Two-photon absorption in semiconducting quantum-well structures

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A theoretical treatment of two-photon absorption is presented for quasi-one-dimensional (Q1D) and quasi-two-dimensional (Q2D) quantum-well structures. It is found that the absorption depends upon the polarization of the radiation field relative to the direction of confinement of the carriers. For radiation polarized perpendicular to the direction of confinement, the absorption is suppressed relative to its value in a bulk semiconductor with the suppression being greater for Q1D than for Q2D structures. When the radiation is polarized along the direction of confinement, the absorption coefficient is enhanced over its value in the bulk. The two-photon absorption in quantum-well systems is found to be a nonmonotonic function of the photon energy with new structure occurring whenever transitions can occur between new pairs of quantized subbands. The threshold for two-photon absorption is shifted towards higher photon energies by the confinement of the carriers.

I. INTRODUCTION

With the recent development of semiconducting quantum-well structures, there has been a growing interest in the optical properties of these structures.[1-8] Most of the interest has focused on the linear optical properties of these structures although there has been some work on the nonlinear optical properties of semiconducting superlattice structures.[9-12] However, not much work has been done on the nonlinear optical properties of single-quantum-well structures or multiple-quantum-well structures in which there is little coupling between the carriers confined in adjacent wells.

Among the nonlinear optical properties which are of interest in bulk semiconductors is their two-photon absorption. If a semiconductor is irradiated by photons whose energy is less than the band-gap energy of the material, then the photons cannot be absorbed (in the absence of defects or free carriers) in a process which involves only a single photon. However, such photons can be absorbed in a two-photon process in which an electron can make a transition from a valence-band to a conduction-band state via a process which involves an intermediate state.[13-16] The absorption coefficient for such a process is proportional to the intensity of the radiation field and therefore such processes will only be important when the material is irradiated by intense fields of photon energies greater than half the band-gap energy such as those obtained from a laser. We wish to present here the results of a simple calculation of the two-photon absorption in a semiconducting quantum-well structure in which the carriers are confined in their motion along either one or two directions in space. In our calculations, we will use a two-band, isotropic parabolic band model for the semiconductor and neglect excitonic effects. Also, we will also neglect any interband transitions associated with the change in crystal symmetry which might be expected in reduced dimensional structures such as the splitting of the degenerate valence band which occurs in GaAs quantum wells. The anisotropy of the energy bands, the change in the crystal symmetry and excitonic effects will have to be taken into account when applying the results of our calculations to a real quantum-well system such as those fabricated from $GaAs/Ga_{1-x}Al_xAs$. However, the calculations presented here will give a rough idea of what can be expected in a semiconducting quantum-well system as compared to the bulk.

II. TWO-PHOTON ABSORPTION COEFFICIENT

The optical absorption coefficient can be directly related to the transition probability that an electron in the valence band will absorb two photons and make a transition to a state in the conduction band.[13-15] In two-photon absorption, there are three types of intermediate states which can play a role. The absorption can occur via intra-valence-band transitions, intra-conduction-band transitions, or transitions in which the intermediate state lies in some other band besides the valence and conduction bands in which the initial and final states occur. Because of the energy difference between the initial and intermediate states which occurs in the denominator of the transition probability, the transition rates will be largest when the intermediate state lies in either the same valence band as the initial state or the same conduction band as the final state since the energy difference will be smallest for these processes, and therefore, we will only consider such processes here.

When the light is polarized perpendicular to the direction of carrier confinement, the matrix elements of the momentum operator are nonvanishing only when the initial and final states have the same set of quantum numbers n, l, and k. Using the same procedure as in the calculation of the two-photon optical-absorption coefficient in bulk semiconductors[13-15] and considering only transitions involving an intermediate state in either the same valence or conduction band, we obtain the results for the absorption coefficients in the following normalized form:

$$K_{\mathrm{Q2D}}=(3y^{1/2}/2)(x-1)^{-3/2}\times\sum_{n=1}^{\infty}(x-1-n^2y)S(x-1-n^2y) \quad (1)$$

and

$$K_{\mathrm{Q1D}}=(3/\pi)(y_a y_b)^{1/2}(x-1)^{-3/2}\times\sum_{n,l=1}^{\infty}(x-1-n^2y_a-l^2y_b)^{1/2}\times S(x-1-n^2y_a-l^2y_b) \quad (2)$$

(Q2D stands for quasi-two-dimensional, etc.), where $K_{\mathrm{Q2D}}=\alpha_{\mathrm{Q2D}}/\alpha_{\mathrm{3D}}$, $K_{\mathrm{Q1D}}=\alpha_{\mathrm{Q1D}}/\alpha_{\mathrm{3D}}$, $x=(2\hbar\omega/E_g)$, $y=(E_d/E_g)$, and $y_{a,b}=(E_{a,b}/E_g)$. Here α_{Q1D}, α_{Q2D}, and α_{3D} are the absorption coefficients for the 1D, 2D, and 3D cases, respectively, E_g is the band-gap energy of the semiconductor, $S(x)$ is the step function, $E_d=(\pi^2\hbar^2/2\mu d^2)$, $E_a=(\pi^2\hbar^2/2\mu a^2)$, and $E_b=(\pi^2\hbar^2/2\mu b^2)$ are the energies of the quantized subbands for Q2D and Q1D quantum-well structures, respectively, μ is the reduced mass of the electron-hole pair, d is the width of the Q2D quantum-well structure, and a and b are the transverse dimensions of the quantum-well wire. The normalized absorption coefficients depend upon the parameters of the material only through the energy gap and the quantized subband energies. The two-photon absorption coefficients for this polarization for Q1D and Q2D structures differ from that in the bulk in two respects. First of all, because of the confinement energies of the electrons and holes in the quantum well, the threshold for two-photon absorption is shifted to higher photon energies. Secondly, the absorption coefficient reflects the difference in the density of states for the electrons which are confined in these quantum-well structures.

When the radiation field is polarized along the direction of carrier confinement, the momentum matrix elements take the form

$$(f\mid \epsilon p\mid I)=(2i\hbar/d)I(n',n)\delta_{k'k}$$

and (3)

$$(f\mid \epsilon p\mid I)=(2i\hbar/a)I(n',n)\delta_{l'l}\delta_{k'k}\ ,$$

where

$$I(n',n)=[1-(-1)^{n'-n}]\{n'n/[(n')^2-n^2]\}\ , \quad (4)$$

when the intermediate state lies in the conduction band for a Q2D or Q1D quantum-well system. The matrix elements given by Eq. (3) assume that the radiation is polarized along the direction where the width of the wire is a. If it is polarized along the direction in which the width of the well is b, then a is replaced by b in this equation. For this polarization, the selection rule that transitions can only occur between subbands having the same set of quantum numbers breaks down. Transitions for this polarization can occur between any two subbands whose quantum numbers differ by an odd integer. When the radiation is polarized along the direction of carrier confinement, the absorption coefficients can be expressed in the following normalized form:

$$K_{\mathrm{Q2D}}=(12y^{3/2}/\pi^2)(x-1)^{-3/2}\sum_{n=1}^{\infty}\sum_{n'(\neq n)}I^2(n',n)\{1-[(n')^2-n^2](2y_c/x)\}^{-2}S[x-1-(n')^2y_c-n^2y_v] \quad (5)$$

and

$$K_{\mathrm{Q1D}}=(24y_a^{3/2}y_b^{1/2}/\pi^3)(x-1)^{-3/2}\sum_{n,l=1}^{\infty}\sum_{n'(\neq n)}I^2(n',n)\{1-[(n')^2-n^2](2y_{ac}/x)\}^{-2}[x-1-l^2y_b-(n')^2y_{ac}-n^2y_{\mathrm{av}}]^{-1/2}\ . \quad (6)$$

Here y_c,y_v and y_{ca},y_{va} are the same as y and y_a,y_b except that the reduced mass of the electron-hole pair is replaced by the electron or hole masses. Again, the normalized absorption coefficients depend upon the parameters of the material only through the energy gap and the quantized subband energies.

In Figs. 1 and 2, the normalized absorption coefficients K_{Q2D} and K_{Q1D} for Q2D and Q1D quantum-well systems are shown as a function of the normalized photon energy x for the polarization either perpendicular or parallel to the direction of confinement. Due to the confinement of the carriers, the threshold for two-photon absorption to occur is shifted to photon energies greater than $E_g/2$ with the shift being greater for quantum-well wires than for Q2D quantum well systems. For this polarization, the absorption is suppressed over its bulk value. As the photon energy increases, the normalized absorption approaches its bulk value. In the limit of large wire thicknesses or well widths, the expressions for the absorption coefficient given by Eqs. (1) and (2) go over to the bulk value using the simple isotropic parabolic model for the energy bands. Also, the normalized absorption coefficients show structure whenever transitions can occur between new pairs of subbands.

For the radiation polarized parallel to the confinement direction, the absorption is enhanced over its bulk value for some ranges of the photon energy unlike the case of the other polarization where it is suppressed. For this polarization, the structure in the absorption which appears when transitions can occur between new pairs of quantized subbands is much sharper than for the case when the radiation is polarized perpendicular to the direction of carrier confinement. In addition, while there is strong structure in the absorption for Q2D structures for this po-

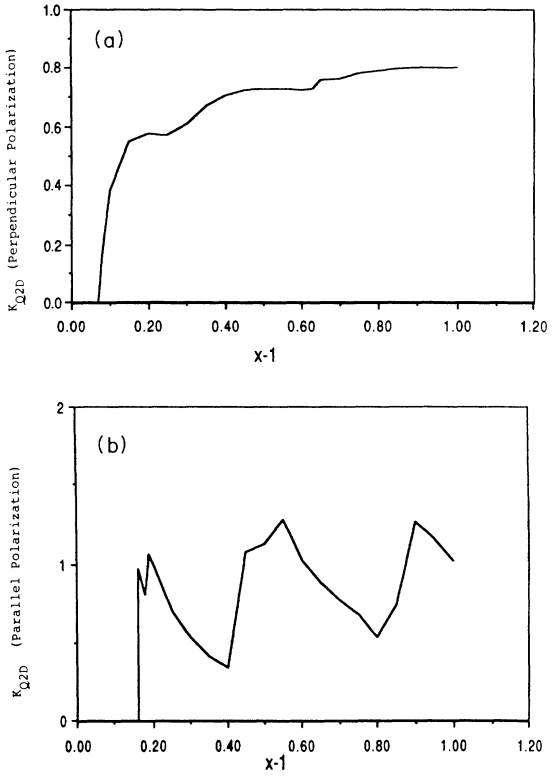

FIG. 1. The ratio of the two-photon absorption coefficients in a Q2D quantum-well system to that in the bulk, K_{Q2D}, is shown as a function of the normalized photon energy x. The parameters used in the calculation are those characteristic of GaAs. (a) Radiation polarized perpendicular to the direction of carrier confinement. (b) Radiation polarized parallel to the direction of carrier confinement.

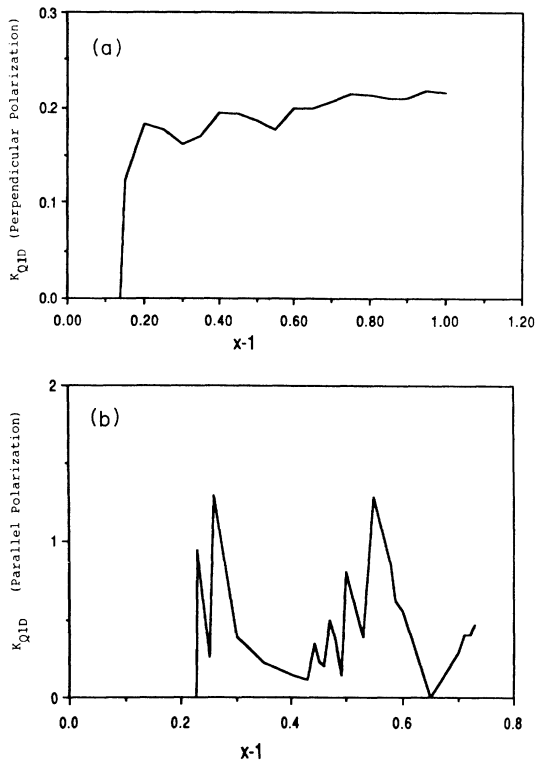

FIG. 2. The ratio of the two-photon absorption coefficients in a Q1D quantum-well system to that in the bulk, K_{Q1D}, is shown as a function of the normalized photon energy x. (a) Radiation polarized perpendicular to the direction of carrier confinement. (b) Radiation polarized parallel to the direction of carrier confinement.

larization, there are sharp peaks in the absorption of Q1D structures when transitions can occur between new pairs of subbands.

III. SUMMARY

The results of our calculations show that the two-photon absorption depends upon the polarization of the radiation field relative to the direction of carrier confinement and that the threshold energy for the absorption is shifted by the quantum confinement energies. The absorption is enhanced over its bulk value for radiation polarized along the direction of carrier confinement and is suppressed in comparison to its bulk value when the radiation is polarized perpendicular to the direction of confinement of the carriers. Although structure appears for both polarizations when transitions between new pairs of quantized subbands can occur, this structure is predicted to be much sharper for radiation polarized along the direction of carrier confinement than for radiation polarized perpendicular to this direction. The shift in the threshold energy is greater for radiation polarized along the direction of carrier confinement because of the different selection rules which hold for this polarization. The structure in the absorption reflects in part the structure which occurs in the density of states for Q1D and Q2D systems. Since the density of states diverges at the bottom (or top) of the subbands for a Q1D system, the absorption for this polarization reflects this divergence. In an actual situation, such divergences would be broadened by the effect of collisions. Also, since the present treatment neglects excitonic effects, these effects would alter to some extent the behavior of the absorption near the thresholds. In addition, we have calculated the absorption only in the region of photon energies where single-photon interband transitions are forbidden. In the transition region where both single-photon and two-photon absorption processes are significant, the total absorption will be different from the bulk because both the single- and two-photon processes are effected by the reduced dimensionality of the structures.

ACKNOWLEDGMENT

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