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Fibonacci invariant and electronic properties of $GaAs/Ga_{1-x}Al_xAs$ quasiperiodic superlattices

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A theoretical study of the electronic properties of $GaAs/Ga_{1-x}Al_xAs$ quasiperiodic Fibonacci superlattices reveals the striking differences to be expected in the band structure, wave-function localization, and optical transitions of superlattices having quasiperiodic modulation either in the barrier width or in the barrier height. We show how these differences are related to the value of the Fibonacci invariant. The results of photoluminescence excitation spectroscopy experiments are also presented.

Periodic semiconductor superlattices (SL's) have been the most intensively studied up to now. But physical systems with controlled nonperiodicity along one direction (the growth axis) can also be easily produced with such SL's. For example, SL's exhibiting quasiperiodic modulation[1] or intentional random disorder[2] have already been grown recently and some of their properties have been investigated. More generally, the electronic properties of such unidimensional systems, which are not invariant by Bloch translation, have been considered in numerous theoretical works which have been devoted to the study of the distinctive features of their band structure and to the localization of their electronic states either in the random[3] or, more recently, in the quasiperiodic[4,5] cases.

We are concerned with $GaAs/Ga_{1-x}Al_xAs$ quasiperiodic Fibonacci SL's. In this system recent experiments have been mainly focused on the study of their structural[6] and dynamical[1,7] properties. To our knowledge only a few studies are available on their electronic and optical properties.[8,9] Therefore we have developed a model to understand the electronic band structure of these SL's: it uses the transfer-matrix technique and it is based on the envelope-function approximation which is known to be quite successful for periodic structures.[10] We show how $GaAs/Ga_{1-x}Al_xAs$ Fibonacci SL's differing by their electronic properties can be identified by examination of the Fibonacci invariant. In particular, we propose a new type of Fibonacci quasiperiodic modulation in which there is a completely delocalized electronic state. We have not yet proved experimentally the existence of this, but we present data of photoluminescence excitation spectroscopy[9] which are in good agreement with our envelope-function calculation of bulk electronic states.

Let us recall that a Fibonacci sequence S_l of order l is obtained by l successive applications of the transformation rule $A \rightarrow AB$ and $B \rightarrow A$. Starting from $S_0 = A$ we obtain $S_1 = AB$, $S_2 = ABA$, $S_3 = ABAAB \cdots$ and shall also consider $S_{-1} = B$. A most important consequence of this rule is self-similarity: We obtain again sequence S_{l-k} just by replacing in S_l each complete S_{k+1} sequence by A and each S_k by B. When $k = 1$ this gives the Fibonacci recurrent relation:

$$S_l = S_{l-1}S_{l-2} \ . \tag{1}$$

The sequence $S_l (l \geq 2)$ comprises F_l elements A and F_{l-1} elements B, F_l being the lth Fibonacci number given by the recurrent law $F_l = F_{l-1} + F_{l-2}$ starting with $F_0 = F_1 = 1$. As l increases the ratio F_l/F_{l-1} converges toward $\tau = (1+\sqrt{5})/2$ which is known as the golden mean.

In this work each of the two basic elements A and B are composed of a GaAs well and a $Ga_{1-x}Al_xAs$ barrier: We have chosen the wells to have the same width but the barriers to have either different widths (case α) or different heights—i.e., different Al composition—(case β). These two cases will appear to be representative of two possible types of quasiperiodic SL's differing by their localization properties. In case α all the electronic states are localized on rather short sequences of elements A and B, whereas in case β one single state is completely delocalized throughout the structure.

The transfer-matrix technique is a very common method for studying nonperiodic systems,[11] and we have used it in the frame of the envelope-function approximation,[10] whereas Fibonacci quasiperiodic unidimensional systems have been rather studied with tight-binding methods. In each slab of the SL we write the envelope wave function as a linear combination of right and left propagating plane waves. The projection coefficients obtained at any couple of points are related by a 2×2 matrix which can be written as follows:

$$T = \begin{pmatrix} X+iY & Re^{-i\psi} \\ Re^{i\psi} & X-iY \end{pmatrix} . \tag{2}$$

If the two points are situated at the middle of GaAs wells of equal width l_w separated by a single barrier of width L_b and height V_b, we have $\psi = \pi/2$ and

$$X = \cos(k_w l_w)\cosh(\kappa_b L_b) - (\xi - \xi^{-1})\sin(k_w l_w)\sinh(\kappa_b L_b)/2 \ , \tag{3}$$

$$Y = \sin(k_w l_w)\cosh(\kappa_b L_b) + (\xi - \xi^{-1})\cos(k_w l_w)\sinh(\kappa_b L_b)/2 \ , \tag{4}$$

$$R = (\xi^{-1} + \xi)\sinh(\kappa_b L_b)/2 \tag{5}$$

with $k_w \equiv \hbar^{-1}(2m_w\varepsilon)^{1/2}$, $\kappa_b = \hbar^{-1}[2m_b(V_b - \varepsilon)]^{1/2}$, and $\xi = (k_w/\kappa_b)(m_b/m_w)$, ε being the energy (counted from

the well bottom), m_w and m_b the effective masses in the well and in the barrier which can be taken as energy dependent in order to take into account the effects of nonparabolic dispersion.[10] The elements X, Y, and R depend on the energy ε and are related by $X^2+Y^2-R^2=1$ (the matrix is unimodular). Such a matrix can be considered for the conduction band or for the valence band (heavy or light holes) as well. If the barrier corresponds to the basic element A or B we use the corresponding subscript for the transfer matrix and its elements. The matrix T_l across all the barriers and wells of sequence S_l is obtained by the recurrent law $T_l=T_{l-2}T_{l-1}$ ($l\geq 1$) resulting from relation (1) starting with $T_{-1}=T_B$ and $T_0=T_A$. A recurrent relation for the half trace X_l is easily deduced: $X_1=2X_{l-1}X_{l-2}-X_{l-3}$ for $l\geq 2$. The band structure of a SL made of sequence S_l is simply obtained—when Born-von Kármán periodic conditions are used—by the condition $|X_l|\leq 1$. From the recurrent relation between successive X_l written above it follows[12] that there is a quantity I:

$$I=-1+X_l^2+X_{l-1}^2+X_{l-2}^2-2X_lX_{l-1}X_{l-2}\ , \qquad (6)$$

which is an invariant independent of order l. In a periodic SL (obtained here by taking $A=B$) I is trivially equal to zero at any energy. The larger the invariant I, the more strongly the quasiperiodicity will affect the properties of the SL: reduction of the bandwidths, increase of localization. We have found that the invariant I given by Eq. (6) has in fact a very simple expression involving only the elements of the two matrices T_A and T_B:

$$I=(Y_AR_B-Y_BR_A)^2\ . \qquad (7)$$

The invariant I is energy dependent here, which is not the case in most models studied theoretically.[4,12] The electron energy dependence of I is displayed in Fig. 1. Note the great difference between cases α and β. In case α, I is large and decreases with energy. In case β, I is smaller and is equal to zero for one value of energy. This is obtained for a very wide range of the SL parameters. If we write $X_l=\cos(\theta_l)$ using Eq. (6) the condition $I=0$ is equivalent for $l\geq 1$ to $\theta_l=\theta_{l-1}+\theta_{l-2}$ (provided that $|X_0|\leq 1$ and $|X_{-1}|\leq 1$ which is verified here, i.e., θ_{-1} and θ_0 are real numbers). This proves that, for this energy, there is an allowed state at any order l.[12]

The electrons band structure is presented in Fig. 2. As is well known[5] the number of bands is equal to the total number of elements of sequence S_l which is equal to F_{l+1}. It is obvious that large differences in the width of the bands and of the gaps are observed between cases α and β. This can be explained by the invariant: When I is close to zero the bands remain wide and the gaps small as order l increases, though ultimately at infinite order the spectrum will in both cases become a Cantor set of Lebesgue measure zero.[4] More quantitatively, it has been shown that there is a scaling[12] of the bands by the quantity

$$\lambda=[1+4(1+I)^2]^{1/2}+2(1+I) \qquad (8)$$

between order l and order $l+3$.[13] Because I is energy dependent, there are some fluctuations in the scaling, but the band structures of Fig. 2 are in agreement with the predictions of relation (8).

It is easy to check that Eq. (6) relating the successive X_l to invariant I is obeyed by the following cycle of period six: $0,0,-(1+I)^{1/2},0,0,+(1+I)^{1/2},\ldots$. The value 0

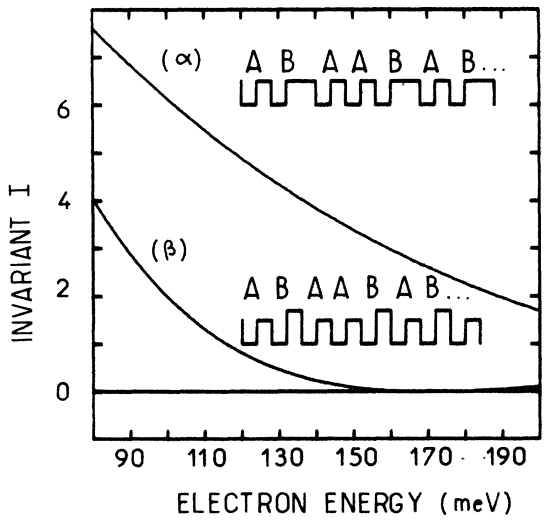

FIG. 1. Electron energy dependence of the invariant I corresponding to the sketched Fibonacci SL (cases α and β). The GaAs wells are 10 monolayers (ML) (1 ML=2.83 Å) thick. The $Ga_{1-x}Al_xAs$ barriers have the following parameters: A (α and β), 11 ML and $x=0.25$; B (α), 24 ML and $x=0.25$; B (β), 11 ML and $x=0.35$.

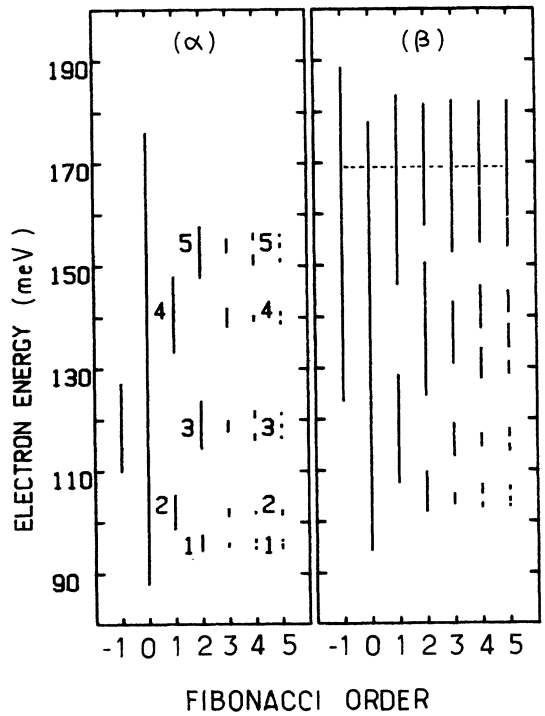

FIG. 2. Electron band structure of Fibonacci SL as a function of Fibonacci order l. Case α (left) labels define groups of bands in energy relation with those of SL $l=1$ and $l=2$. In case β (right) the dotted line indicates the energy for which the invariant I is zero.

corresponds to bands, and because I cannot be negative [see relation (7)] the values $\pm(1+I)^{1/2}$ correspond to gaps. So, at a given energy, when I is large allowed states cannot be obtained for three successive orders l.[14] In case α, in spite of an optimum overlap between the fundamental bands of SL's A ($l=0$) and B ($l=-1$), there is already no overlap between the bands of SL's AB ($l=1$) and ABA ($l=2$). Note that these two SL's have, respectively, either single-A or double-A pattern. Accordingly, at higher order l the bands split with the following rules: (i) The number of bands in the energy ranges of the bands of SL AB (respectively, ABA) denoted as 2,4 (respectively, 1,3,5) on Fig. 2 is equal to the number of single-A (respectively, double-A) patterns in sequence S_l; (ii) in each energy range the Fibonacci series determines the increase of the number of bands. This is a consequence of self-similarity.

All these properties are also reflected in the localization of the wave function. This has been checked by direct calculations[15] using the transfer matrices T_A and T_B. In case α the energy range of an allowed state in the spectrum determines the pattern on which the wave function is localized: single-A pattern (BAB groups) in the energy range 2,4 or double-A pattern ($BAAB$ groups) in the energy range 1,3,5 (see Fig. 2). In case β at the energy at which $I=0$ it has been verified that the wave function is delocalized throughout the structure. This discussion shows the link between the wave function localization (real space) and the band structure (reciprocal space) in different types of quasiperiodic modulation and how these properties are related to the value of the invariant I.

In spite of differences in barrier heights and effective masses the invariant I is roughly similar for electrons, light holes, and heavy holes, and so is therefore the wave-function localization for a given state number n. Indeed we have checked that the wave functions have then a maximum on the same pattern type and at the same position in the sequence irrespective of the carrier type.[15] The wave functions of the same carrier type and different n being orthogonal, it follows that the electron and heavy- or light-hole envelope functions are quasiorthogonalized so that we can predict quasiselection rules $\Delta n \cong 0$ for the optical transitions when I is large in spite of no translation invariance.

Series of samples of increasing order l have been grown by molecular-beam epitaxy. In order to compare SL's of different l but similar thickness ($\cong 0.65$ μm) and to fit the experimental results with our calculations which use Born-von Kármán periodic boundary conditions, the Fibonacci sequences S_l are repeated as necessary. The structures have been characterized by x-ray diffraction,[9] and photoluminescence excitation spectroscopy experiments were performed at liquid-He temperature using a dye laser. The spectra are displayed on Fig. 3 for both cases: the transitions are in good agreement with our calculation based on the model described above (we use the parameters given in Ref. 16 and a constant exciton binding energy of 10 meV). As expected the transitions observed in case α for orders $l=3,4$ and beyond (not shown in the figure) are explained by the transitions of orders $l=1$ and $l=2$ and labeled accordingly. Nothing changes

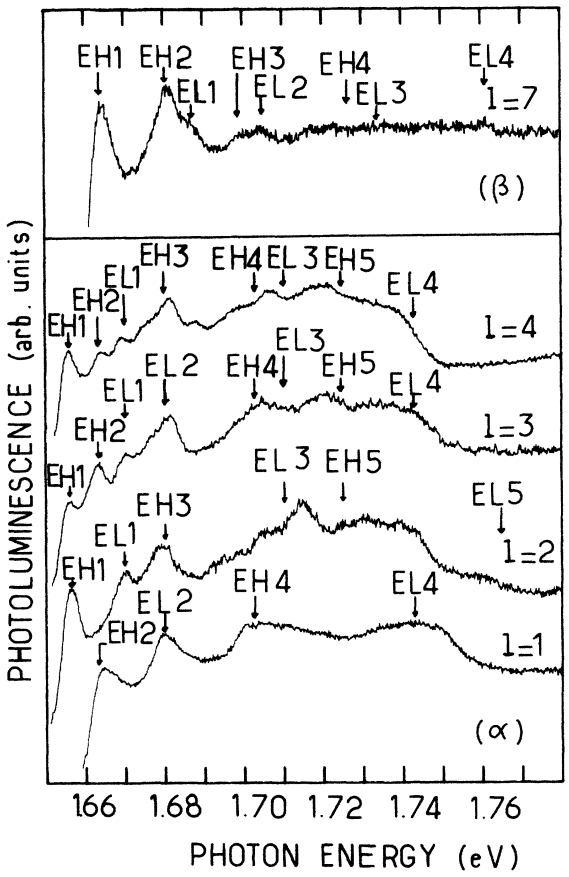

FIG. 3. Photoluminescence excitation spectra of Fibonacci SL of order 1,2,3,4 in case α (bottom) and of order 7 in case β (top) at $T=2$ K. Typical power density is 100 mW/cm^2. Arrows indicate calculated electron-heavy-hole (EH) and electron-light-hole (EL) transition energies for $l=1$ and $l=2$. Labels refer to Fig. 2. The spectra are aligned on the EH1 peak to eliminate the effects of small Al concentration and thickness fluctuations from sample to sample.

at higher order because the new gaps are small and the oscillator strength of new transitions are certainly weak. These observations are in agreement with the predicted selection rules. So we believe that the main features of these spectra can be explained by these bulk state transitions (i.e., calculated in periodized structures) and we do not find any clear evidence in our SL's of the surface states predicted by Nori and Rodriguez.[17] The data of Ref. 8 do not show fine structures at an energy scale comparable to this work, and no detail has been given on the model and the spectrum interpretation. We do believe that our method of following the transitions at successive orders l helps to give strong support to the line assignments. In case β (displayed here only for $l=7$) there is less structure on the high-energy side of the excitation spectrum in agreement with weaker band-gap opening and weaker selection rules expected when the invariant I is small. We do not claim, however, that this is an experimental evidence of delocalization.[18]

The studies presented here show the importance of the

invariant I, which can be calculated in unidimensional quasiperiodic Fibonacci structures in order to understand the band structure, the wave-function localization, and how both are related. We have shown that GaAs/$Ga_{1-x}Al_xAs$ Fibonacci SL's with different types of quasiperiodic modulation are expected to have different electronic properties. We have presented preliminary optical studies of these structures. The observed transitions are well explained by a model using the envelope-function approximation. We hope that the extended electronic state predicted when the quasiperiodic modulation occurs on the barrier height will be seen in further optical or transport experiments.

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[13]Direct examination of the periodic case $A=B$ reveals that the scaling is then $F_{l+3}/F_l=\tau^3$, which is the expected value of λ when $I=0$. This value is equal to the ratio of the number of bands of the two SL's of order l and order $l+3$ because when $A=B$ the gaps are equal to zero. It seems therefore not possible to have scaling by a lower value than τ^3 as could be *a priori* obtained using (8) with a negative value of I.

[14]Again a direct examination of the periodic case $A=B$ indicates that this six-order cycle (here $0,0-1,0,0+1,\ldots$) and this succession band, band, gap, . . . is closely related to particularities of the Fibonacci series for which the successive numbers (giving the number of bands) are odd, odd, even (twice an odd number), odd, odd, even (twice an even number), . . . : 1,1,2,3,5,8, ...

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[16]We use the following: band-gap variation $E_g(Ga_{1-x}Al_xAs)=1.519+1.247x$ (eV); conduction-band offset $\Delta E_c=0.67\Delta E_g$; conduction-band effective mass at $k=0$, $m_e=0.0667m_0$; valence-band effective masses $m_{hh}=(0.34+0.42x)m_0$ and $m_{lh}=(0.094+0.056x)m_0$; spin-orbit splitting $\Delta=0.341-0.066x$ (eV).

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