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Stark ladders in a two-band tight-binding model

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Abstract. The electronic states of a one-dimensional two-band tight-binding model in a uniform electric field, in which the strength of interband coupling is varied, are studied. Eigenenergies, densities of states and wavefunctions are numerically computed for a finite length of the system in order to see the transition from Bloch bands at low electric fields to the eigenstates at high fields explicitly. The results for high electric fields indicate the existence of sets of Stark ladders arising from different bands even when interband coupling is strong.

1. Introduction

The energy spectra of an electron in a crystal under a uniform electric field have been known to be distinctly different from those without an electric field. In a one-dimensional single-band tight-binding model, Katsura *et al* [1] have shown that the wavefunction is represented by a Bessel function and the energy eigenvalue is given by

$$E(n) = nF + \text{constant} \tag{1.1}$$

where n is an integer and $F = eEa$ with $-e$ the electron charge, E the electric field along the chain and a the lattice constant. Subsequently, Kane [2] constructed the wavefunction by the superposition of the Bloch wavefunctions for a general shape of a single band, and obtained exactly the same spectra as in (1.1). The wavefunctions for both models are very similar in that they are localized with a localization length of the order of

$$\lambda = (W/2F)a \tag{1.2}$$

where W is the bandwidth concerned. Unfortunately, however, these pioneering results were not widely appreciated for a long time. Wanner [3–5] was the first to point out that the eigenenergies, in general, have the structure (1.1) because of the periodicity of the crystal potential. Since then the discrete spectra (1.1) have been called Stark ladders. Several authors, however, raised doubt about Wannier's proof. In particular, Zak [6, 7] argued that the interband couplings would completely destroy the discrete character of the spectra and that the spectra might be continuous instead. Saitoh [8] has discussed whether Stark ladders are a physically meaningful concept as resonant levels and has shown that the oscillatory behaviour of the conductivity observed in ZnS crystals by Maekawa [9] can indeed be understood on the the basis of Stark

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ladders. This line of argument was subsequently developed by Sawaki and Nishinaga [10, 11]. Later Avron *et al* [12] proved mathematically that the energy spectra of the Bloch electrons in an electric field are continuous when an infinite number of bands are considered, and that discrete Stark ladders do not exist. This, however, does not necessarily exclude the existence of resonance levels in Stark ladders. Indeed, Avron *et al* [12] have shown that the spectra are point-like, i.e. discrete, when interband coupling among a finite number of bands is considered.

Clearly one of the confusions about the existence of Stark ladders arises from the consideration of an infinite crystal, where the electric field potential is not bounded. For an infinite crystal, a zero electric field could be a singularity of the spectra, and a smooth transition from ordinary Bloch bands to Stark ladders would not be expected. This difficulty can be easily overcome by considering a long but finite length of crystal. The energy spectra of an electron in a finite length of a linear crystal under an electric field were formally solved by Stey and Gusman [13] within the single-band tight-binding model. The eigenenergies and their associated wavefunctions asymptotically approach those obtained by Katsura *et al* as the electric field increases, and they are Bloch-electron-like when the electric field is small. The wavefunction, the energy spectra and the density of states of this system were numerically calculated by Saitoh [14], where a smooth transition from a Bloch character to Stark ladders was observed for which the potential drop across the crystal ϕ was roughly equal to the bandwidth W. Note that the Stark ladders are the exact eigenstates of the system when the electric field becomes infinitely large in this model.

Although it is believed that, when the electric field is large, the general nature of the ladder structure may not be destroyed by the inclusion of interband coupling as long as the overlap between the wavefunctions belonging to different bands is small, it still has to be demonstrated. Fukuyama *et al* [15] investigated briefly the two-band tight-binding model of finite length and concluded that two sets of Stark ladders may exist. In their recent article Leo and Mackinnon [16] numerically investigated a slightly different model from that of Fukuyama *et al* and reported the existence of Stark ladders. With the inclusion of the interband coupling the wavefunction belonging to a particular eigenenergy is a mixture of the original s- and p-band wavefunctions, but the main portion of the wavefunction is either primarily an s- or p-state and the electron is localized in a particular region. The resulting Stark ladders interpenetrate each other. Leo and Mackinnon, however, did not distinguish between the s- and p-components of the wavefunction, and presented the sum of the two envelope functions, which has no clear physical meaning. Moreover, both models are unphysical from our point of view in that the nature of the wavefunctions is not appropriately considered and the signs of the matrix elements are not quite correct; the valence band of both of the earlier models is electron-like at $k = 0$.

The structure of the present article is as follows. First we choose the two-band tight-binding model, which, we believe, reflects the correct character of the wavefunctions. Our model is of the direct-bandgap type; namely the top of the valence band lies at $k = 0$ and the character is hole-like. Second, by studying a finite length of a crystal, the transition of the spectra from being Bloch in character to being Stark ladder type can be fully investigated. Finally, the interband effects will be studied in detail by varying the strengths of interband couplings.

We will present the two-band tight-binding model in section 2, and present the numerical calculations of the wavefunctions, eigenenergies and the density of the states in section 3 together with a discussion on the existence of Stark ladders by changing

the interband coupling strength. Section 4 is devoted to conclusions.

2. Model

We use a one-dimensional tight-binding model, which consists of s and p orbits. The Hamiltonian is given by

$$\begin{aligned}\mathcal{H} = &\sum_{n=1}^{N}(\epsilon_s + nF)S_n^+ S_n - V_s\sum_{n=1}^{N}\{S_{n+1}^+ S_n + S_n^+ S_{n+1}\} \\ &+ \sum_{n=1}^{N}(\epsilon_p + nF)P_n^+ P_n + V_p\sum_{n=1}^{N}\{P_{n+1}^+ P_n + P_n^+ P_{n+1}\} \\ &+ V_{sp}\sum_{n=1}^{N}\{S_n^+ P_{n+1} - S_{n+1}^+ P_n + P_{n+1}^+ S_n - P_n^+ S_{n+1}\} \\ &+ \eta F\sum_{n=1}^{N}\{S_n^+ P_n + P_n^+ S_n\}\end{aligned} \tag{2.1}$$

where S_n and P_n are the annihilation operators of the s and p orbits at the nth site, ϵ_s and ϵ_p are the on-site energies of the s and p orbits, V_s and V_p the transfer integrals of the corresponding orbits, V_{sp} the transfer integral between the s and p orbits, F the electric potential drop across the unit cell, and η is the matrix element of x/a which is very small compared with unity. The fifth term results in the interband coupling. In setting up the model, terms which are smaller than two-site overlap integrals are discarded. Note that the signs of the coupling parameters are different from the models of Fukuyama *et al* and Leo and Mackinnon. In the following we use the unit system in which $\hbar, e$ and a are chosen to be unity, and F will simply be called the electric field. Since the last term is small compared with the first two terms and its major effect is only to widen the gap between the conduction and valence bands, it will be discarded hereafter. The following periodic boundary conditions will be employed here

$$S_{N+1} = S_1 \qquad P_{N+1} = P_1. \tag{2.2}$$

When the electric field is absent ($F = 0$), this Hamiltonian is easily diagonalized and the eigenenergies are explicitly written as

$$E_\pm(k) = \tfrac{1}{2}\left[\epsilon_{sk} + \epsilon_{pk} \pm \sqrt{(\epsilon_{sk} - \epsilon_{pk})^2 + 4\Delta_k^2}\right] \tag{2.3}$$

where

$$\begin{aligned}\epsilon_{sk} &= \epsilon_s - 2V_s\cos k \\ \epsilon_{pk} &= \epsilon_p - 2V_p\cos k \\ \Delta_k &= 2V_{sp}\sin k.\end{aligned} \tag{2.4}$$

and k is the wavenumber of an electron

$$k = (2\pi/N)n \qquad (n = 0, 1, \ldots, N-1). \tag{2.5}$$

When the interband coupling Γ_k is small, the model has the direct gap $E_g = |\epsilon_s - \epsilon_p|$ at the Γ point. Examples of the bands for the bandwidths $W_s = 16$, $W_p = 4$ and several different interband coupling parameters V_{sp} are given in figure 1. The present model is different from those of Fukuyama *et al* (FBF) [13] and Leo and Mackinnon (LM) [14]. The FBF and the LM models give the following dispersion relations

$$E\pm(k) = \tfrac{1}{2}\left[\epsilon_{sk} + \epsilon_{pk} \pm \sqrt{(\epsilon_{sk} - \epsilon_{pk})^2 + 4V_{sp}^2}\right] \qquad \text{(FBF model)} \quad (2.6)$$

$$E\pm(k) = \tfrac{1}{2}\left[\epsilon_{sk} + \epsilon_{pk} \pm \sqrt{(\epsilon_{sk} - \epsilon_{pk})^2 + 4\Delta_k^2}\right] \qquad \text{(LM model)} \quad (2.7)$$

where

$$\begin{aligned} \epsilon_{sk} &= \epsilon_s - 2V_s \cos k \\ \epsilon_{pk} &= \epsilon_p - 2V_p \cos k \\ \Delta_k &= 2V_{sp} \sin k. \end{aligned} \qquad (2.8)$$

Note that the sign of V_p in both the FBF and LM models is different from ours and so the form of the bands is very different from those encountered in semiconductors in which the valence band is hole-like at the Γ point. In our model important mixing occured between the Γ points of the conduction and valence bands. In the previous two models it occured between the conduction Γ point and the valence X point.

To illustrate the numerical results, we fix the band parameters to the following values: $\epsilon_s = 0$, $\epsilon_p = -30$, $V_s = 4$ and $V_p = 1$. These values are chosen somewhat arbitrarily to represent the wide-gap semiconductors. Let us first consider the zero-field case. The conduction band (the s band) and the valence band (the p band) have bandwidths 4 and 16 and a bandgap of 20 for $V_{sp} = 0$. The interband couplings V_{sp} are taken to be 0, 6 and 8.5 which corresponds to the weak, intermediate and strong coupling cases, respectively (figure 1). In the cases of $V_{sp} = 6$ and 8.5, the bands are considerably modified from the weak coupling cases and the new van Hove singularities will appear.

In the general case of non-zero electric fields, it is necessary to diagonalize the Hamiltonian (2.1) numerically in order to obtain eigenenergies and eigenfunctions. This will be performed in the next section.

3. Results

In this section numerical calculations of the density of states (DOS) and the eigenfunctions for non-zero electric fields are given. The DOS is defined by

$$\text{DOS}[E(n)] = \frac{1}{N}\frac{2}{E(n+1) - E(n-1)}g \qquad (3.1)$$

where g represents the g-fold degeneracy of the nth energy state, N the total number of lattice points, and $E(n)$ the nth eigenenergy. In the following, the chain length is taken to be $N = 100$. A large N ensures that the results are represented essentially by bulk properties. The energy origin is taken to be $E_0 = \frac{1}{2}(N+1)F$. We define the potential drop across the system by

$$\phi = NF. \qquad (3.2)$$

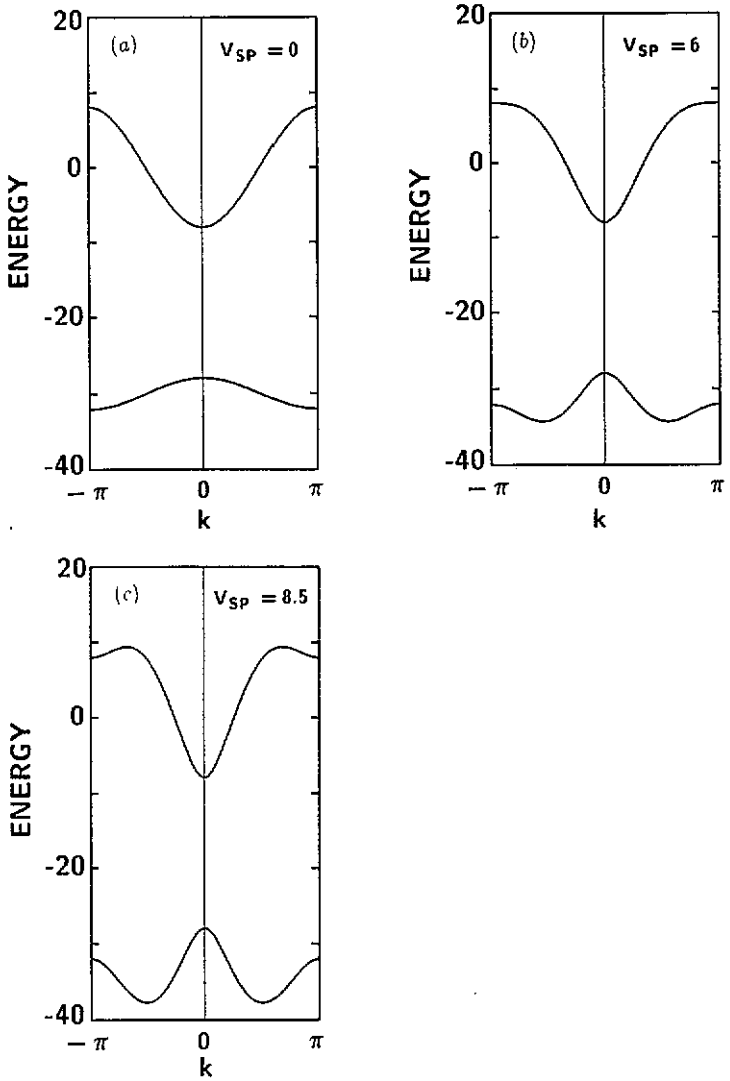


Figure 1. Energy bands for different interband couplings when the electric field is absent. Band parameters are chosen as $V_s = 4, V_p = 1$, and $V_{sp} = 0, 6$ and 8.5 for (a), (b) and (c) respectively, which correspond to the weak, intermediate and strong coupling cases, respectively.

In figures 2(a), (b) and (c), the n dependence of the eigenenergies for $V_{sp} = 0$ are plotted for weak, intermediate and strong electric fields. If the relation between $E(n)$ and n is linear, it means Stark ladders. It is seen that the linear region increases with the electric field, and this indicates that the region of Stark ladders depends on the strength of the electric field for a finite length of the crystal. This is in contrast to the case of an infinitely long crystal in which all the states belong to Stark ladders within the tight-binding model. Note that the slope of the line in the central part of figures 2(b) and (c) is half of the slope at both ends of the line. This will be discussed later.

Figure 3 shows the DOS for a weak field: $F = 0.16$ ($\phi = 16$), where ϕ is equal to the width of the s band for $F = 0$. For $V_{sp} = 0$ (figure 3(a)), the DOS displays the

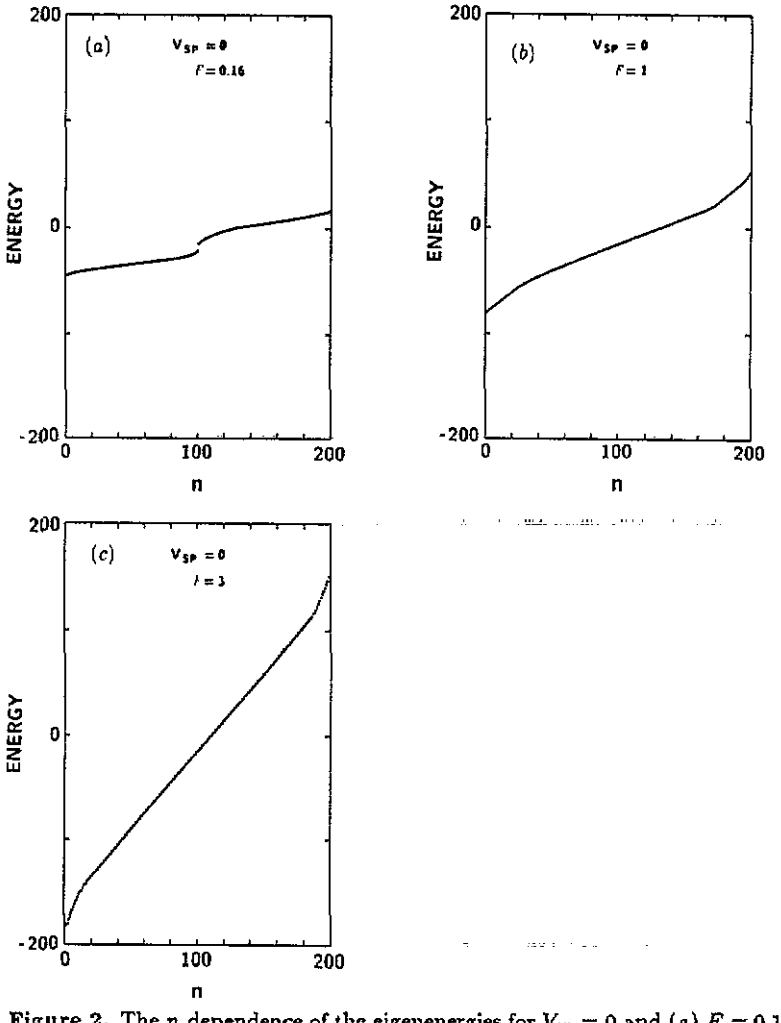


Figure 2. The n dependence of the eigenenergies for $V_{sp} = 0$ and (a) $F = 0.16$, (b) $F = 1$ and (c) $F = 3$.

two detached bands. The characteristic of each band can be understood in terms of the picture of two independent bands in weak fields. The DOS for the lower band is flat in the central region indicating the existence of a Stark ladder. But, for the upper band, there is no flat region, and therefore the Stark ladder is absent. The reason is that the characteristic localization lengths (= (bandwidth)/$2F$) differ in each band because of the difference in bandwidths. Twice the localization length (i.e. the extent of the wavefunction) for the s band is equal to the length of the crystal and the s component of the wavefunction is extended over the crystal, but for the p component twice the localization length is only a quarter of the crystal length and it is localized within the crystal. The band gap becomes narrower than the zero-field case because each if the band is expanded by the electric field. According to the results from the single-band model of Saitoh [14], the electronic states near these band edges have an Airy-like spectrum, i.e. the spectrum is characterized by those of the triangle well potential. The narrowing of the band gap is related to the Franz–Keldysh effect [17,

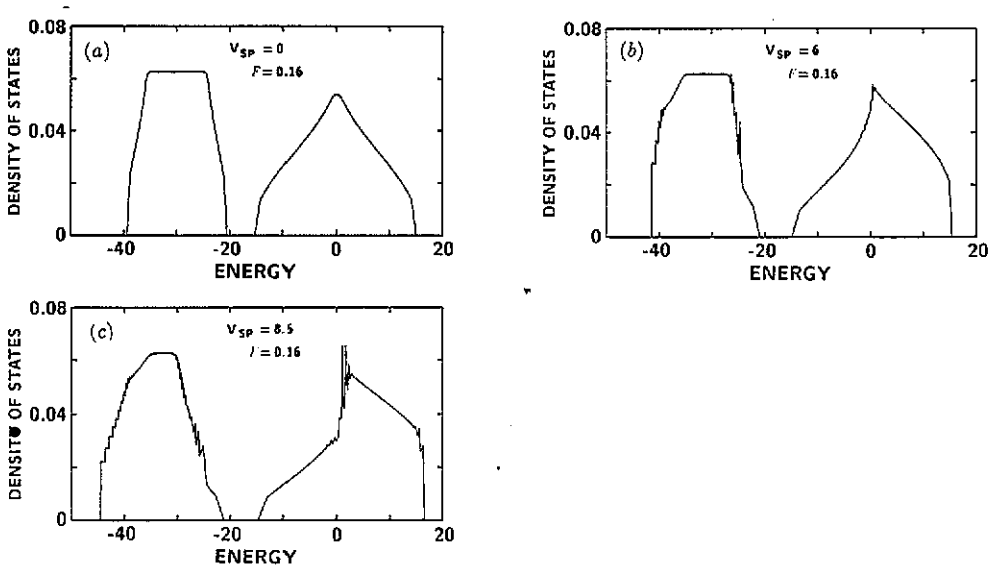


Figure 3. The DOS for various interband couplings: (*a*) $V_{sp} = 0$, (*b*) $V_{sp} = 6$ and (*c*) $V_{sp} = 8.5$ in the weak electric field, where $F = 0.16$ ($\phi = 16$).

18]. Figures 3(*b*) and (*c*) show the case of intermediate and strong interband couplings. In these cases the DOS becomes asymmetric with respect to the band centre because the interband coupling is strong. The flat region in the lower band which indicates the existence of a Stark ladder persists as in figure 3(*a*), even though the interband coupling becomes stronger.

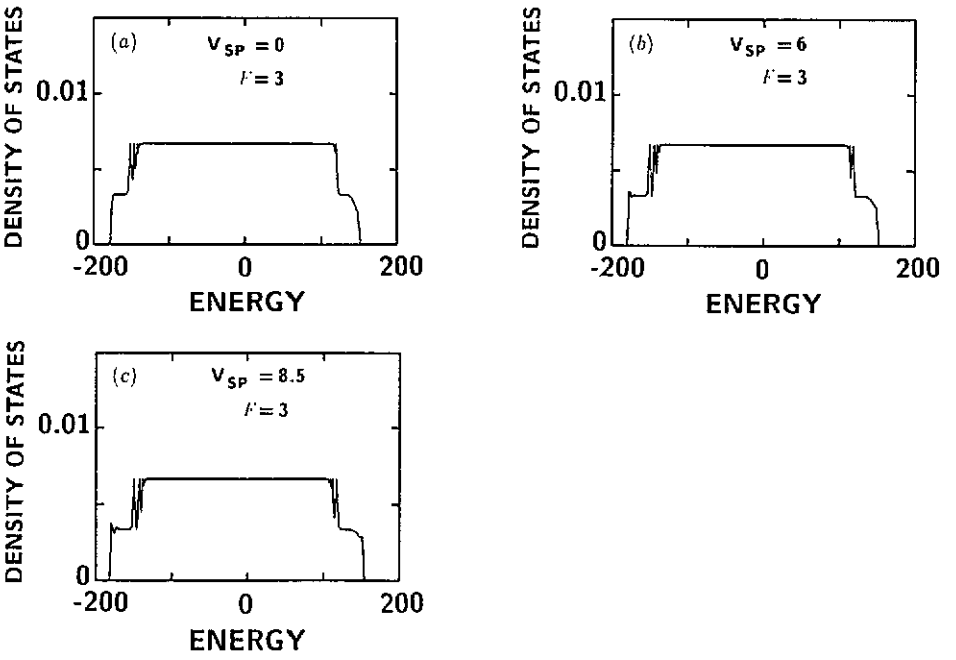


Figure 4. The DOS with (*a*) $V_{sp} = 0$, (*b*) $V_{sp} = 6$ and (*c*) $V_{sp} = 8.5$ for various interband couplings in the high electric field, where $F = 1$ ($\phi = 100$).

Next, the results for a high field, $F = 3$, for which the localization lengths of the wavefunctions for both bands become smaller than the chain length N are shown in figure 4. It should be noted here that two bands merge and the band gap disappears. It is also seen that the shape of the DOS is independent of coupling strength. The DOS has three plateau regions, high in the centre and low at both sides. In the low plateau regions at both sides, the DOS has the value ϕ^{-1} as seen in figure 4. This is understood as the energy levels having equal spacing F as shown in figure 5(a), since

$$\text{DOS} \simeq \frac{2}{N}\frac{1}{2F} = \frac{1}{\phi}. \tag{3.3}$$

In the central flat region, the energy levels do not, in fact, have equal spacing, but a pair of energy levels are equally spaced as shown in figure 5(b). The spacing of every other level is approximately equal to F. The DOS in the figure appears to be flat because of definition (3.1) which averages large (small spacing) and small (large spacing) DOS:

$$\text{DOS} \simeq 2/\phi. \tag{3.4}$$

The difference in the DOS values are reflected in the slopes in figures 2(b) and (c), as mentioned earlier.

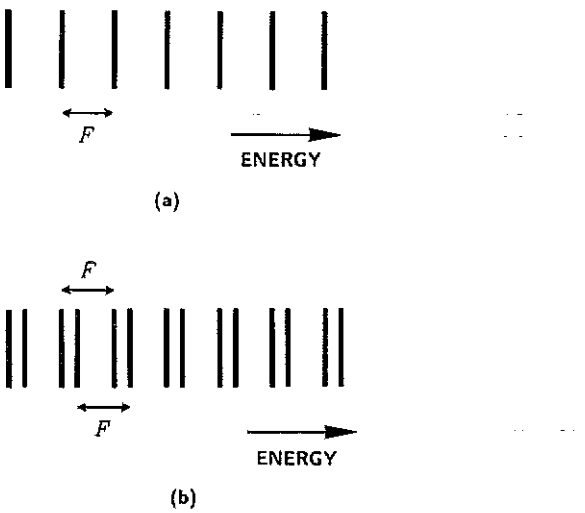
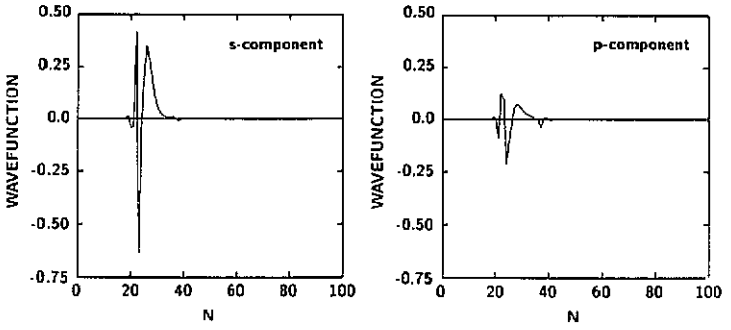
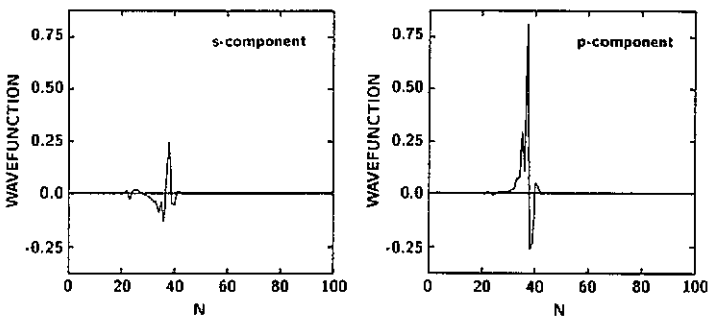


Figure 5. Schematic energy levels in the typical regions of the DOS for figure 4, where (a) is the band edge region and (b) the band centre region.

To see the nature of the wavefunctions let us examine a pair of two neighbouring levels which are shown in figure 5(b). Figure 6 shows an example of the wavefunctions belonging to the 60th and 61st levels in the central region of figure 4(c) with $V_{sp} = 8.5$ and $F = 3$ ($\phi = 300$). The wavefunction corresponding to the 60th level is mainly s in character and is localized around $n \approx 20$ and the wavefunction belonging to the 61st level has mainly the p character located around $n \approx 30$ although the interband



(a) n = 60



(b) n = 61

Figure 6. Typical wavefunctions of adjacent energy levels in the central flat region of figure 4(*c*) where $V_{sp} = 8.5$ and $F = 3(\phi = 300)$: (*a*) the wavefunction belonging to the 60th eigenstate with energy $E(60) = -75.354$; and (*b*) the wavefunction belonging to the 61st eigenstate with energy $E(61) = -74.646$.

coupling mixes the s and p components. The location of the gravity centre of each component is given approximately by

$$\left(n - \frac{N+1}{2}\right) F = E(n) - \epsilon_l \qquad (l = \mathrm{s}, \mathrm{p}). \tag{3.5}$$

Namely, the pair of adjacent energy levels have the character mainly of the s and of the p types, respectively. The next nearest energy distances of s-to-s and p-to-p are given approximately by F. This means the energy levels consist of two interpenetrating Stark ladders as pointed out by Fukuyama *et al* and Leo and Mackinnon. The DOS is given by the sum of these interpenetrating Stark ladders and the DOS in the central region is twice as large as those in the shoulder regions.

It can be concluded that, when the electric field is applied, the dominant component in the Stark ladders is produced more or less from one particular band, and the two types of Stark ladder interpenetrate each other. This behaviour is qualitatively unchanged even if the interband coupling V_{sp} is increased by an unrealistically large amount.

4. Conclusion

We have numerically investigated the electronic states of a linear crystal in an electric field within the two-band tight-binding model by changing the interband coupling. It is concluded that two interpenetrating Stark ladders are observed for strong electric fields irrespective of the strength of the interband couplings. These results are not trivial because of the existence of the interband coupling. The effect of the interband coupling is to mix the s and p components in the resulting wavefunction but this does not destroy the Stark ladders.

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