New approach to study light-emission of periodic structures. Unveiling novel surface-states effects

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An accurate approach to calculate the optical response of periodic structures is proposed. Using the genuine superlattice eigenfunctions and energy eigenvalues, the eigenfunctions parity symmetries, the subband symmetries and the detached surface energy levels, we report new optical-transition selection rules and explicit optical-response calculations. Observed transitions that were considered forbidden, become allowed and interesting optical-spectra effects emerge as fingerprints of intra-subband and surface states. The unexplained groups and isolated narrow peaks observed in high resolution blue-laser spectra, by Nakamura et al., are now fully explained and faithfully reproduced.

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Although the fascinating phenomenon of light emission has been studied for more than a century, the main problem in calculating optical responses of semiconductor periodic structures using, for example, the golden rule

$$|\langle \psi | H_{\text{int}} | \psi \rangle|^2 |(E_i - E_f + i\hbar \omega)^2 + \Gamma_i^2| \quad (1)$$

where $H_{\text{int}}$ describes the light-matter interaction and $\omega$ the emitted photon frequency, has been the lack of explicit knowledge of the initial and final states $|\psi_i\rangle$ and $|\psi_f\rangle$ and of the corresponding energies $E_i$ and $E_f$. In the standard approaches (SAs) to periodic systems, based on models and theories for infinite periodic systems and the continuous subbands description of the standard approach by the most accurate discrete subbands description, we not only replace the continuous subbands description of the standard approach by the most accurate discrete subbands description, we also unveil the surface energy levels (see figure 1), responsible for the, so far, unexplained optical-spectra effects observed in high resolution experiments. At the same time we recover a truly quantum description of optical emissions in periodic structures.

We will show that the detachment of the surface energy levels, apparent in figure 1, is responsible for the groups of peaks observed by Nakamura et al. We will present new selection rules, based on the eigenfunctions’ symmetries and strongly dependent on the quantum numbers $\mu$ and $\nu$, the parity of $n$ and on the surface states. The “forbidden transitions” will become allowed. We will report here two types of selection rules. The first one, based on the eigenfunction’s parity symmetry, will reduce the number of evaluations from $N \simeq (n+1)^2n_c n_s$ to $N/2$, for a SL with $n$ unit cells, $n_c$ SBs in the conduction band (CB) and $n_s$ SBs in the valence band (VB). The second rule, based on the subband symmetry. This rule will reduce the number of evaluations to $\approx n_n n_s n_v/2$.

For simplicity we will refer here to type I SLs. The generalization is direct. It was shown in Ref.\cite{22} that the eigenvalues, for SL bounded by cladding layers like in figure\cite{22} can be obtained from

$$\Re (\alpha_n e^{-i n_k}) - \frac{k^2 - q_w^2}{2q_w k} \Im (\alpha_n e^{i n_k}) - \frac{k^2 + q_w^2}{2q_w k} \Im \beta_n = 0, \quad (2)$$

where $q_w$ and $k$ are the wave numbers at the left (right) and right (left) of the discontinuity point $z_L$ ($z_R$), $\alpha_n = U_n - \alpha^* U_{n-1}$ and $\beta_n = \beta U_{n-1}$ the $n$-cell transfer matrix elements, and $U_n$ the Chebychev polynomial of the second kind evaluated at the real part of the matrix element.
Here $\alpha_{o}$ is a normalization constant and $z$ any point in the $j+1$ cell, i.e. any point between $z_{j}$ and $z_{j+1}$, with $0 \leq j \leq (n-1)$. $\alpha_{j}$, $\beta_{j}$,... are the $j$-cells transfer-matrix elements and $\alpha_{o}$, $\beta_{o}$,...the matrix elements of $M_{o}(z,j)$ that connects the state vectors $\Phi(z_{j})$ and $\Phi(z)$, for $z_{j} \leq z \leq z_{j+1}$. The super-index $q$ refers to quasi-bound superlattice and $b = c, v$ refers to conduction and valence band. The super-index $q$ and the band index will be written only if they are necessary.

To evaluate the SL optical response, especially the photoluminescence (PL) for specific systems, we will consider the golden rule

$$\chi_{PL} = \sum_{\nu',\nu}\frac{f_{ch}}{(h\omega-E_{\mu,\nu}^{e}-E_{g}+E_{\mu',\nu'}^{e}+E_{B})^{2}+\Gamma_{\nu}^{2}},$$

with energies measured from the corresponding band edges. Here $E_{g}$ is the gap energy, $E_{B}$ the exciton binding energy, $\Gamma$ the level broadening energy and $f_{ch}$ the occupation probability.

The parity symmetries, for eigenfunctions of quasi-bounded SLs, are summarized as:

$$\Psi_{\mu,\nu}(z) = \begin{cases} (-1)^{\nu+1}\Psi_{\mu,\nu}(z) & for \ n \ odd \\ (-1)^{\nu+\mu}\Psi_{\mu,\nu}(z) & for \ n \ even \end{cases}.$$ (7)

These relations lead to the following symmetry selection rules (SSRs). For $n$ even, we have:

$$P[\mu + \nu] = P[\mu + \nu + 1].$$ (8)

Here $P[l]$ means parity of $l$. When $n$ is odd the SSRs are:

$$P[\mu + \nu] = P[\mu + \nu].$$ (9)

Similar relations hold for IR transitions, with the additional restrictions $\mu \geq \mu'$ and, whenever $\mu = \mu'$, we must also have $\nu > \nu'$, see Ref. 24. These rules, as mentioned before, effectively reduce the number of possible transitions to $N/2$. Depending on the number of subbands, this can be still a large number. To reduce even more the number of matrix-elements evaluations, we will introduce, some lines below, other rules related with the subband symmetry.

To test our approach, we will consider two specific examples, with results obtained with highest experimental resolution that we could find in the literature. In Ref. 21 the blue emitting SLs $(In_{x}Ga_{1-x}N)\{In_{y}Ga_{1-y}N\}^{N}$ with $x=0.2$, $y=0.05$ and different values of $n$, have been extensively studied. Some results, show spectral features, with groups of narrow spectral widths and peak separations of the order of 0.2nm ($\sim 0.12$meV), that could not be explained so far.

In the upper panel of figure 3, the PL spectrum, first published in Ref. 23, for a SL with $n=10$ and using a monochromator resolution of 0.016nm ($\sim 0.01$meV) is shown. Taking into account this SL parameters, and the appropriate electron and hole effective masses, we obtain the energy eigenvalues, the eigenfunctions and the PL spectrum plotted in the middle panel of figure 3. Some eigenfunctions $\Phi_{1,\nu}$, in the first SB of the CB, are plotted in figure 4. Notice that the eigenvalues $E_{1,10}$ and $E_{1,11}$, that correspond to the surface states $\Phi_{1,10}$ and $\Phi_{1,11}$, are detached from the others energy levels in the SB. To understand the structure of the optical response in figure 3, let us distinguish, in each subband $\mu$ of the CB, the surface energy levels $\{s_{\mu}\}$ from the remaining $n-1$ energy levels $\{g_{\mu}\}$. Similarly, the energy levels $\{s_{\mu'}\}$ from
slightly detached surface states \( \Psi^c \)
eigenfunctions \( \Psi^c \)

FIG. 4. Eigenfunctions and surface states. The eigenfunctions \( \Psi^c \) and \( \Psi^c_1 \) in the first subband of the CB of the blue emitting \( (In_{0.2}Ga_{0.8}N)_{10}\backslash In_{0.05}Ga_{0.95}N) \) SL with \( a=2.5\text{nm} \) and \( b=5\text{nm} \), bounded by GaN cladding layers.

FIG. 5. The subband symmetry. Because of this symmetry the envelopes of the eigenfunctions with indices \( (\mu, \nu) \) and \( (\mu, n+2-\nu) \) are the same. See for example \( \Psi_{2,2} \) and \( \Psi_{2,10} \). Here we plot the eigenfunctions \( \Psi_{2,\nu} \) for a SL with \( n=10 \), thus with \( \nu=1, 2, \ldots, 11 \).

the levels \( \{g\mu'\} \), in the subband \( \mu' \) of the VB. The transitions \( g1 \rightarrow g2' \) are responsible for the group of peaks with larger wavelengths, between 419.24nm and 420.747nm, the transitions \( g1 \rightarrow s2' \) and \( s1 \rightarrow g2' \), for the group of peaks in the middle, and the transitions \( s1 \rightarrow s2' \) for the isolated peak at the left.

This non-obvious resonant structure is a consequence of the presence of surface states, whose detachment determines the shift and the appearance of groups of peaks, as well as, of the isolated peak at a higher energy. It is clear that in order to observe this effect we need high-resolution experiments.

A rather general characteristic of the PL and IR spectra, measured or calculated, is the small number of peaks, much smaller than the \( N/2 \). One reason is, of course, the low experimental resolution. From the explicit calculations, we found out that, besides the parity symmetry, we have also the subband symmetry, glimpsed in Ref. [22], playing an important role in the relative values of the transition-matrix elements. In fact, when the surface levels detach, the matrix elements that fulfill the conditions

\[
|\mu - \mu'| = 1, 3, 5, \ldots
\]

and

\[
\langle \mu', \nu' | \frac{\partial}{\partial z} | \mu, \nu \rangle \text{ with } \nu + \nu' = n, \nu' = 1, 2, \ldots, \mu = 1, 2, \ldots,
\]

are leading order transitions. When the surface levels do not detach, the leading order transitions are

\[
\langle \mu', \nu' | \frac{\partial}{\partial z} | \mu, \nu \rangle \text{ where } |\mu - \mu'| = 1, 3, 5, \ldots
\]

\[
\nu + \nu' = n, n + 2.
\]

Because of the subband symmetry, the envelope curve of \( \Psi_{\mu,\nu} \) is similar to that of \( \Psi_{\mu,n+2-\nu} \), when the SSs do not...
clear that without the detachment of the surface energy
terms that contribute to the PL in the middle panel. It is
asymmetric confining potential, thus larger SSs detachment.
In the lower panel we plot separately, and indicate, the tran-

duction is from matrix-elements evaluations. For a PL spectrum, the
studied here the SSs detach, see figure 4.

The eigenfunctions in figure 5, correspond to a system
in the first case, and to \( | \Psi_{1,20} \rangle \) and \( | \Psi_{1,21} \rangle \) in s1 become localized at the opposite sides
of the superlattice. The same happens with \( \Psi_{1,20}' \) and
\( \Psi_{1,21}' \) to \( g_{21} \rightarrow g'_{21} \). Even so, using the SSRs and the LORs we end
up calculating 78 matrix-elements and with the spectrum
in the panel at the middle of figure 6. As shown in the
lower panel of this figure, the transitions \( E_{1,21} \rightarrow g_{22}' \)
and \( E_{1,20} \rightarrow g_{21}' \) lead to the most visible structures, referred
as “broaden emission lines” in [26]. In these graphs we

do not show the transition \( E_{1,21} \rightarrow \rightarrow g_{1}' \), which occurs
at higher energy.

Interesting surface-states effects were unveiled and the
eigenfunctions’ parity and subband symmetries role, on
the selection rules and leading order rules, were shown.
High-accuracy PL experimental results, with features
that could not be explained before, are now fully under-
stood. The improved optical response theory opens up
the possibility to enhance the optical techniques for spe-
cific applications. We expect that the relation between
surface states, cladding layers’ energy gap and the novel
group structure and isolated peak in the PL spectra, will
be further studied and experimentally confirmed.

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