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ELECTRONIC AND OPTICAL PROPERTIES OF QUASIPERIODIC FIBONACCI SUPERLATTICES

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RESUME : Les propriétés optiques d'une série de superréseaux quasiperiodiques de Fibonacci d'ordre croissant sont étudiées par spectroscopie d'excitation de la photoluminescence et comparées à un calcul de la structure de bande électronique.

ABSTRACT: The optical properties of a series of Fibonacci quasiperiodic superlattices of increasing order are studied by photoluminescence excitation spectroscopy and compared with calculations of the electronic band structure.

Most of studies concerning GaAs/GaAlAs superlattices (SL) have considered up to now periodic structures, but non periodic one raise interesting problems because there is no Bloch translation invariance and localization effects are expected. To begin with, we have focused our attention on Fibonacci quasiperiodic SL, which are attractive because using a simple recurrent law we can obtain series of increasing period starting from usual simple periodic SL and ending with a complete non periodic SL. Pioneering studies by Merlin et al. (1) have pointed out the very peculiar structural, dynamical and electronic properties that can be observed in these SL.

Our concern in this paper is with the electronic and optical properties of these Fibonacci quasiperiodic SL. In part I we explain briefly how a transfer matrix technique can be used to calculate the band structure of any non periodic heterostructure. In part II this method is applied to the case of the Fibonacci SL and we discuss the localization of the wave functions and show how it is related to the band structure. Finally in part III we present experimental results on a series of Fibonacci SL grown by molecular beam epitaxy (MBE), namely X-ray diffraction data and photoluminescence excitation spectroscopy. We discuss the selection rules of optical transitions and the coherence length of the electronic wave function.

1 TRANSFER MATRIX IN SEMICONDUCTOR HETEROSTRUCTURES

The transfer matrix technique is very common for studying non periodic systems (2) and we have applied it to the calculations of the electronic band structure of semiconductor SL (unpublished). In the effective mass approximation the envelop wavefunction can be written as the sum of a right propagating and a left
propagating plane wave of \( k \) vector determined by the energy. Using the continuity of the wave function and of the quantum current at the interfaces we can obtain a 2x2 transfer matrix between the projections of the wave function on these plane waves of any pair of elements (barrier or well) of the structure. To be specific in the case of transfer between two wells of equal width \( L_w \) separated by a single barrier of width \( L_b \), we obtain the following expressions for the elements of matrix \( T \):

\[
\begin{align*}
T_{11} &= \left[ \exp(i k_w L_w) \right] \left[ \cosh(k_b L_b) \right] - \left( 1/2 \right) \left( 1/\xi - \xi \operatorname{sh}(k_b L_b) \right) \\
T_{12} &= -\left( 1/2 \right) \left( 1/\xi + \xi \operatorname{sh}(k_b L_b) \right) \\
T_{21} &= T_{11}^* \\
T_{22} &= T_{11}^* \\
T_{21} &= T_{12} \\
T_{22} &= T_{11} \\
\end{align*}
\]

In these relations we have \( \xi = (k_w/k_b)(m_b/m_w) \) where \( k_w \) is the wave vector in the wells, \( k_b = i k_b \) with \( k_b \) the wave vector in the barrier where the effective mass \( m_w \) in the well and \( m_b \) in the barrier can be taken energy dependent to take into account non parabolic effects (3).

The electronic spectrum is determined (4) for given boundary conditions by the value \( X \) of the half trace of the total transfer matrix between the first and last wells of a period (finite or infinite) of the structure.

![Figure 1: Electronic band structure of Fibonacci SL. The groups of bands are indexed by reference to order \( l=1 \) (bands 2,4) and \( l=2 \) (bands 1,3,5).](image1)

![Figure 2: Electronic energy dispersion curves as a function of reduced Bloch index for Fibonacci SL for order \( l=1 \) to \( l=5 \) (the energy origin is arbitrary shifted for each curve).](image2)

II ELECTRONIC BAND STRUCTURE AND WAVEFUNCTION LOCALISATION

We recall that a quasiperiodic Fibonacci (5) SL of order \( l \) is constructed by \( l \) applications of the Fibonacci transformation law \( A \rightarrow AB \) and \( B \rightarrow BA \) starting from \( A \). We obtain the following sequences: \( A(1=0), AB(1=1), ABA(1=2), ABAB(1=3), \ldots \) and assign to sequence \( B \) the order \( l=-1 \). The half trace \( X_1 \) of the transfer matrix corresponding to Fibonacci sequence \( l \) is very easily obtained because the following recurrent relation holds (4): \( X_1 = 2X_{1-1} - X_{1-2} - X_{1-3} \) at a given energy. Fig.1 displays the band
structure of the electrons for Fibonacci SL with Born-von Karman boundary conditions in the energy range corresponding to the fundamental bands of SL A and B. We have chosen A and B to consist each of the same GaAs well and a Ga$_{1-x}$Al$_x$As barrier thinner in A than in B and of composition $x=0.23$. The number of bands is equal to the total number of basic elements of the corresponding sequence. Note that the complete overlap of the bands of SL A and B obtained with our choice of elements A and B gives states in a large energy range and wide gaps and this will be very convenient for optical studies. A very important particularity to be noticed is that there is on the contrary no overlap at all between the bands of the SL AB (1=1) and ABA (1=2) and that for larger order l all the bands fall in the energy range of either one of these two SL. As a consequence of this fact the localization of the wave function will be very simple to understand at any order l.

Using the method described in ref.4, it is convenient to study the electronic band structure of Fibonacci SL in an extended zone scheme in reciprocal space using a reduced Bloch index spanning a range of values independent of l (up to l infinite). We observe (see Fig.2) that as l is increased the main gaps are rapidly fixed altogether in energy and position in reciprocal space beyond $l=2$ (the new gaps which appear at higher order are much smaller) and our calculation of the wave function (unpublished) show that for any state at any order $l \geq 3$ its localization is either around a BAAAB if the state correspond in energy to a band of the SL ABAABA...($l=2$) or around a BAB if it corresponds to a band of the SL ABAB...($l=1$)(the exact pattern in the sequence where localization takes place depends on the state considered). The same is also true in the valence band for heavy holes and light holes: therefore for a given state corresponding to a value of the Bloch index the wave function is localized on the same pattern at the same position of the total sequence independently of the effective mass or the potential barrier height.

**III. OPTICAL PROPERTIES**

In the series of samples which have been grown by MBE the GaAs wells have a nominal width of 10 monolayers and the Ga$_{1-x}$Al$_x$As barriers ($x=0.23$) have a width of 11 monolayers in A and 24 monolayers in B. This series consisted of Fibonacci order $l=-1,0,...,5$ and $l=9$ and in order to compare SL with approximately the same total thickness ($\approx 0.65\mu m$) the Fibonacci sequence for $l \leq 5$ are repeated as necessary. X-ray diffraction spectra of two samples are displayed Fig.3. We have observed that as the order l increases the diffraction angle of the main peaks are rapidly fixed (unpublished).

Low temperature (2 K) photoluminescence spectra exhibit a single sharp line 5 meV wide at half maximum. Photoluminescence excitation spectroscopy was done with a dye laser tunable between 780 and 680 nm (pyridine 2 pumped by an Ar$^+$ laser). Fig 4 shows the different spectra for $l=1$ to 4 and $l=9$. The new minigaps beyond order $l=3$ being very small, no new feature can be resolved in the spectrum above this order.
The resolved excitonic peaks are assigned to transitions between electronic and light or heavy hole states with $\Delta k=0$ ($k$ being the Bloch index). This selection rule results from the quasi orthogonality of the electron and hole wave functions because the pattern on which they are localized is determined only by $k$ as explained above. Using notations of Fig.1, electronic states of mini-bands 1,3,5 (resp.2,4) are localized around a BABAB pattern (resp. 300 Å,240 Å) so that the coherence length cannot be related to the length of the sequence for which all expected transitions are observed (600 Å for $l=3$). The fact that the energy differences of the observed peaks is constant beyond $l=4$ proves that there is no effect due to the non negligible value of the $k$ vector of the light in comparison with the Brillouin zone extension in reciprocal space.

We wish to thank D. Paquet for many stimulating discussions.

Fig 3 (above): Diffraction spectra of two Fibonacci SL ($l=1,l=9$) obtained with CuKα radiation around the 002 order of GaAs.

Fig 4 (right): Photoluminescence excitation spectra of Fibonacci SL of order 1,2,3,4, 9 at $T=2$ K (excitation power 100 mW/cm²). The relative shift of the peaks at high order is explained by a two percent variation of the aluminium concentration. The peaks are assigned to electrons (E) and heavy (H) or light (L) holes excitonic transitions and the labels correspond to the indices of Fig.1.

REFERENCES