Oscillator strength and sum rule for intersubband transitions in a superlattice

F.M. Peeters* and A. Matulis†
Departement Natuurkunde, Universiteit Antwerpen (UIA), Universiteitsplein 1, B-2610 Antwerpen, Belgium

M. Helm, T. Fromherz, and W. Hilber
Institut für Halbleiterphysik, Universität Linz, A-4040 Linz, Austria
(Received 19 May 1993)

The oscillator strength for inter-minisubband optical absorption is calculated for a superlattice. For periodic structures the matrix element $\langle n|x|n'\rangle$ is ill defined and we show how the definition of the position operator has to be modified in order to obtain the correct result which then leads to the same result as a calculation of the oscillator strength using the matrix element $\langle n|p|n'\rangle$. A new sum rule for superlattices is derived, which involves *averaging* over wave vectors in the first Brillouin zone.

I. INTRODUCTION

Optical spectroscopy is a very important experimental tool to investigate the energy levels in semiconductors.[1–3] This knowledge is highly important both for a fundamental understanding of the band structure as well as for technological applications, e.g., in optoelectronics (quantum-well lasers, photodetectors). The quantity that determines the strength of an optical transition, if it is experimentally observable, is the dipole matrix element between initial and final states. For comparison of transition strengths in different physical systems, the dimensionless quantity of the so-called oscillator strength proves to be very useful. Historically, it was introduced for optical transitions within atoms and molecules where electrons are bound. In this case it is usually defined through the coordinate (x) matrix element between the one electron wave functions by[4]

$$f_{n'n} = \frac{2m\omega_{n'n}}{\hbar}|\langle n'|x|n\rangle|^2 , \tag{1.1}$$

where m is the electron mass and $\omega_{n'n} = E_{n'n}/\hbar = (E_{n'} - E_n)/\hbar$ the angular frequency of the corresponding electron transition.

The direct way of coupling the radiation field to the electron Hamiltonian is, however, through the $\mathbf{A}\cdot\mathbf{p}$ interaction,[5] where $\mathbf{A}$ is the vector potential of the electromagnetic field and $\mathbf{p}$ is the momentum operator. This leads to a definition for the oscillator strength of

$$f_{n'n} = \frac{2}{m\hbar\omega_{n'n}}|\langle n'|p|n\rangle|^2 . \tag{1.2}$$

For bounded systems both definitions (1.1) and (1.2) are equivalent, which is a direct consequence of the commutation relation $p = i\frac{m}{\hbar}[H, x]$, valid for velocity-independent potentials and in the absence of any magnetic field.

Furthermore, the oscillator strengths satisfy the so-called f sum rule

$$\sum_{\nu'} f_{\nu'\nu} = 1 . \tag{1.3}$$

Here ν specifies one set of quantum numbers. The ν' summation has to be performed over all possible final-state quantum numbers. When the final state has a higher energy than the initial state, f is positive (absorption); in the opposite case f is negative (emission). This sum rule actually does not rely on specific matrix elements, but follows from general analytic properties of the dielectric function and therefore is a fundamental property of matter. Equivalently, it can be written as[6]

$$\int_0^\infty d\omega\ \omega\epsilon_2(\omega) = \frac{\pi}{2}\omega_p^2 , \tag{1.4}$$

in which form it is called the Thomas-Reiche-Kuhn[7] sum rule. In Eq. (1.4) $\epsilon_2(\omega)$ is the imaginary part of the dielectric function and ω_p is the plasma frequency. The above sum rule is valid within linear response in the electric field of the radiation and for arbitrary order in anything else (i.e. multiphonon processes, scattering with impurities, etc.).

In the case of electron states which are not bounded, such as for a free particle or electrons in a periodic system (i.e., superlattice), the two expressions (1.1) and (1.2) lead to different results. For a bounded system the first-order processes exhaust completely the sum rule (1.3). We found that this is no longer true for an unbounded system.

The purpose of this paper is (1) to show that in periodic systems (we will take a superlattice as an example) Eq. (1.1) is not applicable, but instead it is mandatory to use the $\mathbf{A}\cdot\mathbf{p}$ interaction with Eq. (1.2) as a definition for the oscillator strength. The reason for this is that the x operator is not well defined in periodic systems. (2) We show under which conditions (1.2) and (1.1) are equivalent and how the x operator has to be modified to yield the correct results in the other cases. (3) Furthermore, we apply the formalism to transitions between minibands in semiconductor superlattices, where we are able to prove a new sum rule, which contains an average over the wave vectors in the first Brillouin zone.

0163-1829/93/48(16)/12008(8)/$06.00

©1993 The American Physical Society

II. OPTICAL TRANSITIONS IN BOUNDED SYSTEMS

In order to pose the problem clearly we will give a concise review of the relevant results for optical transitions in bounded systems. In classical electrodynamics an electromagnetic field is described by the electric $\mathbf{E}(\mathbf{r},t)$ and magnetic $\mathbf{B}(\mathbf{r},t)$ fields which satisfy the Maxwell equations. These fields can be expressed in terms of the four vector $[\mathbf{A}(\mathbf{r},t),\phi(\mathbf{r},t)]$ as follows: $\mathbf{E} = -\frac{1}{c}\frac{\partial \mathbf{A}}{\partial t} - \text{grad}\phi$ and $\mathbf{B} = \text{rot}\mathbf{A}$, where $\mathbf{A}(\mathbf{r},t)$ is called the vector potential and $\phi(\mathbf{r},t)$ the scalar potential which are determined up to a gauge transformation. A nonrelativistic particle of mass m and charge q interacting with an electromagnetic field is described by the Hamiltonian[5]

$$H = \frac{1}{2m}\left(\mathbf{p} - \frac{q}{c}\mathbf{A}\right)^2 - q\phi \ . \tag{2.1}$$

One can choose a gauge in which $\phi = 0$ and $\text{div}\mathbf{A} = 0$. This leads to the interaction Hamiltonian for a plane wave $\mathbf{A}(\mathbf{r},t) = \mathbf{A_0}\exp(i\mathbf{k}\cdot\mathbf{r} - i\omega t)$,

$$H' = i\frac{e}{m\omega}\mathbf{E}\cdot\mathbf{p}e^{i(\mathbf{k}\cdot\mathbf{r}-\omega t)} \ . \tag{2.2}$$

For most systems one can expand the exponential $\exp(i\mathbf{k}\cdot\mathbf{r})$ leaving only the zeroth order, i.e., $\exp(i\mathbf{k}\cdot\mathbf{r}) \sim 1$. This is valid as long as the wavelength of the light is much larger than the characteristic dimension of the electron system (electron wavelength). In atoms this inequality is fulfilled very well, but also for subband transitions in semiconductor superlattices, where $\lambda_{\text{photon}} \sim 10\ \mu\text{m} \gg \lambda_{\text{electron}} \sim 100$ Å. This is usually called the electric dipole approximation.

Through a gauge transformation, which is exact within the dipole approximation,[8] an interaction Hamiltonian

$$H' = e\mathbf{E}\cdot\mathbf{r}e^{-i\omega t} \tag{2.3}$$

is achieved which is the more commonly used electric dipole interaction Hamiltonian. $\mathbf{D} = e\mathbf{r}$ is the electric dipole moment associated with the electron. So we see that both interactions ($\mathbf{A}\cdot\mathbf{p}$ as well as $\mathbf{E}\cdot\mathbf{r}$) appear well justified. But for periodic systems (such as, e.g., in a superlattice where the electron states extend to infinity) Eq. (2.3) contradicts the periodic boundary condition while Eq. (2.2) contains only the momentum operator and is consequently periodic.

A bounded system is one in which the electron wave function satisfies the boundary conditions $\psi(x)\,|_{x\to\pm\infty}\to 0$. For future purposes we will introduce dimensionless units by making the following transformations: $x \to a_0 x$, $p \to \hbar p/a_0$, where a_0 is some characteristic length in the system which we do not have to specify at this moment. The energy is measured in units of $E_0 = \hbar^2/ma_0^2$ (= 11 meV for $a_0 = 100$ Å and $m/m_0 = 0.068$, which is a typical value for GaAs).

For simplicity we will consider the operators to be one dimensional and use only one quantum number. First we will derive a relation between the x and p matrix elements. Therefore we consider the following commutator: $p = \dot{x} = i[H,x]$, which may be written in matrix form

$$\langle n'|p|n\rangle = iE_{n'n}\langle n'|x|n\rangle \tag{2.4}$$

from which we easily derive the identity of the oscillator strengths (1.1) and (1.2),

$$f_{n'n} = 2E_{n'n}|\langle n'|x|n\rangle|^2 = \frac{2}{E_{n'n}}|\langle n'|p|n\rangle|^2. \tag{2.5}$$

The sum rule (1.3) is obtained by considering the commutator $i[p,x] = 1$, which in matrix representation reads

$$\begin{aligned} 1 &= i\langle n|[p,x]|n\rangle = i\sum_{n'}[\langle n|p|n'\rangle\langle n'|x|n\rangle \\ &\qquad -\langle n|x|n'\rangle\langle n'|p|n\rangle] \\ &= 2\sum_{n'}\text{Im}\langle n'|p|n\rangle\langle n'|x|n\rangle^* = \sum_{n'} f_{n'n} \ . \end{aligned} \tag{2.6}$$

III. SYSTEM WITH A PERIODIC POTENTIAL

In order to check the validity of Eqs. (2.4) and (2.6) in periodic potentials $V(x) = V(x+1)$ we consider the simple one-dimensional Schrödinger equation

$$\left[\frac{d^2}{dx^2} + 2[E - V(x)]\right]\psi(x) = 0, \tag{3.1}$$

where the unit of length a_0 is taken to be equal to the period of our system. The generalization to problems in higher dimensions is straightforward. For semiconductor superlattices the above equation is valid within the envelope function approximation[2] where we have neglected the mass difference between the barriers and the wells for simplicity. The solution of Eq. (3.1) has a Bloch form

$$\psi_{n,k}(x) = e^{ikx}u_{n,k}(x), \tag{3.2}$$

where the function $u_{n,k}(x)$ is periodic $u_{n,k}(x+1) = u_{n,k}(x)$ and obeys the following Schrödinger equation:

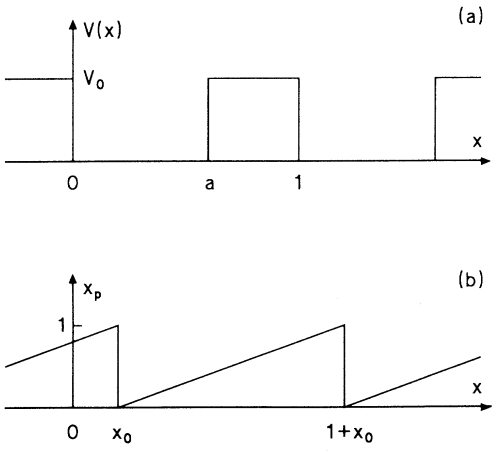

FIG. 1. (a) Superlattice potential and (b) periodic coordinate operator in dimensionless units.

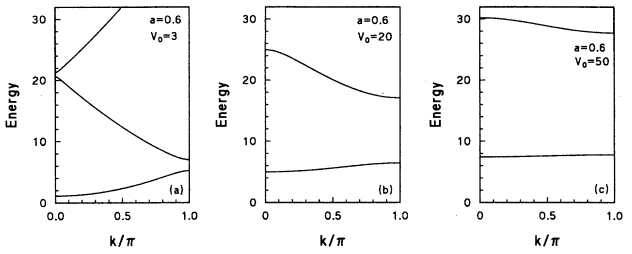

FIG. 2. The first two minibands of the energy spectrum (in units of $\hbar^2/ma_0^2$) of a superlattice with well width of $a = 0.6$ and potential strength: (a) $V_0 = 3$, (b) $V_0 = 20$, and (c) $V_0 = 50$.

$$[H(k) - E_{n,k}]\, u_{n,k}(x) = 0 \tag{3.3}$$

in the interval $0 \leq x \leq 1$ with the Hamiltonian

$$H(k) = -\frac{1}{2}\frac{d^2}{dx^2} - ik\frac{d}{dx} + \frac{1}{2}k^2 + V(x). \tag{3.4}$$

We will assume the following normalization condition:

$$\langle n,k|n,k\rangle = \int_0^1 dx|u_{n,k}|^2 = 1. \tag{3.5}$$

As a numerical illustration we took the simple potential $V(x) = 0(0 \leq x \leq a), V_0(a \leq x \leq 1)$, shown in Fig. 1(a), which models a typical superlattice. In both intervals the electron wave function can be written as linear combinations of trigonometric and hyperbolic functions. The eigenvalues (see Figs. 2 and 3) and corresponding wave functions are obtained by matching numerically the above-mentioned functions at the potential steps. For optical transitions the wavelength of light is much larger than any characteristic dimension in the superlattice and consequently we can restrict ourselves to vertical intersubband transitions (i.e., in which $\Delta k = 0$). In the present paper we will also neglect photon-phonon transitions. The matrix elements relevant to us are

$$P_{nn'}(k) = \langle n,k|p|n',k\rangle = \int_0^1 dx\; u_{n,k}(x)^* p u_{n',k}(x)$$
$$= -i\int_0^1 dx\; u_{n,k}(x)^* \dot{u}_{n',k}(x) \tag{3.6}$$

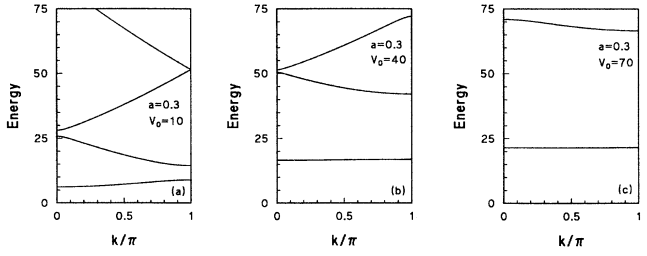

FIG. 3. The same as Fig. 2 but now for $a = 0.3$ and (a) $V_0 = 10$, (b) $V_0 = 40$, and (c) $V_0 = 70$.

and

$$X_{nn'}(k) = iE_{n,n'}(k)\langle n,k|x|n',k\rangle$$
$$= iE_{n,n'}(k)\int_0^1 dx\; u_{n,k}(x)^* x u_{n',k}(x), \tag{3.7}$$

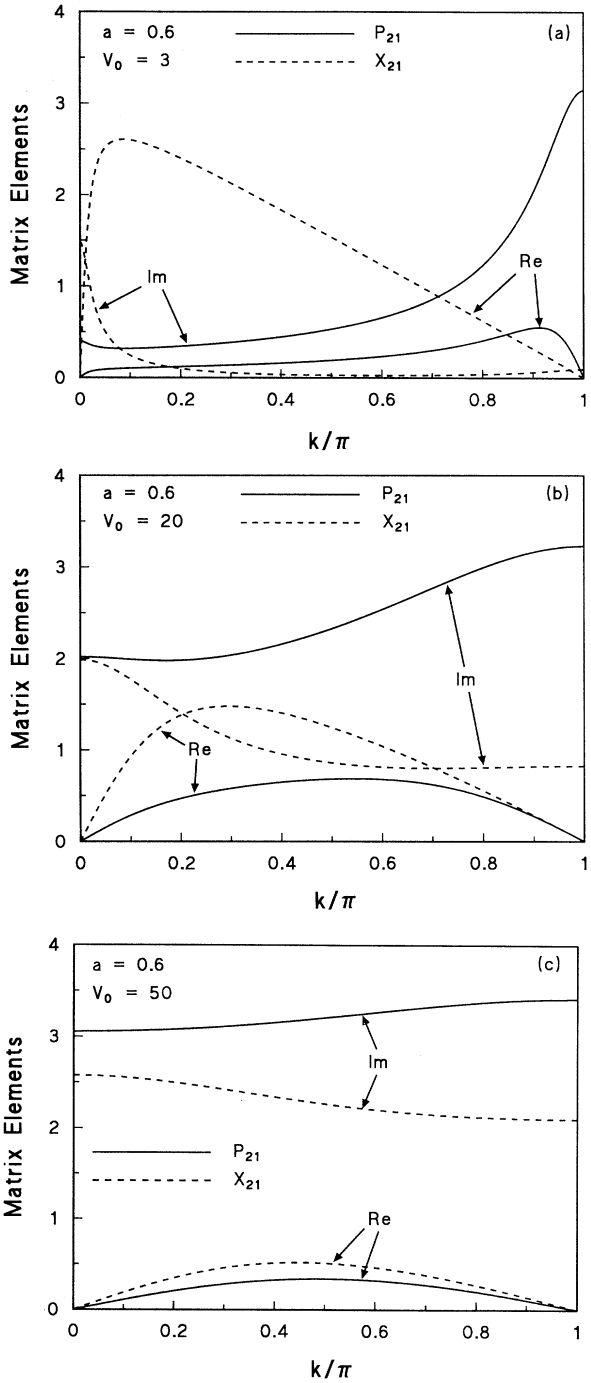

FIG. 4. Real and imaginary part of the coordinate ($X_{n,n'}$) and momentum ($P_{n,n'}$) matrix elements as a function of the wave vector for $n = 2, n' = 1$, quantum well width of $a = 0.6$ and potential strength of (a) $V_0 = 3$, (b) $V_0 = 20$, (c) $V_0 = 50$.

which we defined such that it enables us a direct comparison with $P_{nn'}(k)$. In Eq. (3.7) we used $E_{n,n'}(k) = E_{n,k} - E_{n',k}$. In a bounded system the matrix elements $P_{nn'}(k)$ and $X_{nn'}(k)$ coincide according to Eq. (2.4). In the case of a periodic system the real and imaginary part of the matrix elements (3.6) and (3.7) are depicted in Fig. 4 for $n = 2, n' = 1$, and for a superlattice with

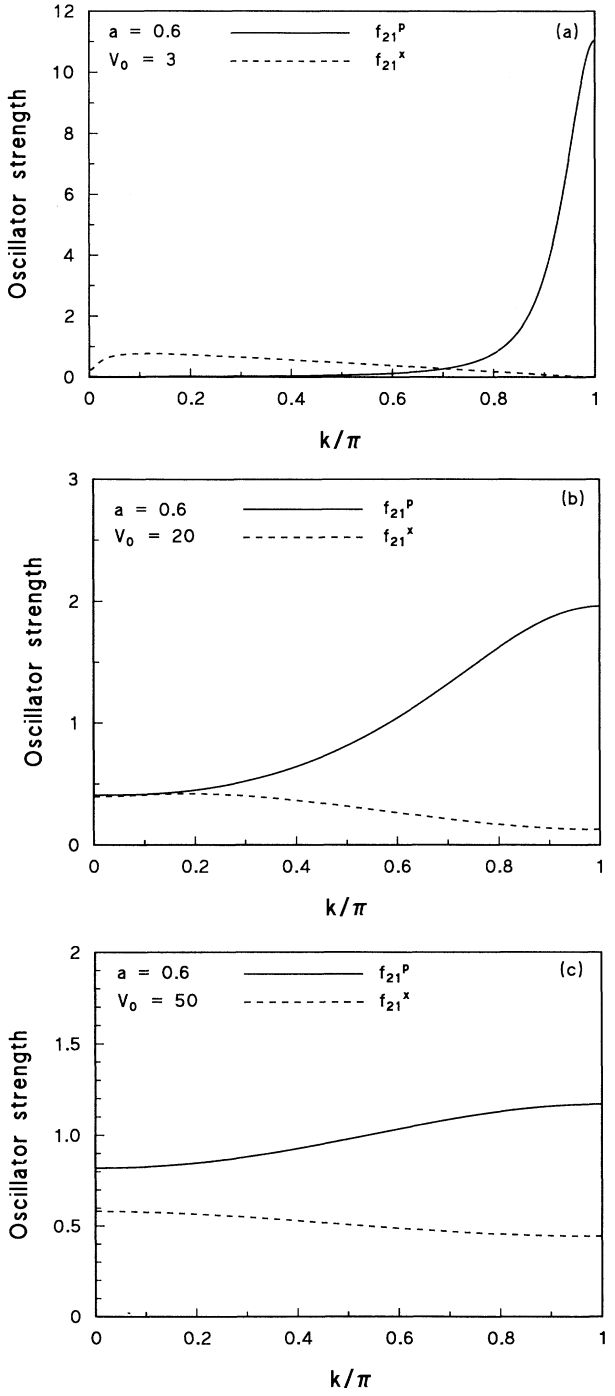

FIG. 5. The oscillator strengths corresponding to the matrix elements of Fig. 4 as defined by Eqs. (1.1) and (1.2).

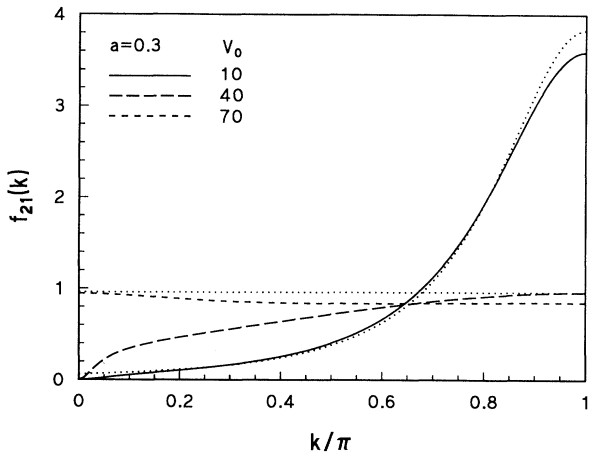

FIG. 6. The oscillator strength $f_{21}(k)$ as function of the electron wave vector for a well width of $a = 0.3$ and three different values of the height of the potential barrier. The results for $V_0 \to 0$ and for a single quantum well are given by the dotted curves.

well width $a = 0.6$ and for various values of the potential strengths: (a) $V_0 = 3$, (b) $V_0 = 20$, and (c) $V_0 = 50$. It is apparent that the relation (2.4) is not valid in the present case. The corresponding results for the oscillator strength defined by Eq. (2.5) as obtained from the momentum matrix elements $f^p_{nn'}$ and position matrix element $f^x_{nn'}$ are shown in Fig. 5. In Fig. 6 the oscillator strength f^p_{21} is plotted for a narrower well of $a = 0.3$ and three different values of the barrier height. Notice that the oscillator strength f^p_{21} is an increasing function of the electron wave vector, which is a consequence of the fact that $E_{21}(k)$ decreases with increasing k (see Fig. 2). This is more apparent for strongly coupled superlattices [Fig. 5(a)] where near the Brillouin-zone boundary the oscillator strength can become more than ten times larger in comparison to its value at the zone center. The function f^x_{21} is less than 1 and it is a uniformly decreasing function of the wave vector except for very strongly coupled superlattices [see Fig. 5(a)].

IV. SOLUTION TO THE PROBLEM

The discrepancy between the matrix elements $P_{nn'}$ and $X_{nn'}$ is due to the fact that the electron coordinate operator is unbounded and its matrix element is ill defined in the case of a free particle or electrons in a periodic system. This problem was also encountered in other areas in solid-state physics, e.g., in the definition of the Wannier position operator in the discussion on Wannier states in crystals. Because of the imposed periodic boundary condition in such a system all operators should have at least the same periodicity.[9]

Following Ref. 9 we remedy this by replacing the coordinate operator x by a periodic function $x_p(x)$, which is shown in Fig. 1(b). It coincides with the coordinate x in the elementary cell and is composed of a periodic rep-

etition of the "sawlike" function $q(x) = x\theta(1-x)$ where $\theta(x)$ is the step function $\theta(x) = 1(x \geq 0), 0(x < 0)$. Note that the elementary cell can be chosen with an arbitrary shift x_0 relative to the periodic potential $V(x)$.

Now we have to calculate the following commutator:

$$[H(k), q(x-x_0)] = -\left(\frac{d}{dx} + ik\right) + \delta(x-x_0)\left(\frac{d}{dx} + ik\right) + \dot{\delta}(x-x_0), \tag{4.1}$$

from which we obtain the corresponding matrix element

$$E_{nn'}(k)\langle n,k|x|n',k\rangle = -i\{\langle n,k|p|n',k\rangle + k\delta_{nn'}\} + w_{nn'}(k;x_0), \tag{4.2}$$

where we defined the following function:

$$\begin{aligned} w_{nn'}(k;x) &= \frac{1}{2}\{u_{n,k}(x)^* \dot{u}_{n',k}(x) - \dot{u}_{n,k}(x)^* u_{n',k}(x)\} \\ &\quad + iku_{n,k}(x)^* u_{n',k}(x) \\ &= \frac{1}{2}\{\psi_{n,k}(x)^* \dot{\psi}_{n',k}(x) - \dot{\psi}_{n,k}(x)^* \psi_{n',k}(x)\}, \end{aligned} \tag{4.3}$$

which is actually half the Wronskian of two solutions of Eq. (3.1). For $n = n'$ this Wronskian is nothing else but the matrix element of the current operator for an electron in subband n with wave vector k. In a periodic potential the relation between the x and p matrix elements becomes

$$P_{nn'} + k\delta_{nn'} = X_{nn'} - iw_{nn'}(k;x_0), \tag{4.4}$$

where the term linear in the wave vector disappears for the nondiagonal elements.

We have checked numerically the validity of Eq. (4.4) for the above-mentioned model system. Note that in a periodic system the Wronskian (4.3) has to be added to the standard relation (2.4), which is only valid for a bounded system. The Wronskian (4.3) is in general a complex quantity and we found numerically that the absolute value of it is less when (1) the elementary cell edges x_0 are in the barrier region ($a \leq x \leq 1$), (2) V_0 is large; in the limit $V_0 \to \infty$ the system turns into a bound system and the Wronskian is zero, and (3) for $k = 0$ the Wronskian is real and there exist x values for which the Wronskian is zero and Eq. (4.4) accidentally coincides with the relation (2.4) for bounded system. This is no longer true when $k \neq 0$. Thus it follows that when the potential is sufficiently strong and the elementary cell edges are in the barrier region, the $\mathbf{E}\cdot\mathbf{r}$ and $\mathbf{A}\cdot\mathbf{p}$ interaction become equivalent again. The system is then still periodic, but the unit cells are completely decoupled from each other.

Next we will follow the approach of Sec. II in order to derive the sum rule valid for periodic systems. Therefore consider the following commutator:

$$i[p, q(x-x_0)] = \left[\frac{d}{dx}, q(x-x_0\right] = 1 - \delta(x-x_0), \tag{4.5}$$

with its corresponding matrix elements

$$\begin{aligned} i\langle n,k|[p,q(x-x_0)]|n',k\rangle &= i\sum_{n'}\{\langle n,k|p|n',k\rangle\langle n',k|x|n,k\rangle - \langle n,k|x|n',k\rangle\langle n',k|p|n,k\rangle\} \\ &= -i\sum_{n'}\{\langle n',k|p|n,k\rangle\langle n',k|x|n,k\rangle^* - \langle n',k|p|n,k\rangle^*\langle n',k|x|n,k\rangle\} \\ &= 1 - |u_{n,k}(x_0)|^2 \,. \end{aligned} \tag{4.6}$$

Finally we obtain the sum rule

$$2\sum_{n'} \mathrm{Im}\{\langle n',k|p|n,k\rangle\langle n',k|x|n,k\rangle^*\} = 1 - |u_{n,k}(x_0)|^2. \tag{4.7}$$

Notice that this sum rule (4.7) and relation (4.4) depend on the way we have chosen the elementary cell relative to the potential $V(x)$. Furthermore, the terms in the sum on the left-hand side of the above equation are not simply the oscillator strengths for optical absorption. In order to find an expression which contains only $P_{n'n}$ and not $X_{n'n}$, we insert Eq. (4.4) into Eq. (4.7) to get

$$\begin{aligned} \sum_{n'} f_{n'n}(k) &= \sum_{n'} \frac{2}{E_{n'n}(k)}|P_{n'n}(k)|^2 \\ &= 1 - |u_{n,k}(x_0)|^2 \\ &\quad + 2\,\mathrm{Im}\sum_{n'} P_{n'n}(k)\frac{w_{n'n}(k;x_0)}{E_{n'n}(k)} \,. \end{aligned} \tag{4.8}$$

Making use of the Wronskian properties one easily proves that the right-hand side (RHS) of Eq. (4.8) does not depend on x_0. The above expression can hardly be considered as a sum rule because the RHS depends in a very complicated way on the detailed features of the band

structure. However, this is not a real surprise, since in (4.8) a summation is only performed over the miniband index n' and not over the final wave vectors k'. The complete f sum rule must have the form

$$\sum_{k'} \sum_{n'} f_{n'k',nk} = 1 \ . \tag{4.9}$$

This cannot, however, be proven explicitly, since the transitions to states with different final wave vectors $k' \neq k$ are second-order (and higher-order) processes, where the momentum transfer has to be provided by phonon emission/absorption or impurity scattering. Physically, these processes correspond to *intra*-miniband (or free-carrier) absorption/emission, when $n' = n$ (for a related experiment, see Brozak *et al.*[11]). In contrast, the transitions with $k' = k$ and $n' \neq n$ are *inter*-miniband transitions. In order for the complete f sum rule to be satisfied, all possible transitions have to be summed up, no matter if they are higher-order processes. In the next section we will show that it is possible to obtain a new sum rule, which involves inter-miniband transitions. This is done by averaging Eq. (4.8) over the first Brillouin zone.

V. AVERAGED SUM RULE

In order to prove this new sum rule we will first need a number of relations which we obtain by differentiating Eq. (3.3) by d/dk and integrating it over the elementary cell

$$\int_0^1 dx \ u_{n',k}(x)^* \left(\left[\frac{dH(k)}{dk} - \frac{dE_{n,k}}{dk} \right] u_{n,k} + [H(k) - E_{n,k}] \frac{du_{n,k}}{dk} \right) = 0, \tag{5.1}$$

which, by using Eq. (3.4), can be reduced to

$$\int_0^1 dx \ u_{n',k}(x)^* \left(-i\frac{d}{dx} \right) u_{n,k}(x) - \left(\frac{dE_{n,k}}{dk} - k \right) \delta_{n'n} + (E_{n',k} - E_{n,k}) \int_0^1 dx \ u_{n',k}(x)^* \frac{d}{dk} u_{n,k}(x) = 0. \tag{5.2}$$

Finally we obtain

$$\langle n', k|p|n, k\rangle = \delta_{n'n} \left(\frac{dE_{n,k}}{dk} - k \right) - E_{n'n}(k) \left\langle n', k \left| \frac{d}{dk} \right| n, k \right\rangle \ . \tag{5.3}$$

which does not depend on the choice of the position of the unit cell. The first term on the RHS of Eq. (5.3) is the velocity of the electron state in the miniband n minus the contribution from the Bloch term Eq. (3.2). The last term is the nondiagonal contribution.

An analogous mathematical trick is applied to the complex conjugate of Eq. (3.3) and we obtain a similar expression

$$\langle n', k|p|n, k\rangle = \delta_{n'n} \left(\frac{dE_{n,k}}{dk} - k \right) + E_{n'n}(k) \left\langle n', k \left| \frac{\overleftarrow{d}}{dk} \right| n, k \right\rangle, \tag{5.4}$$

where the symbol $\overleftarrow{d}/dk$ indicates that the differential operator acts on the left function.

Next we make use of Eqs. (5.3) and (5.4) for the nondiagonal case ($n' \neq n$) and calculate the sum over the oscillator strengths for inter-miniband transitions

$$\begin{aligned} F_n(k) &= \sum_{n' \neq n} f_{n'n}(k) = \sum_{n' \neq n} \frac{2}{E_{n'n}(k)} \langle n, k|p|n', k\rangle \langle n', k|p|n, k\rangle \\ &= -\sum_{n' \neq n} \left[\langle n, k|p|n', k\rangle \left\langle n', k \left| \frac{d}{dk} \right| n, k \right\rangle + \left\langle n, k \left| \frac{\overleftarrow{d}}{dk} \right| n', k \right\rangle \langle n', k|p|n, k\rangle \right] \\ &= -\left\langle n, k \left| p \frac{d}{dk} \right| n, k \right\rangle - \left\langle n, k \left| \frac{\overleftarrow{d}}{dk} p \right| n, k \right\rangle. \end{aligned} \tag{5.5}$$

In the last step we used the fact that the wave functions with the same k vector satisfy the completeness relation and

$$\left\langle n, k \left| \frac{d}{dk} \right| n, k \right\rangle + \left\langle n, k \left| \frac{\overleftarrow{d}}{dk} \right| n, k \right\rangle = \frac{d}{dk} \langle n, k|n, k\rangle = 0. \tag{5.6}$$

Finally, we arrive at the following sum rule:

$$F_n(k) = \sum_{n' \neq n} f_{n'n}(k) = -\frac{d}{dk} \langle n, k|p|n, k\rangle \ , \tag{5.7}$$

which tells us that the sum of the oscillator strengths for vertical transitions is in general not equal to 1 anymore

and depends on the initial state $|n,k\rangle$.

The derivative d/dk on the RHS of Eq. (5.7) suggests that the above equation can be simplified by averaging it over the first Brillouin zone. Indeed, integrating Eq. (5.7) over k we obtain

$$F_n = \frac{1}{2\pi}\int_{-\pi}^{\pi} F_n(k)dk = \frac{1}{2\pi}\Big[\langle n,-\pi|p|n,-\pi\rangle - \langle n,\pi|p|n,\pi\rangle\Big]. \quad (5.8)$$

This can be further simplified if we use the definition for the average electron velocity [see the first term on the RHS of Eq. (5.3)]

$$v_n(k) = k + \langle n,k|p|n,k\rangle \quad (5.9)$$

and the fact that the velocity is zero at the Brillouin-zone edges. We finally obtain the average sum rule

$$F_n = \frac{1}{2\pi}\int_{-\pi}^{\pi} dk \sum_{n'\neq n} f_{n'n}(k) = 1\ , \quad (5.10)$$

which now is independent of the band index n. Note that we integrate over k ($\frac{1}{2\pi}\int dk = \sum_k$ in our units), whereas in Eq. (4.9) the integral goes only over the final states k'. The physical meaning of this sum rule is that now the influence of intra-miniband emission and absorption is averaged out, and we regain a sum rule similar to a single quantum well. Now it is also easy to see why the oscillator strength at the Brillouin-zone center has to be smaller than one, whereas at the edge it can be larger (see Figs. 5 and 6). The center of the Brillouin zone of the bottom band is the lowest energy in the system. Therefore, only absorptive transitions are possible, of which some fraction are $n \neq n'$, $k = k'$ interband transitions, and some are $n = n'$, $k \neq k'$ intraband transitions. Clearly, for both parts, the oscillator strength must be smaller than one. In contrast, at the edge of the Brillouin zone, the intraband transitions are emission processes and count negative in the oscillator sum. Therefore, the interband oscillator strength at the zone edge has to be greater than one. This asymmetry has recently been proven experimentally by some of the present authors.[10]

In Figs. 7(a) and 7(b) we illustrate this averaged sum rule by plotting f_{21} (dotted curve), $f_{21}+f_{31}$ (dashed curve), and $f_{21}+f_{31}+f_{41}$ (solid curve) as a function of the strength of the potential step V_0 for the case of (a) $a = 0.3$ (i.e., the well width is roughly half that of the barrier) and (b) $a = 0.6$ (i.e. the well width is 1.5 times the barrier width). Notice that in the limit of $V_0 \to 0$ (the nearly-free electron case) the main contribution to the sum rule comes from the transition f_{21}, which is concentrated in the $k \sim \pi$ region [see Figs. 5(a) and 6]. When V_0 increases the oscillator strengths are redistributed into the upper bands and we notice that f_{21} passes through a minimum while the other oscillator strengths attain a maximum. Notice also that f_{31} is different from zero and can be even larger than f_{41} for not too large values of V_0,

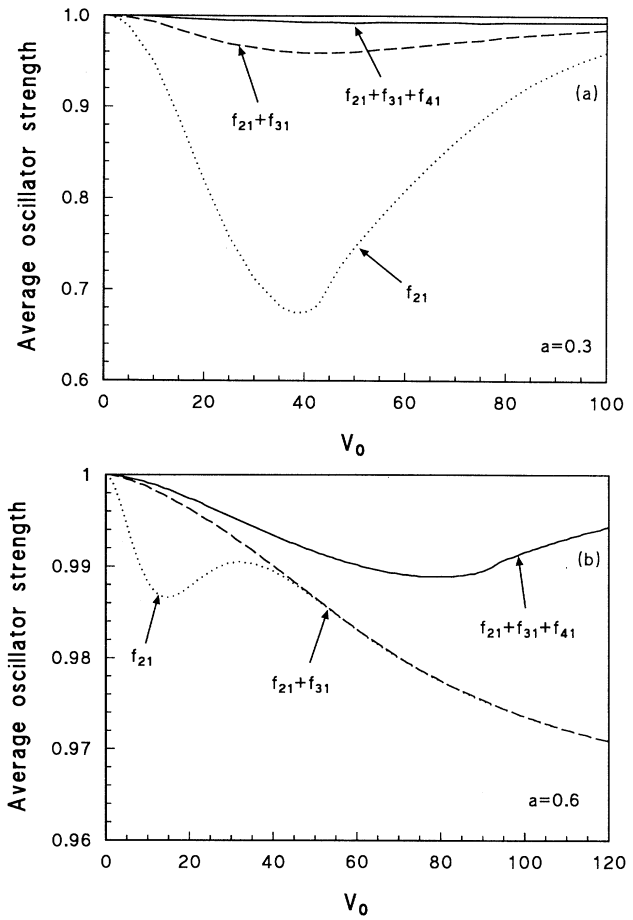

FIG. 7. The sum of the oscillator strengths averaged over the first Brillouin zone as function of the height of the potential barrier for a well width of (a) $a = 0.3$ and (b) $a = 0.6$.

i.e., when the system still exhibits wide minibands. This is in contrast to the quantum-well case where $f_{31} = 0$ for symmetry reasons. The minimum in f_{21} becomes deeper with decreasing well width [e.g., for $a = 0.2$ we have $(f_{21})_{\min} = 0.34$ at $V_0 \sim 85$ where we have $f_{31} = 0.42$ and $f_{41} = 0.18$] and it is less pronounced for larger a values [e.g., for $a = 0.6$ we found $(f_{21})_{\min} = 0.987$ at $V_0 \sim 20$ where $f_{31} = 0.011$]. We can understand the behavior of f_{21} in Fig. 7 as follows. For small values of V_0 the electron in the first level is quasifree in the superlattice direction, but with increasing V_0 it becomes more and more localized in the well region. As a consequence the overlap with the wave function of the second miniband, which is still quasifree, decreases and thus f_{21} also decreases. For $a = 0.3$ the energy of the first miniband is below the barrier when $V_0 > 20$. Further increase of V_0 will start to trap the wave function of the second miniband into the wells of the superlattice; in fact for $V_0 \sim 55$ the bottom of the second miniband becomes equal to V_0 and the top of the second miniband is completely below the barrier for $V_0 > 73$ in the case of $a = 0.3$. At the same time the wave function of the second miniband $|2\rangle$ becomes more and more trapped into the wells and the overlap with

the wave function $|1\rangle$ increases leading to a concomittant increase of f_{21}. The substantial difference between Figs. 7(a) and 7(b) is a consequence of the fact that in superlattices with small well widths (i.e., small a values) there is a larger contrast between bound and extended states, which makes the matrix elements substantially different and also different from the quantum-well case.

The distribution of the oscillator strength $f_{21}(k)$ in wave-vector space is illustrated in Figs. 5 and 6 for $a = 0.6$ and $a = 0.3$, respectively, and three different values of the barrier height. For small V_0 values the oscillator strength is strongly determined by the energy term $E_{21}(k)$ [see the definition of $f_{21}(k)$ in Eq. (4.8)], which becomes very small near $k = \pi$ and which results in a $f_{21}(k)$ which is strongly peaked near the edge of the Brillouin zone. It is possible to obtain an analytic expression for f_{21} in the nearly-free-electron approximation,[12] i.e., $V_0 \to 0$. In this approximation the two lowest-energy minibands are approximated by free-electron bands, except near the zone boundary where the splitting of the band is calculated using the superlattice potential as a perturbation. In doing so, we obtain

$$f_{21}(k) = \frac{\gamma}{\{1 + [\gamma(1 - k/\pi)]^2\}^{3/2}}, \tag{5.11}$$

where

$$\gamma = \frac{\pi^3}{V_0 \sin(\pi a)}, \tag{5.12}$$

which for $V_0 = 10$ is depicted in Fig. 6 by the dotted curve. The agreement becomes exact in the limit $V_0 \to 0$. Similarly we can prove, within the same limit, that $f_{n1} \sim V_0^2$ for $n \geq 3$. With increasing V_0 the difference $E_{21}(k = \pi)$ increases and $f_{21}(k = \pi)$ decreases as is apparent, e.g., in Fig. 6 when we compare the $V_0 = 10$ result with the $V_0 = 40$ result. With increasing V_0 the energy bands are becoming more and more dispersionless (see Figs. 2 and 3), which results in an almost constant difference $E_{21}(k)$ and consequently in an oscillator strength $f_{21}(k)$ which is independent of the wave vector and which approaches the quantum-well result (dotted horizontal curve in Fig. 6).

VI. CONCLUSIONS

We have shown that the standard expressions for the calculation of the oscillator strength as used in atomic physics, which is based on the x-matrix element, are not applicable in systems where the electron wave function is unbounded. The correct calculation has to be based on the p-matrix elements. The fundamental Thomas-Reiche-Kuhn sum rule of course still has to be satisfied, but both intra- and inter-miniband transitions have to be taken into account. A new sum rule has been derived, which only contains interband transitions. This is achieved by averaging the oscillator strengths over the first Brillouin zone of the superlattice.

In conclusion, we would like to point out that, although we considered only the simple one-dimensional case (i.e., superlattice) of periodic potentials, the main results and the averaged sum rule can be easy generalized to two-dimensional and three-dimensional periodic systems.

ACKNOWLEDGMENTS

This work is supported by the Belgian National Science Foundation (NFWO), the Interuniversity Microelectronics Center (IMEC, Leuven), the "Fonds zur Förderung der Wissenschaftlichen Forschung" (FWF), Vienna, and by the "Jubiläumsfonds der Österreichischen Nationalbank." One of us (A.M.) acknowledges the support of the EEC through the program "Cooperation in Science and Technology with Central and Eastern European Countries." M.H. acknowledges elucidating conversations with M. Kriechbaum and P. Vogl.

* Electronic address: peeters@nats.uia.ac.be
† Permanent address: Semiconductor Physics Institute, Gostauto 11, 2600 Vilnius, Lithuania.

[1] J. Pankove, *Optical Processes in Semiconductors* (Dover, New York, 1975).
[2] G. Bastard, *Wave Mechanics Applied to Semiconductor Hetrostructures* (Les éditions de physique, Les Ulis Cedex, 1988), p. 237.
[3] For recent reviews see, e.g., S. Schmitt-Rink, D.S. Chemla, and D.A.B. Miller, Adv. Phys. **38**, 89 (1989); *Physics and Applications of Quantum Wells and Superlattices*, edited by E.E. Mendez and K. von Klitzing (Plenum, New York, 1987).
[4] H. Friedrich, *Theoretical Atomic Physics* (Springer-Verlag, Berlin, 1990), p. 111.
[5] J.J. Sakurai, *Advanced Quantum Mechanics* (Addison-Wesley, Reading, MA, 1967), p. 15.
[6] D.Y. Smith, in *Handbook of Optical Constants of Solids*, edited by E.D. Palik (Academic, New York, 1985), p. 35.
[7] M. Mizushima, *Quantum Mechanics of Atomic Spectra and Atomic Structure* (Benjamin, New York, 1970), p. 91.
[8] C. Cohen-Tannoudji, B. Diu, and F. Laloë, *Quantum Mechanics* (Wiley, New York, 1977), p. 1308.
[9] See, e.g., G. Weinreich, *Solids: Elementary Theory for Advanced Students* (Wiley, New York, 1965), p. 136.
[10] M. Helm, W. Hilber, T. Fromherz, F.M. Peeters, K. Alavi, and R.N. Pathak, Phys. Rev. B **48**, 1601 (1993).
[11] G. Brozak, M. Helm, F. DeRosa, C.H. Perry, M. Koza, R. Bhat, and S.J. Allen, Jr., Phys. Rev. Lett. **64**, 3163 (1990).
[12] G. Burns, *Solid State Physics* (Academic, New York, 1990), p. 257.