Electronic Structure of a Quasiperiodic Superlattice

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The electronic structure of a quasiperiodic superlattice with slabs of A- and B-type layers is studied within a tight-binding model by use of the transfer matrix and a perturbative approach. It is found that the band gaps vary strongly within the two-dimensional Brillouin zone and, in the case of more than one layer in a slab, there are a few eigenvalues for each $\mathbf{k}_\parallel$ for which the eigenstates appear to be extended in the quasiperiodic direction. The latter is true even for the Fibonacci chain with more than one atom per cell.

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Recently Merlin *et al.*[1] have fabricated quasiperiodic superlattices (QPS) of GaAs-AlAs with layers of GaAs and AlAs arranged in a Fibonacci sequence (FS). Similar metallic superlattices of Cu-Nb have been fabricated by Hu *et al.*[2] Vacancy-stabilized phases of systems like Al-Pd, where planes of Al, Pd, and vacancies are arranged in a FS, are other examples.[3] The aim of this paper is to investigate the electronic structure of such systems. Intuitively, one would expect the electronic structure normal to the layers to be similar to the Fibonacci chain $ABAABABA\ldots$, whose spectrum is a Cantor set and for which the states are critical.[4,5] However, the behavior of the spectrum when each A (B) is replaced by more than one A (B) in the Fibonacci chain is not known. This is relevant for the superlattice as each slab can have more than one layer.[1,2] Furthermore the dependence of the spectrum on (i) the wave vector parallel to the layers $\mathbf{k}_\parallel$ and (ii) the layer atomic arrangement is not *a priori* clear. We address these questions with the aid of a single-band tight-binding Hamiltonian on a superlattice with (001) layers of the simple cubic[6] (sc) and the face centered cubic (fcc) lattices. Two complimentary methods are used: (a) the transfer-matrix method which is very effective in obtaining accurate numbers and (b) a perturbative scheme which allows us to obtain approximate *analytical* expressions for the band gaps and their positions. These serve a useful role in understanding the results obtained in (a). We demonstrate that the spectrum depends strongly on $\mathbf{k}_\parallel$ and that there are a few states for each $\mathbf{k}_\parallel$ which appear to be *extended* along the QP direction also.

Consider a superlattice with slabs of N layers of A and M layers of B atoms arranged in a FS. Then the equation of motion for an electron in the tight-binding model is

$$[E-\epsilon(\mathbf{R})]\psi(\mathbf{R})=\textstyle\sum_{\boldsymbol{\delta}} t(\mathbf{R},\mathbf{R}+\boldsymbol{\delta})\psi(\mathbf{R}+\boldsymbol{\delta}), \tag{1}$$

where $\boldsymbol{\delta}$ runs over all the nearest neighbors. Using $\mathbf{R}=(\mathbf{R}_\parallel,s)$ with $\mathbf{R}_\parallel$ denoting the component parallel to the layers and s the layer index, we have $\epsilon(\mathbf{R})=\epsilon(s)$ for the site energies, and $t(\mathbf{R},\mathbf{R}+\boldsymbol{\delta})=t^{\parallel}(s)$ for the intralayer and $t(\mathbf{R},\mathbf{R}+\boldsymbol{\delta})=t^{\perp}(s,s+1)$ for the interlayer hopping integrals. On taking the Fourier transform of (1) with respect to $\mathbf{R}_\parallel$, we get

$$[E-\epsilon(s)-T_s^{\parallel}]\psi(\mathbf{k}_\parallel,s)=\sum_{l=\pm 1} T^{\perp}_{s,s+l}\psi(\mathbf{k}_\parallel,s+l), \tag{2}$$

where $T_s^{\parallel}=2t^{\parallel}(s)(\cos k_x a+\cos k_y a)$ and $T^{\perp}_{s,s\pm 1}=t^{\perp}(s,s\pm 1)$ for the sc lattice, and

$$T_s^{\parallel}=4t^{\parallel}(s)\cos(k_x a/2)\cos(k_y a/2),$$

$$T^{\perp}_{s,s\pm 1}=2t^{\perp}(s,s\pm 1)[\cos(k_x a/2)+\cos(k_y a/2)]$$

for the fcc lattice with the lattice constant a. In the matrix form we can write (2) as

$$\begin{pmatrix}\psi(\mathbf{k}_\parallel,s+1)\\ \psi(\mathbf{k}_\parallel,s)\end{pmatrix}=\begin{pmatrix}\dfrac{E-T_s^{\parallel}-\epsilon(s)}{T^{\perp}_{s,s+1}} & -\dfrac{T^{\perp}_{s,s-1}}{T^{\perp}_{s,s+1}}\\ 1 & 0\end{pmatrix}\begin{pmatrix}\psi(\mathbf{k}_\parallel,s)\\ \psi(\mathbf{k}_\parallel,s-1)\end{pmatrix}. \tag{3}$$

The above transfer matrix $\mathcal{T}$ can be used to obtain necessary information about the electronic spectra much as in the 1D case. If the sequence of layers repeats after p slabs, then the spectrum is given by the product M_p of transfer matrices of p slabs (here p is taken to be a Fibonacci number).

We first consider the case $N=M=1$ (case I) for which the site energies $\epsilon(s)$ are ϵ_A (ϵ_B) for A (B) layers. The hopping integrals $t^{\parallel}(s)$ within a layer are either t_{AA} or t_{BB} whereas the interlayer hopping integrals $t^{\perp}(s,s+1)$ take on two values t_{AA} (here taken to be the same as for the A layers) or t_{AB} which is chosen to be $(t_{AA}t_{BB})^{1/2}$. There are four different transfer matrices

corresponding to the local sequences *ABA*, *BAB*, *AAB*, and *BAA* of layers. It is convenient to define three new transfer matrices

$$\mathcal{T}_1' = \mathcal{T}(t_{AB},t_{BA})\mathcal{T}(t_{BA},t_{AB}),$$
$$\mathcal{T}_2' = \mathcal{T}(t_{AA},t_{AB}), \quad (4)$$
$$\mathcal{T}_3' = \mathcal{T}(t_{AB},t_{BA})\mathcal{T}(t_{BA},t_{AA}),$$

where the arguments of $\mathcal{T}$ denote $t^{\perp}(s,s-1)$ and $t^{\perp}(s,s+1)$. Then starting with $\mathcal{T}_1'$ (corresponding to $p=2$), transfer matrices for successive FS can be obtained by the following replacements: $\mathcal{T}_1' \rightarrow \mathcal{T}_2'\mathcal{T}_3'$, $\mathcal{T}_2' \rightarrow \mathcal{T}_3'$, and $\mathcal{T}_3' \rightarrow \mathcal{T}_2'\mathcal{T}_1'$. Since the determinant of the transfer matrix is unity, the allowed energy regions are obtained by the trace condition $x_p = \frac{1}{2}\mathrm{tr}M_p \leq 1$.

Figure 1 shows the results for the sc superlattice along the three high-symmetry directions in the 2D Brillouin zone with $p=9$ and $\epsilon_A=-0.25$, $\epsilon_B=0.25$, $t_{AA}=-1.0$, and $t_{BB}=-1.2$ (in units of $|t_{AA}|$). Although for a given $\mathbf{k}_{\parallel}$ triadic Cantor structure persists as in the case of the QP chain (QPC), the dependence of gaps on $\mathbf{k}_{\parallel}$ is strong.[6] Whereas at the Γ point the gaps are quite small, they are large around the M point. There is a crossover of the largest gap from the lower side to the upper side as we go from Γ to X or the M point. These features will be explained later with use of an approximate analytical scheme for the band gaps.

To illustrate the effect of more than one layer in each slab (case II) we have taken for simplicity either $\Delta t = t_{BB}-t_{AA}=0$ or $\Delta\epsilon=\epsilon_B-\epsilon_A=0$. Also when $\Delta t \neq 0$ the interlayer hopping integrals are assumed to take only two values, namely t_{AA} and t_{BB} with $t_{A\rightarrow B}=t_{BB}$ and $t_{B\rightarrow A}=t_{AA}$. With this choice it is possible to write for the pth $(p>2)$ FS the recursion relation $M_p = M_{p-2}M_{p-1}$. Here M_0 and M_1 are the products of M and N transfer matrices for B and A layers, respectively. The above recursion relation allows us to use the method of trace map for which

$$I = x_{p+1}^2 + x_p^2 + x_{p-1}^2 - 2x_{p+1}x_p x_{p-1} - 1$$

is a conserved quantity. In the simplest case for the sc lattice with $t_{AA}=t_{BB}=t$ and $N=M=1$,

$$I_1 = (\epsilon_B-\epsilon_A)^2/4t^2, \quad (5)$$

which is the same as for the QPC and interestingly is independent of $\mathbf{k}_{\parallel}$. The spectrum shown in Fig. 2(a) is thus the same for the entire 2D Brillouin zone except for an energy shift.

For $M=N=2$,

$$I_2 = I_1 \frac{\{(w/2)^2 - [2t(\cos k_x a + \cos k_y a) - E]^2\}^2}{t^4}, \quad (6)$$

where we have used $\epsilon_B = -\epsilon_A = w/2$. The fact that the dependence of I_2 on $\mathbf{k}_{\parallel}$ appears in the above form suggests that the gaps remain the same for different values of $\mathbf{k}_{\parallel}$, except for a shift in the energy. This is in fact borne out by our numerical calculations. The effect of doubling the thickness of the slabs is to introduce [Fig. 2(b)] mini bands, each of which develops the Cantor-set structure as the FS progresses. An important difference is that I_2 is zero at $E = 2t(\cos k_x a + \cos k_y a) \pm w/2$ and is therefore an eigenstate for successive FS. For $N=M=3$,

$$I_3 = I_1(1-D_A^2)^2(1-D_B^2)^2, \quad (7)$$

where $D_i = (\epsilon_i + T_s^{\parallel} - E)/t$ with $i=A,B$. Again, I_3 vanishes at $D_A^2=1$ or $D_B^2=1$. For $w=|t|=1$, in both these cases the gap vanishes at $E=-3.5$ and -4.5. Therefore

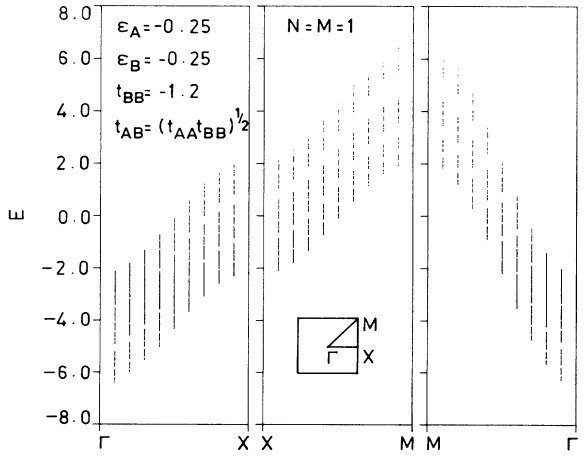

FIG. 1. Allowed energy states for a (001) simple cubic Fibonacci superlattice along the three high-symmetry directions in the 2D Brillouin zone. N (M) represents the number of A (B) layers in a slab. Energies are in units of $|t_{AA}|$.

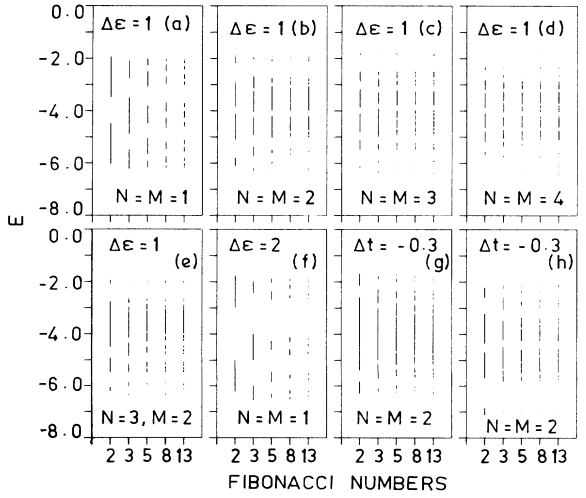

FIG. 2. Allowed energy states for a (001) sc superlattice at $\mathbf{k}_{\parallel}=0$. (a)–(f) $\Delta t=0$; (g),(h) $\epsilon_A=\epsilon_B=0$. (g) corresponds to the spectrum of a chain shifted by -4.0.

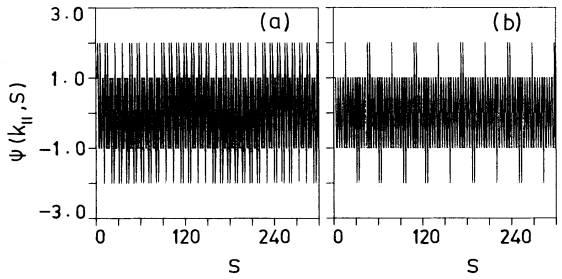

FIG. 3. Variation of the wave function on successive layers for $E=-3.5$. (a) $N=M=2$, (b) $N=M=3$. $\psi(0)=\psi(1)=1$.

we expect the corresponding states to be extended, as indeed we found them to be in our calculations. This is shown in Fig. 3. (We have checked this up to 2500 layers.) For $N=M=3$, I_3 vanishes also at $E=-2.5$ and -5.5. Similar states are also found for $N=M=4$. It appears that the number of such states increases with N,M. Also close to such energies several states are almost periodic. The envelope of the corresponding wave function has the character of a standing wave whose "wavelength" decreases as one moves away from energies for which $I=0$. From these results we can say in general that *for $N,M>1$, there are energies for each $\mathbf{k}_\parallel$ where the gap disappears and for which the states are extended.*

The two major gaps move toward the edges of the spectrum with increasing N and M; and their magnitude appears to be insensitive to the number of layers, and it scales approximately with $\Delta\epsilon$ [compare Figs. 2(a) and 2(f)]. Similar results have been obtained for $N\neq M$ and, as an example, Fig. 2(e) shows the results for $N=3$ and $M=2$. In the case when $\Delta\epsilon=0$ and $\Delta t\neq 0$, for a given $\mathbf{k}_\parallel$, the spectrum is asymmetric about $E=0$ for the QPS in contrast to the QPC where it is symmetric. This is because the intralayer hopping integrals act as site energies of an "equivalent chain" problem [see Eq. (13)]. For the same reason the gaps are also much larger as compared to the QPC [see Fig. 2(g) and 2(h)].

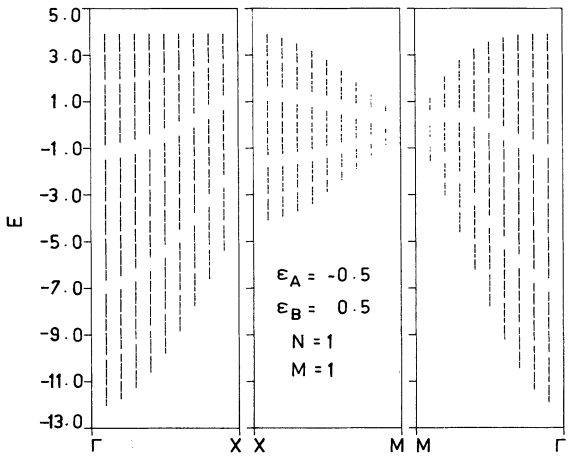

FIG. 4. Same as in Fig. 1 for a (001) fcc superlattice with $\Delta t=0$.

The spectrum of an fcc QPS with $N=M=1$ and $\Delta t=0$ is shown in Fig. 4. It can be seen that the gaps depend on $\mathbf{k}_\parallel$ in contrast to the sc case. The most dramatic situation arises at the M point where the spectrum has just two points separated by $\Delta\epsilon$, one corresponding to all the states on A sites and the other to all the states on B sites, their ratio being the golden mean $\tau=(\sqrt{5}+1)/2$. These are the two singularities in the spectrum of the QPS corresponding to the singularities in the density of states of the pure A or B component with the fcc lattice.

These results are better understood by an approximate analytical scheme to obtain the band gaps. As the method is perturbative in nature, the actual values of the gaps are good only for small or moderate Δt and/or $\Delta\epsilon$. However, in the absence of any analytical scheme to calculate the exact band gaps, our method is very useful. Following our earlier work,[5,7] (2) can be cast in the following form by Fourier transformation with respect to s. For the sc lattice we have

$$E\psi(\mathbf{k}_\parallel,k_z)=\int dk'\{\epsilon(k')+[\exp(-ik_za)+\exp(i(k_z-k')a)]t^\perp(k')+2(\cos k_xa+\cos k_ya)t^\parallel(k')\}\psi(\mathbf{k}_\parallel,k_z-k'), \quad (8)$$

where $t^\perp(k')$, $t^\parallel(k')$, and $\epsilon(k')$ are the structure factors of the interlayer, intralayer, and site integrals. These can be obtained by projection method for any kind of aperiodic sequence following Refs. 5 and 7. This enables us to write (8) as

$$E\psi(\mathbf{k}_\parallel,k_z)=\sum_{m,n}U_{nm}\psi(\mathbf{k}_\parallel,k_z-q_{nm}), \quad (9)$$

where $q_{nm}=2\pi(m+n\tau)/(M+N\tau)a$, $0\le q_{nm}<2\pi$, denote the reciprocal lattice vectors in the z direction (m,n integers) and U_{nm} is given by the expression within the curly braces with $k_z=q_{nm}$ in (8). It is clear that the band gaps occur at $k_z=q_{nm}/2$ and to the lowest order these can be obtained by evaluation of (9) at $k_z=\pm q_{nm}/2$ as in the periodic case. The magnitude ΔE_{nm} of the gap is simply $2|U_{nm}|$. Here we consider two cases corresponding to the results presented in Figs. 1 and 2 with $N=M$. Following Refs. 5 and 7 we have for case I

$$t_{nm}^{\perp}=\{2t_{AB}\exp(-2i\theta_{nm})\sin\theta_{nm}+t_{AA}\exp[-i(1+4\tau)/\tau\theta_{nm}]\sin(\theta_{nm}/\tau)\}/[(1+\tau)\theta_{nm}], \quad (10)$$

and in general for cases I and II

$$f_{nm}=\sum_{\lambda=0}^{N-1} e^{-i\lambda q_{nm}a}[f_1 e^{-i\theta_{nm}(2+\tau)}\sin(\theta_{nm}\tau)+f_2 e^{-i\theta_{nm}}\sin\theta_{nm}]/[N\theta_{nm}(1+\tau)], \tag{11}$$

where $f_{nm}=t_{nm}^{\parallel}(\epsilon_{nm})$ with $f_1=t_{AA}(\epsilon_A)$ and $f_2=t_{BB}(\epsilon_B)$, and $\theta_{nm}=\pi(m-n)/(1+\tau)$.

Using Eqs. (8)–(11), we obtain for case I

$$\Delta E_{nm}=4\{(\Delta t^{\perp}\sin 2\theta_{nm})^2+[\Delta t^{\parallel}(\cos k_x a+\cos k_y a)+\Delta\epsilon/2]^2\sin^2\theta_{nm} + \{\cos[(q_{nm}a+2\theta_{nm})/2]\}\sin 2\theta_{nm}\sin\theta_{nm}[2\Delta^{\perp}\Delta t^{\parallel}(\cos k_x a+\cos k_y a)+\Delta t^{\perp}\Delta\epsilon]\}^{1/2}/[(1+\tau)\theta_{nm}], \tag{12}$$

and for case II

$$\Delta E_{nm}=2|\Delta E|\,[\sin(\theta_{nm})/(1+\tau)\theta_{nm}]\,|\sin(Nq_{nm}a/2)/N\sin(q_{nm}a/2)|, \tag{13}$$

where

$$E_i=\epsilon_i+2t_{ii}[\cos k_x a+\cos k_y a+\exp(iq_{nm}a/2)], \qquad i=A,B;$$

$\Delta E=E_A-E_B$, and $\Delta t^{\perp}=t_{AB}-t_{AA}$.

From (12), the dependence of ΔE_{nm} on $\mathbf{k}_{\parallel}$ is clearly seen. Also its dependence on $\Delta\epsilon$, $\Delta t^{\parallel}$, and $\Delta t^{\perp}$ is not simple. At the Γ point the $\Delta\epsilon$, $\Delta t^{\parallel}$, and $\Delta t^{\perp}$ terms compete leading to small gaps, whereas at the M point they add and hence the dominant gaps are large. The predicted values of all the dominant gaps at the Γ point are found to be in good agreement[8] with those obtained by the transfer-matrix method. At the M point there is a 15% mean error which is expected in the perturbative theory. However, (12) predicts all the features correctly including the reversal of the dominant gaps. For the case $\Delta t=0$, it follows from (13) that ΔE_{nm} is independent of $\mathbf{k}_{\parallel}$ and it scales with $\Delta\epsilon$. Also for $N>1$ since $0\le q_{nm}<2\pi$, there are N sets of bands, the first one ending at $M=N=1$, where $q_{11}=2\pi/Na$. The dominant gaps now occur at $(m,n)=(1,0)$ and $(0,1)$ at the lower end and $(N-1,N)$ and $(N,N-1)$ at the upper end. Hence as the number of layers increases the gaps move towards the extreme ends of the spectrum. The insensitivity of these gaps as a function of N is due to the fact that $|\sin(Nq_{nm}a/2)/N\sin(q_{nm}a/2)|$ is a slowly varying function of N. Indeed in the limit of large N, this takes a value of $\sin[\pi/(1+\tau)][\pi/(1+\tau)]^{-1}$. Thus the perturbative approach explains most of the features of the spectrum obtained in Figs. 1 and 2.

In summary, we find a strong variation of band gaps in the 2D Brillouin zone of a quasiperiodic superlattice and this should be important in understanding the electronic properties of such superlattices. The variation of gaps has been shown to be due to competition between site, interlayer, and intralayer integrals. An important finding is the occurrence of extended states in the QP direction when there is more than one layer per slab. The same is true even for a QPC when there is more than one atom per cell. This should be important for understanding the transport properties of such quasiperiodic systems. The electronic spectrum of the superlattice shows van Hove singularities which correspond to the underlying periodic lattice. Some of the results presented here are expected to be true for more realistic models and we hope that this paper will stimulate further work on such systems.

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