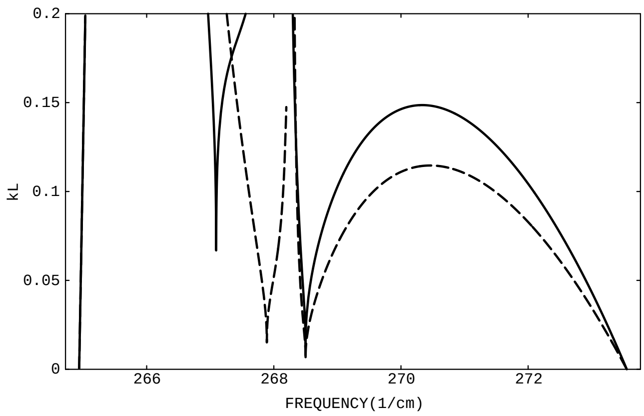


**Figure 5.** Dispersion relations of the optical interface phonon in an $\mathrm{Al}_{0.4}\mathrm{Ga}_{0.6}\mathrm{As}/\mathrm{Al}_x\mathrm{Ga}_{1-x}\mathrm{As}/\mathrm{Al}_{0.4}\mathrm{Ga}_{0.6}\mathrm{As}$ GQW for GaAs-type modes with $x_0 = 0.1$ (dashed curves) and $x_0 = 0.2$ (solid curves). It can be seen that the dispersion relation depends on the gradient of the GQW.

Above all, let us see the frequency responses in the GQW. Figures 5 and 6 show the dispersion relations of phonons for GaAs- and AlAs-type modes in the GQW, respectively. Firstly, they are the same as those in the SQW [10] in that there are non-single values of frequency for $kL$ on the dispersion curves in the GQW. This is because the solutions of equations (2) (i.e. the Maxwell equations) must satisfy the condition of boundaries at both $z = 0$ and $z = L$ of these structures. Actually we can see that if they are extended to the non-physical region, the dispersion curves in the GQW are similar to those in the SQW. What is evident is that the dispersion curves in the GQW are more complicated than those in the SQW. This is because of the dependency of structures. They will tend to the simpler shape like that in the SQW when $x_0 \to 0$ (cf figures 11, 12). In contrast to the case in the SQW, there are not evident narrow maximum values of $k$ but minimum ones. From figures 5 and 6 we can also see that all interface modes exist between $\omega_T(0.4)$ and $\omega_L(0.4)$, which are 264.94 $\mathrm{cm}^{-1}$ and

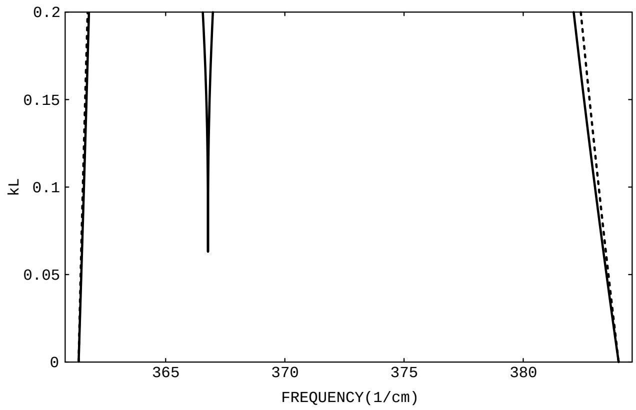


**Figure 6.** Dispersion relations of the optical interface phonon in an $Al_{0.4}Ga_{0.6}As/Al_xGa_{1-x}As/Al_{0.4}Ga_{0.6}As$ GQW for AlAs-type modes with $x_0 = 0.1$ (dashed curves) and $x_0 = 0.2$ (solid curves). It can be seen that the dispersion relation depends on the gradient of the GQW.

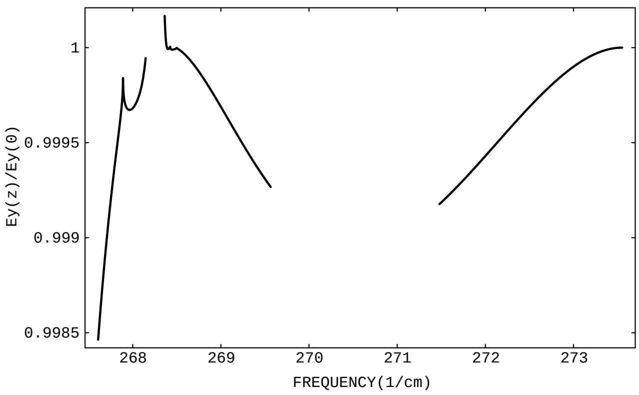


**Figure 7.** Electric field components $E_y$ of optical interface modes in the $Al_{0.4}Ga_{0.6}As/Al_xGa_{1-x}As/Al_{0.4}Ga_{0.6}As$ GQW of GaAs-type versus the frequency $\omega$, where $x_0$ and $z/L$ are taken to be 0.1 and 0.3 respectively.

273.55 $cm^{-1}$ for GaAs-type modes (figure 5) and 361.35 $cm^{-1}$ and 384.00 $cm^{-1}$ for AlAs-type modes (figure 6). Moreover for AlAs-type modes (figure 6) there are three narrow frequency regions existing of the dispersion relation curves. This means that the interface AlAs-type modes exist only in these narrow frequency regions.

It is shown that the frequency responses of electric field components $E_y$ in the GQW (figures 7, 8) are quite different from those in the SQW (figure 9), and especially they vary very slightly with the frequency $\omega$, but those in the SQW vary very greatly.

On the other hand, the values of $E_y$ in the GQW (figure 10) are little changed along with $z$. Nevertheless in the SQW they are greatly changed; sometimes the absolute values of $E_y$ change from 0 to a maximum. Moreover the curves of $E_y$ in the GQW have lost the symmetries (i.e., symmetric and antisymmetric properties) which the curves of $E_y$ in the SQW possessed. This is reasonable because of the asymmetric structure.

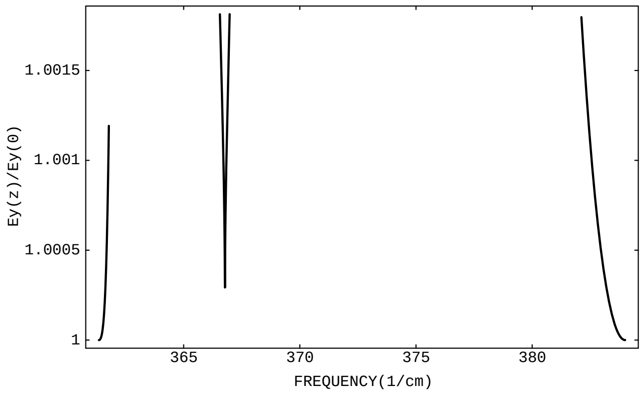


**Figure 8.** Electric field components $E_y$ of optical interface modes in the $Al_{0.4}Ga_{0.6}As/Al_xGa_{1-x}As/Al_{0.4}Ga_{0.6}As$ GQW of AlAs type versus the frequency $\omega$, where $x_0$ and $z/L$ are taken to be 0.1 and 0.3 respectively.

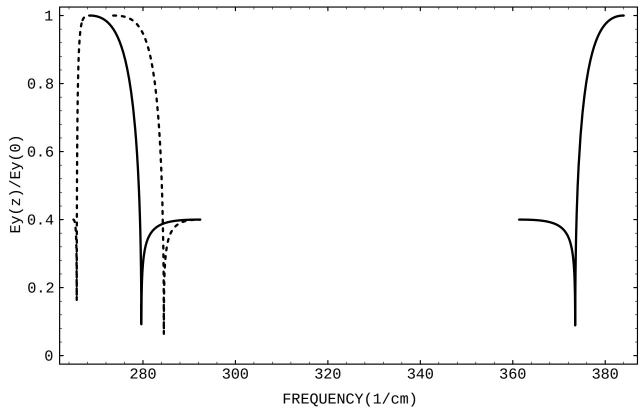


**Figure 9.** Electric field components $E_y$ of the optical interface modes in the SQW versus the frequency $\omega$, where $z/L$ is taken to be 0.3. The dashed and solid curves refer to that for GaAs and AlAs types of $Al_{0.4}Ga_{0.6}As$ respectively.

Finally, of course, the dispersion relations and $E_y$ in the GQW are also dependent on the gradient of the GQW (figures 5, 6, 10). This is a peculiar characteristic of the GQW and it increases the selectivity, so that may be significant for applications.

It should be noticed that the properties of the optical interface phonon in the GQW for GaAs-type modes are quite different from those for AlAs-type modes, which is similar to the case in the SQW. For example, the dispersion relations for AlAs-type modes are in three narrow frequency regions but those for GaAs-type ones are not, and so on. These are because the dependencies of $\omega_{L1}(x)$, $\omega_{T1}(x)$ and $\omega_{L2}(x)$, $\omega_{T2}(x)$ on $x$ (cf (27), (28) and (29), (30)) are different from each other. So are the frequency responses of $E_y$. This is also significant for applications.

It must be pointed out that when $x_0 \to 0$ the dispersion curves of the phonon in the GQW for GaAs type tend to those in the SQW more slowly than for the AlAs type. Calculation shows (figures 11, 12) that when $x_0 = 0.02$, the dispersion curves of the phonon in the GQW for the AlAs type are very close and very similar to those in the SQW, but for the GaAs type they

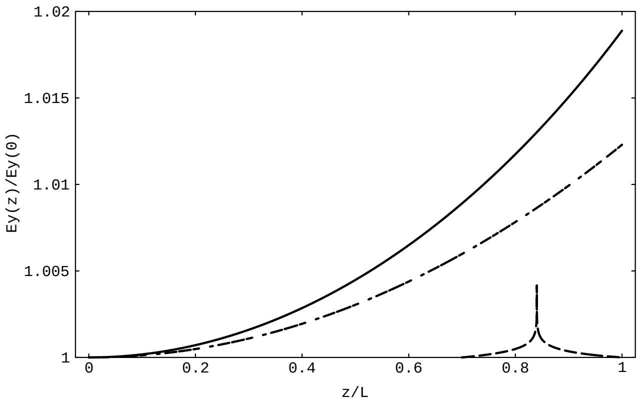


**Figure 10.** Electric field components $E_y$ of optical interface modes in the $Al_{0.4}Ga_{0.6}As/Al_xGa_{1-x}As/Al_{0.4}Ga_{0.6}As$ GQW versus $z$. The dashed curves refer to that for GaAs-type modes with $x_0 = 0.2$ and $\omega = 268$ cm$^{-1}$. The solid and dashed–dotted curves refer to that for AlAs-type modes with $x_0 = 0.2$ and $x_0 = 0.1$ respectively; the frequency of both is 382.5 cm$^{-1}$. Their dependence on the gradient of the GQW is evident.

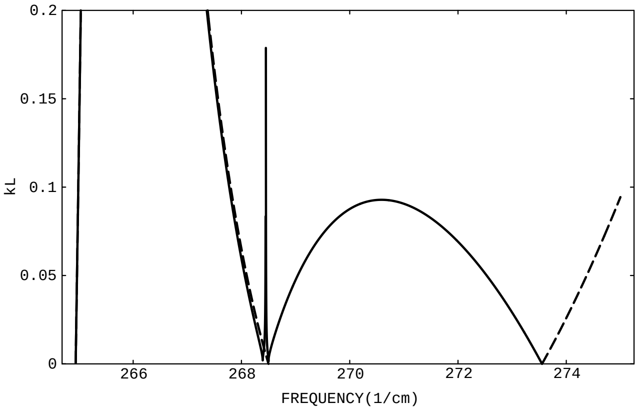


**Figure 11.** Dispersion relations of optical interface phonon in $Al_{0.4}Ga_{0.6}As/Al_xGa_{1-x}As/Al_{0.4}Ga_{0.6}As$ GQW for GaAs-type modes with $x_0 = 0.02$ (solid curves). The dashed curves refer to that in the $Al_{0.4}Ga_{0.6}As/GaAs/Al_{0.4}Ga_{0.6}As$ SQW. Two lines on the left of the dispersion curves of the phonon in the GQW coincide with those in the SQW in the main. It can be seen that when $x_0 \to 0$ the dispersion curves of the phonon in the GQW tend to those in the SQW.

are somewhat different; this can be explained by the fact that $Al_xGa_{1-x}As$ plays an important role even it is a smaller rate. It is worth while noticing that when $x_0 \to 0$, $\varepsilon(x) \to \varepsilon_G$, i.e. $Al_xGa_{1-x}As \to GaAs$, for GaAs type; but when $x_0 \to 0$, $\varepsilon(x) \to \varepsilon_\infty$ (not $\varepsilon_G$) for AlAs type and this actually coincides with the fact that there is no GaAs of AlAs type, although $Al_xGa_{1-x}As$ of AlAs type does exist. But it can be seen from figure 12 that the dispersion curves in the GQW for AlAs type tend to those in the SQW very well in the region of the dispersion curves. Where $\varepsilon_G$ is the dielectric function of GaAs, $\varepsilon_\infty$ is the high-frequency dielectric constant (i.e., the optical dielectric constant).

When $x_0 \to 0$ the convergence of equations (24) and (25) is also evident since then $S(z)$ does not depend on $z$ and $U(z, 0)$ is easily integrated.

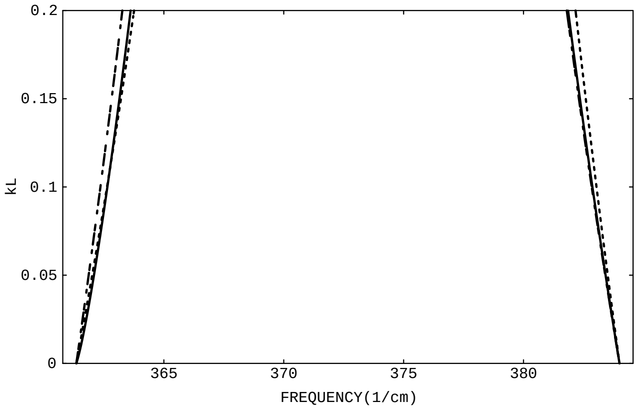


**Figure 12.** Dispersion relations of the optical interface phonon in the $Al_{0.4}Ga_{0.6}As/Al_xGa_{1-x}As/Al_{0.4}Ga_{0.6}As$ GQW for AlAs-type modes with $x_0 = 0.02$ (solid curves). The dotted curves refer to those in the $Al_{0.4}Ga_{0.6}As/GaAs/Al_{0.4}Ga_{0.6}As$ SQW in which the dielectric function of GaAs is $\varepsilon_G$; the dashed and dotted ones refer to those in the $Al_{0.4}Ga_{0.6}As/GaAs/Al_{0.4}Ga_{0.6}As$ SQW in which the dielectric function of 'GaAs' is $\varepsilon_\infty$. It can be seen that some segments of solid ones coincide with dotted or dashed and dotted. This shows that when $x_0 \to 0$ the dispersion curves of the phonon in the GQW tend to those in the SQW.

The discussion on the electric displacement component $D_z$ is similar to that on the electric field $E_y$.

The properties of the GQW can be verified by the method of Raman scattering as in [19, 20].

Since the interface phonon scattering mechanism is an important scattering mechanism in quantum well systems [16, 17], it can be expected that a high mobility may be achieved in a GQW structure with the gradient, the width and the type of $Al_xGa_{1-x}As$ chosen properly.

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