Noncanonical Wannier-Stark ladders and surface state quantization in finite crystals subjected to a homogeneous electric field

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(Received 10 November 2000; revised manuscript received 26 March 2001; published 29 May 2001)

In the one-particle single band approximation, which is the basis of the original Wannier result, commonly referred to as the Wannier-Stark ladder (WSL), we have extended the concept by predicting the existence of noncanonical WSLs which are a set of evenly spaced levels (in the middle of the tilted band) with *noncanonical level spacing* equal to the Plank constant times $(1-2m'/m)^{-1}$ times Bloch oscillation frequency. To observe a particular WSL, the certain voltage must be applied. The latter is related to the numbers $m=3,4,\ldots$ and $m'=1,2,\ldots<m/2$. We also show that, if the electrostatic energy due to applied voltage is larger than the zero-field band width, the quantization of surface localized states smoothly changes from the Airy type (at the spectrum edges) to the Wannier-Stark type with a pronounced energy interval in between, where the level spacing doubles that of canonical WSL. Analytical results are derived within the exactly solvable model of finite tilted tight-binding band. Their experimental implications and further-to-go directions are addressed to dielectric crystalline layers and superlattices, whose thickness (length) admits the direct tunneling.

DOI: 10.1103/PhysRevB.63.235410 PACS number(s): 73.21.−b, 73.61.−r, 73.90.+f

I. INTRODUCTION

Since a notion of the Stark ladder was proposed by Wannier in 1960 (Ref. 1) and after many years of debates and controversy, the quantization of electronic band states in a constant electric field [the Wannier-Stark ladder (WSL)] should have developed into one of basic concepts of solid state. During last decade an impressive progress has been achieved in the understanding of Wannier-Stark (WS) quantization and related phenomena such as Zener tunneling[2] and Franz-Keldysh effect,[3] especially, in superlattices,[4–7] see also reviews in Refs. 8–10, and driven optical lattices,[11] see also references therein.

Along with models approaching various aspects of real systems (multiband structure, electron-electron and electron-phonon interactions, edge effects, etc.) the one- and two-band tight-binding approximation has been widely used in discussions of WSLs.[12–16] Even the one band model reflects salient essentials of real systems, admits obtaining analytical results and may serve, therefore, as a starting point for more accurate theories. It is also often used for checking and interpreting numerical results which are dominant in the field because of the problem complexity.

One band tight-binding model of an *infinite* crystal subjected to a constant electric field was solved exactly by Hacker and Obermair[9] to prove that the eigenvalues do form WSL while the corresponding eigenfunctions are localized within the tilted band. The electron state localization is thus shifting in space along the field. Using the same model but for a *finite* crystal Saitoh[10] pointed out that for a WSL to appear, the potential energy of the applied voltage must exceed some critical value shown likely to be (but not proved) the band width. The coexistence of the Airy-type and Wannier-type spectra in finite crystals was also discussed but neither of these has been derived consistently from the basic Schrödinger equation. For open boundaries the existence of a WSL was explicitly demonstrated by Stay and Gusman[14] on the basis of the exact formal solution to the Schrödinger problem. However, they left out of focus the surface state spectrum and, as is shown below, their estimate of the bulk states energies (that is WSLs in finite crystals) needs to be corrected.

Fukuyama *et al.*[15] and in a more recent work Yakovenko and Goan[17] have obtained both the Airy and Wannier-Stark spectra from one and the same equation for a semi-infinite tight-binding chain. For this reason (see details in Sec. VI) their results are limited only to the case, where the total electrostatic energy is larger than the electron band width. The Airy-type quantization has been obtained for the very edge of the spectrum. Thus it is not representative for the most of edge states. As to the bulk states spectrum, both works just repeated the earlier result obtained for infinite systems.

The current presentation removes the above mentioned weaknesses. It offers a more detailed and, in certain aspects, more accurate description of bulk and surface states, which is based on the mentioned above exact *formal* solution of the spectral problem for a finite tight-binding chain. Under the conditions not limiting its applicability much, that solution is advanced further to an explicit form of eigenvalues for a number of cases of interest. The obtained conclusions are valid for practically any crystal thickness and applied voltage attainable in real experiments.

Recently, some of our findings have been briefly reported.[18] The aim of this article is to give a complete classification of electronic states subjected to a constant electric field in a one-particle single band approximation with an emphasis on explicit analytical results and their experimental implications. We predict the existence of WSLs with noncanonical level spacing controlled by the applied voltage and new types of quantization for surface states, namely, with Wannier and doubled Wannier level spacing.

The article is organized as follows. An exact secular equation for the problem in focus is introduced in Sec. II. The analysis of bulk states spectrum at low and high voltages is performed in Secs. III and IV, respectively. Section V classifies surface localized states and gives explicit expressions for the eigenvalues in different energy intervals. The results obtained for a finite crystal thickness at high voltages are rederived for a semi-infinite crystal in Sec. VI, which also links our results with previous studies. Section VII summarizes the main findings of the work, outlines the conditions for their experimental verification, and points to directions for their further usage.

II. PROBLEM STATEMENT AND BASIC EQUATIONS

The gross structure of a single band spectrum in presence of a constant electric field can readily be understood in terms of a finite tilted band represented in Fig. 1. The middle and lower diagrams distinguish, respectively, the cases of *low* [the electrostatic energy difference eV is smaller than the unperturbed band width $E^0_{\rm bw}$ $(eV<E^0_{\rm bw})$] and *high* $(eV>E^0_{\rm bw})$ applied voltages; while the upper exhibits the unperturbed band $(eV=0)$.

We consider a symmetric tight-binding band under the influence of a constant electric field. Since the spectrum of the system is symmetric with respect to its center (denoted in Fig. 1 as E_c) it is convenient to use E_c as the energy reference point. Equivalently, one can assume that the electrostatic potential energy varies symmetrically with respect to the crystal center and use the zero energy as the reference point. With the latter choice, the Hamiltonian matrix has the form

$$H_{mn}=-eFa\left(\frac{\mathcal{N}+1}{2}-n\right)\delta_{m,n}+\beta\delta_{|m-n|,1}, \qquad (1)$$

where $n,m=\overline{1,\mathcal{N}}$, $\mathcal{N}a$ is the crystal thickness (see Fig. 1), and e, F, and a are, respectively, the absolute value of electron charge, electric field strength, and lattice constant; β is the energy of electron resonance transfer between neighboring sites; the electron site energy in zero field is set equal to zero. In what follows, β serves as the energy unit, notation $\mathcal{E}=eFa/\beta$ stands for the field parameter, $E^0_{\rm bw}=4$, and $eV=\mathcal{E}(\mathcal{N}-1)$ is the energy of electrostatic potential applied to the bounding surfaces which are supposed to be perpendicular to one of the crystal principal axes.

It has been shown by many authors[13–16,19] that finding the eigenvalues of matrix H_{mn}, which are given by zeros of the determinant

$$\mathcal{D}_{\mathcal{N}}(E)=|E\delta_{m,n}-\beta^{-1}H_{mn}|=0, \qquad (2)$$

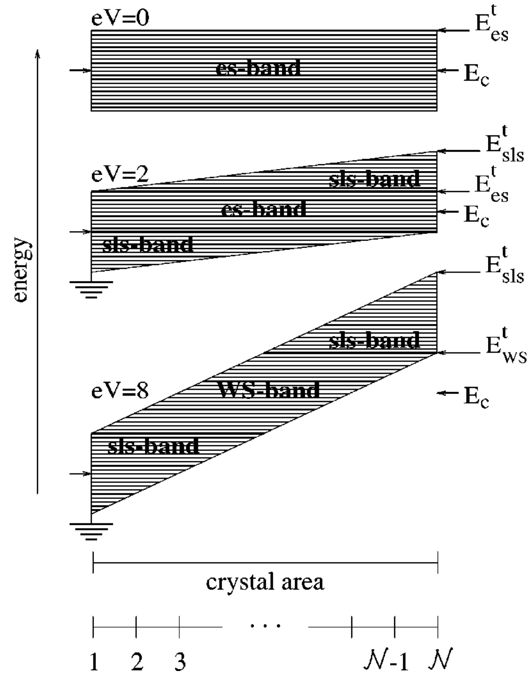

FIG. 1. Untilted (upper) and tilted (middle and lower) bands of electronic levels in a crystal of finite thickness. The crystal area in the direction of electric field is occupied by $\mathcal{N}$ monoatomic layers. In the division of the band spectrum at a particular voltage V, the center of the spectrum E_c serves as a reference point. At $V=0$ all states within the band are extended from the left-hand side of the crystal to the right-hand side. These states form a band of *extended states* or es band which is indicated by shaded rectangles in the upper and middle diagrams. The zero-field band width $E^0_{\rm bw}=4$ in units of the electron resonance transfer energy. At $eV_{\rm min}<eV<4$, see Eq. (26), in addition to the (narrowed) es band, two bands of surface localized states emerge (sls bands indicated by shaded triangles). At $eV>4$ the es band shrinks to zero and the Wannier-Stark band appears instead (WS band is indicated by a shaded parallelogram). Index t (top) labels the highest possible energy within the es $(E^t_{es}=2-eV/2)$, WS- $(E^t_{\rm WS}=eV/2-2)$, and sls band $(E^t_{sls}=2+eV/2)$.

can be reduced to the solution of a certain transcendent equation. For the matrix defined in Eq. (1), the functional form of the determinant is represented by the equation[19]

$$D_{\mathcal{N}}(E)=J_{\mu+(\mathcal{N}+1)/2}(z)Y_{\mu-(\mathcal{N}+1)/2}(z)-Y_{\mu+(\mathcal{N}+1)/2}(z)J_{\mu-(\mathcal{N}+1)/2}(z), \qquad (3)$$

where $\mu\equiv E/\mathcal{E}$, $z\equiv 2/\mathcal{E}$, and $J_\mu(z)$ and $Y_\mu(z)$ are the Bessel functions of the first and second kind, respectively. For other forms of Eqs. (2) and (3) see, e.g., Refs. 13–16.

Our main purpose is to give (with a minimal loss of accuracy) a more clear form of Eq. (3) which admits obtaining analytical solutions to Eq. (2). This is done under the condition $\mathcal{E}\ll 1$ and $\mathcal{N}\gg 1$ covering most cases of interest. All underlying derivations are based on the standard approximations of Bessel functions with large arguments and small or large orders. The particular approximation to be used depends on the energy interval. Basically our approach is similar to that used in Ref. 15. However, as it will be seen very soon, an analytical description of electric field effect on the electron spectrum goes far beyond the previous results.

For the low and high voltages, as is specified above, the resulting equations for the bulk electronic states have different properties. Also the bulk (extended or Wannier-Stark) and surface states are distinct. Therefore, the cases of bulk states at low voltages, at high voltages, and surface states are considered separately. In the two next sections, we concentrate at the bulk states spectrum.

III. BULK STATES, LOW VOLTAGES SPECTRUM

If $eV<4$ and $0<E<2-eV/2$, any part of the crystal is classically accessible for electrons. But it is not so, if $2-eV/2<E<2+eV/2$, in which case the upper sloped line in Fig. 1 (mid diagram) divides classically accessible (shaded) and forbidden (unshaded) areas. In the former energy interval, the electron states are extended over the entire length $\mathcal{N}$, the crystal thickness and, in the latter, they have a tendency to be preferably localized near the right hand side of the crystal. Correspondingly, the lower-energy interval is appropriate to refer as an *extended states* band or es band, whereas the upper interval will be called *surface localized states* band or sls band. Because of the spectrum symmetry, only positive energies and positive sign of the field parameter will be discussed, i.e., only the upper half of the es band (or Wannier-Stark band at high voltages) and the upper sls band.

By making use of well-known properties of Bessel functions,[20] which appear in Eq. (3), and restricting ourselves by energies not too close to the top of the es band $0<E<2-eV/2-\mathcal{E}$ (the excluded $\mathcal{E}$ interval contains one or no levels at all) the expression of determinant (3) can be transformed into the following:

$$\mathcal{D}_{\mathcal{N}}(E)\approx\frac{\mathcal{E}}{\pi}\sqrt{\frac{1}{\sin\xi\sin\xi'}}\sin\left\{\xi+\xi'-\frac{2}{\mathcal{E}}(\Phi_{\xi}-\Phi_{\xi'})\right\}, \tag{4}$$

where

$$2\cos\xi=E+\frac{eV}{2},\quad \Phi_{\xi}=\sin\xi-\xi\cos\xi \tag{5a}$$

and

$$2\cos\xi'=E-\frac{eV}{2},\quad \Phi_{\xi'}=\sin\xi'-\xi'\cos\xi'. \tag{5b}$$

Represented in Fig. 2 energy dependence of determinant (4) shows that significant distinctions from the exact dependence $\mathcal{D}_{\mathcal{N}}(E)$ are observed only in the excluded $\mathcal{E}$ interval near the top of es band.

From Eqs. (2) and (4) it is obvious that within the es band the eigenvalues are given by solutions to equation

$$\sin\{(\mathcal{N}+1)\xi^{es}\}=0, \tag{6}$$

where

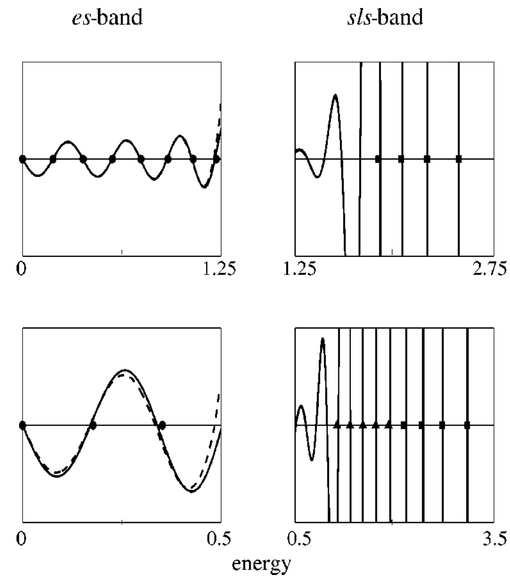

FIG. 2. Exact dependencies $\mathcal{D}_{\mathcal{N}}(E)$ (solid lines) and their approximations (dashed lines) calculated from Eq. (4) for the es band, and from equation $\sin\xi^{sls}=0$ for the sls band. In calculations $\mathcal{N}=31$, $\mathcal{E}=0.05$ ($eV=1.5$, upper curves), and $\mathcal{E}=0.1$ ($eV=3$, lower curves). The level energies are determined by intersections with the horizontal axis. The number of states in the sls band ($n^{sls}=8$ for $\mathcal{E}=0.05$ and $n^{sls}=13$ for $\mathcal{E}=0.1$) coincides with that calculated from Eq. (24a). Approximate values of energy levels (27) and (28) are marked by filled squares and triangles, respectively. Filled circles correspond to approximation $E_n=2\cos\xi_n^{es}$, where ξ_n^{es} is given by Eq. (9).

$$\xi^{es}=\frac{1}{\mathcal{N}+1}\left\{\xi+\xi'-\frac{2}{\mathcal{E}}(\Phi_{\xi}-\Phi_{\xi'})\right\}. \tag{7a}$$

Equation (6) is formally identical to that determines the quantized values of the quasi-impulse $\xi^{(0)}$ in an unperturbed (untilted) tight-binding band, $\sin\{(\mathcal{N}+1)\xi^{(0)}\}=0$. Thus, in presence of a constant electric field the es band states may be classified by a field-dependent *pseudo*-quasi-impulse ξ^{es} which is quantized exactly in the same way, as the quasi-impulse in zero field. It lists only those electronic states, which are within the es band

$$\xi_n^{es}=\frac{\pi n}{\mathcal{N}+1},$$

$$n=\left[\frac{1}{2}(\mathcal{N}+1)\right],\left[\frac{1}{2}(\mathcal{N}+1)\right]-1,\ldots,\left[\frac{1}{2}(\mathcal{N}+1)\right]-n^{es}+1. \tag{7b}$$

Here (and in subsequent expressions) square brackets indicate the integer part of the argument. The maximal possible value of ξ^{es} is equal or close to $\pi/2$. The minimal possible value of ξ^{es} is controlled by the applied voltage. It determines the number of states in a half of es band $n^{es}=[(\mathcal{N}+1)/2]+1-(\mathcal{N}+1)\xi^{es}(E=2-eV/2)/\pi$ (see a more convenient definition of n^{es} in Sec. V).

A. Extended states spectrum, perturbative treatment

From the definitions given above it follows that in general, the pseudo-quasi-impulse ξ^{es} can be represented either by a function of a single energy variable E or by a function of two quasi-impulses ξ and ξ' defined in Eqs. (5a) and (5b). In zero field, both quantities ξ and ξ' coincide with the quasi-impulse $\xi^{(0)}$ which is related to the untilted-band energy by the dispersion relation $E^{(0)}=2\cos\xi^{(0)}$. It is clear that at sufficiently low voltages ξ^{es} must be related to $\xi^{(0)}$. To find the relation, let us pass from ξ and ξ' in Eqs. (5a) and (5b) to a single quasi-impulse $\overline{\xi}$ such that $E=2\cos\overline{\xi}$. Then, expanding $\overline{\xi}$ and $\Phi_{\overline{\xi}}$ in powers of eV up to $(eV)^3$ and substituting the result in Eq. (7), one can obtain in the lowest approximation

$$\xi^{es}\approx\overline{\xi}-\frac{(eV)^2}{96}\frac{\cos\overline{\xi}}{\sin^3\overline{\xi}}\frac{\mathcal{N}+5}{\mathcal{N}+1}. \tag{8}$$

Now using Eq. (8) in Eq. (6) and solving the latter in linear approximation with regard to $(eV)^2$, we get

$$\xi_n^{es}\approx\xi_n^{(0)}+(eV)^2\frac{\mathcal{N}+5}{96(\mathcal{N}+1)}\frac{\cos\xi_n^{(0)}}{\sin^3\xi_n^{(0)}}, \tag{9}$$

where $\xi_n^{(0)}=\pi n/(\mathcal{N}+1)$ stands for the solution to Eq. (6) in zero field. In Fig. 2, the values of $E_n=2\cos\xi_n^{es}$ are marked by filled circles.

It can be seen from the above equation that the field effect on es-band energies is different for the inner (E is close to the es-band center, where $E=0$) and outer (E is close to the es-band edge $E=E_{es}^t=2-eV/2$) levels. If, for example, $\mathcal{N}$ is an odd number $\mathcal{N}=2N+1$, the inner levels are $\xi_{n'}^{(0)}=\pi/2-\pi n'/[2(N+1)]$, where $n'=1,\ 2,\ \dots,$ and is restricted by the condition $n'/(N+1)\ll 1$. In this case, we have from Eq. (9)

$$\xi_{n'}^{es}=\frac{\pi}{2}\left\{1-\frac{n'}{N+1}\left(1-\frac{(eV)^2}{96}\frac{\mathcal{N}+5}{\mathcal{N}+1}\right)\right\}, \tag{10}$$

showing that the shift of inner levels by the field is *towards* the band center and *does not depend* on the level number. Thus the *decrease* of level spacing in the middle of the es band is mostly controlled by the crystal thickness. For $\mathcal{N}\gg 1$, the correction to the zero-field value of $\xi_{n'}^{es}$ is smaller than, or of the order of 1% up to $eV=1$. Interestingly (and not at all self-obviously), the applied voltage makes the mid-band states more densely packed, though as a whole, es band plus sls band, the spectrum widens.

In contrast to inner levels, the field effect on the outer levels of es band does depend on the level number. Again the general tendency is a *decrease* of the level spacing by the field. The condition of the applicability of Eq. (9) for the outer levels ($\xi_n^{(0)}\ll 1$) requires much lower voltages, since, as is seen from that equation, the second term in its right hand side can be regarded as small only if $\mathcal{E}\ll 4\sqrt{6}\pi^2 n^2/\mathcal{N}^3$. The rigorous estimate of the accuracy of Eq. (9), which is based on its derivation, gives even a more demanding condition $\mathcal{E}\ll 3.2\pi^2 n^2/\mathcal{N}^3$. Hence, strictly speaking, if the electrostatic potential energy is of the order or larger than $30/\mathcal{N}^2$, to describe accurately the outer es-band levels, one has to use Eq. (7). However, it appears that even when eV is of the order of $30/\mathcal{N}^2$, approximation (9) works reasonably well, see Fig. 2.

B. Nonperturbative treatment, noncanonical WSLs

In this section, the smallness of the field parameter is not used. Under the only condition $\mathcal{N}\gg 1$, some explicit solution to Eq. (2) will be found for $eV<4$. As it has been reported recently,[18] at the particular values of energy $E_n=n\mathcal{E}$ with n ($\ll N$) an integer, determinant (3) is accurately approximated by the expression

$$\mathcal{D}_{2N+1}(E_n)\approx(-1)^{N+n+1}\frac{\mathcal{E}}{\pi}\frac{\sin\{2n\arccos(\mathcal{E}N/2)\}}{\sin\{\arccos(\mathcal{E}N/2)\}}, \tag{11}$$

where $\mathcal{N}$ is set to be odd. The case of even $\mathcal{N}=2N$ is treated similarly.

It follows from Eq. (11) that if $eV<4$ (i.e., $\mathcal{E}N/2<1$), some (or none) of electronic states have the energies which are exactly equal to $n\mathcal{E}$. Moreover, for the voltages $2n\arccos(\mathcal{E}N/2)=\pi k,\quad k=0,1,2,\dots,$ some of electronic states do have the energy $n\mathcal{E}$. The voltage (electric field strength) required for $\mathcal{D}_{2N+1}(E_n)=0$ is thus equal to

$$eV_{m'/m}=(\mathcal{N}-1)\mathcal{E}_{m'/m}=4\cos\left(\pi\frac{m'}{m}\right), \tag{12}$$

where $m=3,4,\dots;\ m'=1,2,\dots<m/2$; and m and m' are relatively prime numbers, i.e., they do not have any common divisor.

An analysis of the above condition leads to a conclusion that there exist sequences of eigenvalues (specific series) $E_j=j\mathcal{E}$, where $j=0,m,2m,\dots,$ if m is odd, and $j=0,m/2,m,\dots,$ if m is even. It may be convenient to distinguish these as odd $E_j^{o\{(m-1)/2,m'\}}$ (m is odd) and even $E_j^{e\{(m-2)/2,m'\}}$ (m is even) series. As is seen from Eq. (12), the odd series $E_j^{o(1,1)}$ appears at $eV_{1/3}=2$ ($\mathcal{E}_{1/3}=1/N$) with the level spacing equal to $3\mathcal{E}_{1/3}=3/N$. This odd series lists all levels in the middle part of the es band. The same is true about the even series $E_j^{e(1,1)}=2j\mathcal{E}_{1/4}$, which appears at $eV_{1/4}=2\sqrt{2}>eV_{1/3}$. In general, the odd series $E_j^{o\{(m-1)/2,m'\}}$ will coincide with each $(m-2m')$th spectrum level, whereas the energies $E_j^{e\{(m-2)/2,m'\}}$ will coincide with each $(m/2-m')$th spectrum level. For instance, the odd series $E_j^{o(2,1)}$, $E_j^{o(3,1)}$, and so on (which appear at ever higher voltage) will coincide with each third, fifth, and so on spectrum level. Some of odd and even series are compared with exact level energies in Fig. 3.

Next observation makes the spectrum regularities just described especially simple. The comparison with the exact spectrum shows that between levels E_j and $E_{j\pm 1}$, an approximation of equidistant levels is appropriate, unless the level energies are not too far from the spectrum center. For instance, when series $E_j^{o(2,1)}$ and $E_j^{o(2,2)}$ appear, the level spacing in the band middle is (approximately) equal to

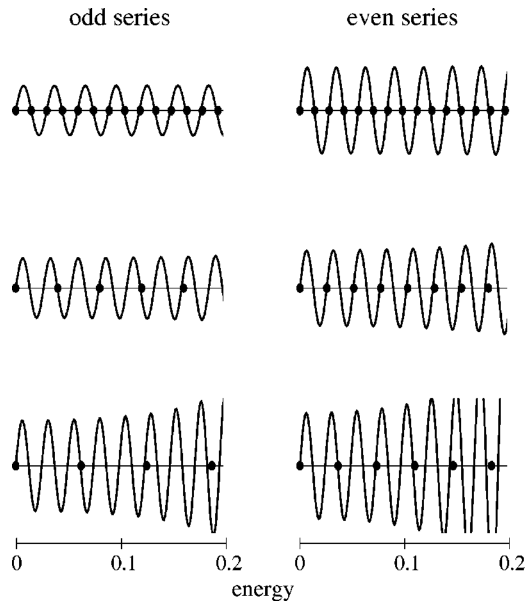

FIG. 3. Exact dependence $\mathcal{D}_{\mathcal{N}}(E)$ (solid lines) for $\mathcal{N}=401$ and different values of applied potential $eV=4\cos\pi/m$, where (from top to bottom) $m=3,5,7$ (to the left) and $m=4,6,8$ (to the right). The odd and even series $E_j^{o\{(m-1)/2,1\}}=jm\mathcal{E}$ and $E_j^{e\{(m-2)/2,1\}}=j(m/2)\mathcal{E}$, $j=0,1,2,\ldots$, respectively, are indicated by filled circles.

$5\mathcal{E}_{5/1}/3$ and $5\mathcal{E}_{5/2}$, respectively. Using the equidistant approximation, it can be shown that for the voltages defined in Eq. (12) the middle part of one-electron spectrum has a WSL form

$$E_l = l\,\frac{1}{1-2m'/m}\mathcal{E}_{m'/m}, \quad l=0,1,2,\ldots, \tag{13}$$

but with *noncanonical level spacing*.

Thus a proper tuning of the applied voltage ($eV<4$) should give rise to a *noncanonical* WSL. Depending on the applied potential, the level spacing within such WSLs can be, in particular, of double, triple, or fractional (>1) value of that would be in the corresponding WSL in a hypothetic infinite crystal. Changing the tuning (i.e., changing m, m') should result in a predictable replacement of one noncanonical WSL by the other. Let, for instance, at the given voltage V, $\mathcal{E}=eV/(\mathcal{N}-1)$, a double-$\mathcal{E}$ spaced WSL is observed, i.e., there are levels with energies $0, \pm2\mathcal{E}, \pm4\mathcal{E}, \pm6\mathcal{E},\ldots$. Then, according to Eq. (12) $m=4$, $m'=1$. If we decrease the voltage to the value of V' such that $\arccos(eV'/4)/\arccos(eV/4)=4/3$, a triple-$\mathcal{E}$ spaced WSL $[0,\pm3\mathcal{E}',\pm6\mathcal{E}',\pm9\mathcal{E}',\ldots,\mathcal{E}'=eV'/(\mathcal{N}-1)]$ has to emerge instead.

The established property of the es band is a reminiscence of approximately equidistant levels (with the spacing equal to π/N) near the center of zero-field band. The electric field changes the level spacing but not the spectrum symmetry. Hence the equidistant middle spectrum is expected rather than surprising. We just found the values of the applied voltage needed for the level spacing to be equal to a ‘‘certain amount of $\mathcal{E}$.’’

Similar results can be obtained for $\mathcal{N}=2N$. In this case, the noncanonical WSLs are described by Eq. (13), where l is replaced by $l+1/2$, and $\mathcal{E}_{m'/m}$ is determined by the equality $N\mathcal{E}_{m'/m}=2\cos[\pi(m'/m)]$.

IV. HIGH VOLTAGES, WANNIER-STARK SPECTRUM

If the electrostatic potential energy is larger than $eV=4$ [at which the es band shrinks to *one* ($\mathcal{N}$ is odd) or *no* ($\mathcal{N}$ is even) states], the extension of all tilted-band states is smaller than the crystal thickness, see lower diagram in Fig. 1. The middle part of the tilted band is separated from both sides of the crystal by classically forbidden regions. The corresponding energy interval of the spectrum $0<E<eV/2-2$ will be referred henceforth as the Wannier-Stark band or WS band. To simplify further discussion, we assume that $\mathcal{N}$ is an odd positive integer. However, all the conclusions of this section remain valid also for $\mathcal{N}=2N$.

At high voltages and for energies $E=n\mathcal{E}$ we have instead of Eq. (11) (Ref. 18)

$$\mathcal{D}_{2N+1}(n\mathcal{E})\approx(-1)^{N+n+1}\frac{\mathcal{E}}{\pi}\frac{\sinh\{2n\cosh^{-1}(\mathcal{E}N/2)\}}{\sinh\{\cosh^{-1}(\mathcal{E}N/2)\}}, \tag{14}$$

implying that the energy levels within the WS-band are *not described exactly* by equation $E_n=n\mathcal{E}$. Hence the WSL in finite systems must always be understood in an asymptotic sense. This point has been emphasized by Stey and Gusman,[14] who showed that corrections to the WS energies in finite crystals are exponentially small, if $\mathcal{N}\gg1$. Below we obtain a fully determined expression of the correction term.

For the WS-band energy interval, the second summand in Eq. (3) is dominating for any E, i.e.,

$$\mathcal{D}_{2N+1}(E)\approx -Y_{(E+N\mathcal{E})/\mathcal{E}+1}\left(\frac{2}{\mathcal{E}}\right)J_{(E-N\mathcal{E})/\mathcal{E}-1}\left(\frac{2}{\mathcal{E}}\right). \tag{15}$$

In terms of the positive orders of the Bessel functions the above equation may be rewritten as

$$\begin{aligned}\mathcal{D}_{2N+1}(E)\approx(-1)^N Y_{(E+N\mathcal{E})/\mathcal{E}+1}\left(\frac{2}{\mathcal{E}}\right)\\ \times\left\{J_{(N\mathcal{E}-E)/\mathcal{E}+1}\left(\frac{2}{\mathcal{E}}\right)\cos\left(\frac{\pi E}{\mathcal{E}}\right)\right.\\ \left.+Y_{(N\mathcal{E}-E)/\mathcal{E}+1}\left(\frac{2}{\mathcal{E}}\right)\sin\left(\frac{\pi E}{\mathcal{E}}\right)\right\}.\end{aligned} \tag{16}$$

Thus the roots of $\mathcal{D}_{2N+1}(E)$ are given by

$$J_{(N\mathcal{E}-E)/\mathcal{E}+1}\left(\frac{2}{\mathcal{E}}\right)\cos\left(\frac{\pi E}{\mathcal{E}}\right)+Y_{(N\mathcal{E}-E)/\mathcal{E}+1}\left(\frac{2}{\mathcal{E}}\right)\sin\left(\frac{\pi E}{\mathcal{E}}\right)=0. \tag{17}$$

Using in the above relation the Debay asymptotics of Bessel functions,[20] we arrive at

$$\tan\left(\frac{\pi E}{\mathcal{E}}\right)=\frac{1}{2}\exp\left(-\frac{4\Phi_{\delta'}}{\mathcal{E}}-2\,\delta'\right), \tag{18}$$

where $\delta = \cosh^{-1}\{(N\mathcal{E}-E)/2\}$, and $\Phi_{\delta'} = \delta' \cosh \delta' - \sinh \delta'$.

For the energies, which satisfy the condition $\Phi_{\delta'} \gg \mathcal{E}$, the solution to the above equation reads

$$E = E_n = n\mathcal{E} + \frac{\mathcal{E}}{2\pi} \exp\left(-\frac{4\Phi_{\delta'_n}}{\mathcal{E}} - 2\delta'_n \right), \qquad (19)$$

where $\delta'_n = \cosh^{-1}\{\mathcal{E}(N-n)/2\}$. The above expression appears to be accurate except the energies close to the value of $eV/2-2$. If, in addition, $\mathcal{E}(N-n) \gg 1$ ($2\cosh \delta'_n \approx \exp \delta'_n$), Eq. (19) takes an especially simple form

$$E = E_n = n\mathcal{E} + \frac{2\mathcal{E}}{\pi(eV-2\mathcal{E}n)^2} \left\{ \frac{e}{\mathcal{E}(N-n)} \right\}^{2(N-n)}, \qquad (20)$$

giving another explicit expression of corrections to the Wannier energies $E = n\mathcal{E}$.

Unlike the related result of Stey and Gusman,[14] Eqs. (19) and (20) are derived in a straightforward manner and do not contain an undefined constant, as does Eq. (47) in the cited work. Moreover, the exponential correcting term in Ref. 14 lacks the necessary restriction $\mathcal{E}(N-n) > e$.

V. SURFACE LOCALIZED STATES BAND $|2-eV/2| < E < 2 + eV/2$

As distinct from the bulk states spectrum discussed so far, the treatment of the sls-band spectrum is performed identically in the cases of low and high voltages. Within the energy interval $|2-eV/2| + \mathcal{E} < E < 2 + eV/2 - \mathcal{E}$, determinant (3) takes the form

$$\mathcal{D}_{\mathcal{N}}(E) \approx \frac{\mathcal{E}}{\pi} \sqrt{\frac{1}{\sin \xi' \sinh \delta}} \cos\left(\frac{2\Phi_{\xi'}}{\mathcal{E}} + \xi' - \frac{\pi}{4} \right) \times \exp\left(\frac{2\Phi_\delta}{\mathcal{E}} + \delta \right), \qquad (21)$$

where

$$2\cosh \delta = E + \frac{eV}{2}, \quad \Phi_\delta = \delta \cosh \delta - \sinh \delta. \qquad (22)$$

Notice that the energy in the sls band $E = 2\cosh \delta - eV/2$ corresponds to an imaginary value of quasi-impulse $\xi = i\delta$, which is defined by Eq. (5a). The latter transforms into Eq. (22) under replacements $\xi \to i\delta$ and $\Phi_\xi \to i\Phi_{i\delta}$. It also deserves emphasizing that, in contrast to the bulk states, which are classified by the pseudo-quasi-impulse at low voltages and by canonical WSLs at high voltages, the quantization of sls-band energies is determined by one and the same quantity.

As it follows from Eq. (21), the quantum numbers of sls band states $\xi_n^{sls} = \pi n$, $n = 1,2,\ldots,n^{sls}-1$, n^{sls}, are given by solutions to equation

$$\xi^{sls} \equiv \frac{2}{\mathcal{E}} \Phi_{\xi'} + \xi' + \frac{\pi}{4} = \pi n, \quad n = 1,2,\ldots,n^{sls}, \qquad (23)$$

where the number of states in the sls band is defined as $\pi n^{sls} = \xi^{sls}(E = |2-eV/2|)$.

At low voltages ($eV < 4$), we have

$$n^{sls} = \left[\frac{2}{\pi\mathcal{E}} \left\{ \sqrt{eV\left(1-\frac{eV}{4}\right)} - \left(1-\frac{eV}{2}\right) \times \arccos\left(1-\frac{eV}{2}\right) \right\} + \frac{1}{\pi} \arccos\left(1-\frac{eV}{2}\right) + \frac{1}{4} \right]. \qquad (24a)$$

For odd $\mathcal{N}$, the $E=0$ level is included in the above definition. Therefore the number of states in both upper and lower sls bands is equal to $2n^{sls}-1$. The quantities n^{es} and n^{sls} must satisfy the identity $n^{sls} + n^{es} = [(\mathcal{N}+1)/2]$ (the state number conservation) which, in particular, can be used to check the accuracy of the above description.

Equation (24a) tells us that the number of states in the sls band is controlled by both the crystal thickness and field strength. For instance, at very low voltages $eV \ll 1$, to the first approximation

$$n^{sls} = \left[\frac{2\mathcal{N}}{3\pi} \sqrt{eV} \right], \qquad (24b)$$

i.e., n^{sls} depends on the crystal thickness more strongly than on the field strength.

At high voltages (when the sls-band levels are within the energy interval $eV/2-2 < E < eV/2+2$), the sls band is of the maximal width. The latter is equal to the unperturbed band width. In this case $\xi^{sls}(E = eV/2-2) = 2\pi/\mathcal{E} + 5\pi/4$, so that instead of Eq. (24a), we now have

$$n^{sls} = \left[\frac{2}{\mathcal{E}} + \frac{5}{4} \right], \qquad (24c)$$

which contains an extra term 5/4 as compared to the result obtained for a semi-infinite crystal by Yakovenko and Goan.[17] Thus for $eV > 4$, the number of sls-band states is fully controlled by the field strength. A simple comparison of the latter equation with Eq. (24b) clearly indicates that the manifestation of electric field effects in thin crystals may be very much different from that is learned from semi-infinite models.

A. Analytical solutions for eigenvalues in sls bands

In certain energy intervals Eq. (23) or, equivalently, equation $\sin \xi^{sls} = 0$ can be solved analytically. To proceed with finding such solutions, we first answer a relevant question: which minimal voltage $V_{\min}$ is to be applied for the first localized state (on either side of the es band) to appear?

Note that, if the $n=1$ on-site energy in Eq. (1) were fixed at zero value and $\mathcal{E}$ is positive, the lower edge of the spectrum would be field independent and has the energy $E' = -2$. On the other hand, it is well known that a perturbation of band states results in the appearance of a local state whenever the corresponding secular equation has a solution outside the band, in the given case, for $|E'| \leq 2$. Thus to find $V_{\min}$, we have to solve the secular equation with respect to $\mathcal{E}$ at the band edge $E' = 2$. Alternatively, with the choice of the energy reference point used throughout the discussion, we have to find the solution to Eq. (2) with $E=2-eV/2$. This is equivalent to solving the equation

$$\frac{2}{\mathcal{E}}\frac{(eV)^{3/2}}{3}+\sqrt{eV}=\frac{3\pi}{4}, \tag{25}$$

which follows from Eq. (23) under conditions $n^{sls}=1$ and $eV\ll 1$. The only real root of the above equation is well approximated by

$$eV_{\min}\approx(3/4)3^{1/3}(\pi\mathcal{E})^{2/3}-\mathcal{E}. \tag{26}$$

Now we restrict ourselves to the energy intervals near the bottom, middle, and top of sls band. These are (1) $E^t_{sls}-E\ll 1$; (2) $|E-eV/2|\ll 1$ $(eV>2)$; and (3) $E-E^t_{\mathrm{WS}}\ll 1$ $(eV>4)$. Clearly, the latter of the indicated intervals exists only in the case of high voltages.

(1) $E^t_{sls}-E\ll 1$. Under the condition $\xi'\approx\sqrt{E^t_{sls}-E}\ll 1$, $\Phi_{\xi'}$ can be expanded in powers of ξ'. Using the expansion in Eq. (23) yields

$$E^t_{sls}-E_n=\left\{\frac{3}{2}\pi\left(n-\frac{1}{4}\right)\mathcal{E}\right\}^{2/3}-\mathcal{E} \tag{27}$$

which is nothing but the Airy spectrum, see, e.g., Ref. 21.

Because of the condition of its derivation, Eq. (27) is applicable only to sls-band top levels; a comparison with exact solutions to Eq. (2) is represented in Fig. 2(b). As was shown by Fukuyama *et al.*,[15] the same relation also can be obtained for a semi-infinite tight-binding band, see below. It is more familiar for the free electron or effective mass approximation.[22] To get a more common representation of Eq. (27), one has to replace in Eq. (27) the energy scale β by its equivalent in the continuous limit: $\beta\to\hbar^2/(2m^*a^2)$, m^* is the electron effective mass.

(2) $|E-eV/2|\ll 1$, $eV>2$. For the energies close to $eV/2$ we find that equation

$$E_m\approx\frac{eV}{2}+2\mathcal{E}\left(\frac{2}{\pi\mathcal{E}}+\frac{3}{4}-\left[\frac{2}{\pi\mathcal{E}}+\frac{3}{4}\right]\right)+2\mathcal{E}m, \tag{28}$$

where $m=0,\pm1,\pm2,\ldots$, $|m|\ll 1/(2\mathcal{E})$, gives the eigenvalues of matrix (1) with a good accuracy. Thus, for energy levels lying above (m is zero or positive) and below (m is negative) $E=eV/2$, the level spacing is equal to the *doubled* spacing in the corresponding (canonical) WSL.

(3) $E-E^t_{\mathrm{WS}}\ll 1$ $(eV>4)$. In this case, it is easy to see from Eq. (23) that $E-E^t_{\mathrm{WS}}=2-\mathcal{E}(n-5/4)$, where n is of the order of n^{sls} defined by Eq. (24c). This gives the Wannier quantization rule

$$E_{n+1}-E_n=\mathcal{E}. \tag{29}$$

However, here it refers to the bottom of sls band having a triangular shape. We stress that the validity of Eq. (29) for energies outside, though close to the WS band, is not at all obvious.

As is mentioned in the introduction, for a semi-infinite crystal Eq. (27) was repeatedly derived earlier by different techniques.[13,15,17] At the same time, Eqs. (28) and (29) have never been reported in the given context, at least to the best of our knowledge.

VI. SEMI-INFINITE TILTED TIGHT-BINDING BAND

The main purpose of this section is to establish the relationship between electric field effects on the electronic spectrum in finite and semi-infinite crystals, and to make clear links with previous results. For a semi-infinite crystal, the energy reference used in all above derivations is irrelevant. With the $n=1$ site energy fixed at $E=0$, Hamiltonian (1) takes the form $\beta^{-1}H_{nm}=\mathcal{E}(n-1)+\delta_{|n-m|,1}$, and the eigenvalue equation (2) can be rewritten as

$$\mathcal{D}_{\mathcal{N}}(E)=J_{\mu+1}(z)Y_{\mu-\mathcal{N}}(z)-Y_{\mu+1}(z)J_{\mu-\mathcal{N}}(z)=0. \tag{30}$$

In the limit $\mathcal{N}\to\infty$ taken at $\mathcal{E}>0$, one electron levels are determined by

$$J_{-\mu-1}(2/\mathcal{E})=0, \tag{31}$$

and fill a semi-infinite interval $E>-2$.

Equation (31) (or its equivalents) has been used for a description of electric field effect on the electronic spectrum by Fukuyama *et al.*[15] and in subsequent publications. In terminology of present discussion, Eq. (31) is relevant *only to the high voltage case*, since $eV\to\infty$ as $\mathcal{N}\to\infty$. As is intuitively expected, all principal results regarding the sls band (the high voltage case) and WS band must be possible to obtain from Eq. (31). The following confirms the expectation and thus, links our results with previous studies.

As in the case of a finite $\mathcal{N}$, the explicit expression of solutions to Eq. (31) depends on which part of the spectrum is described. By analogy with the preceding discussion, the set of electronic levels within the energy interval $-2<E<2$ is referred to as the sls band, and the remaining (infinite) part of the spectrum is called the WS band.

First we consider the energy interval $-2<E<-\mathcal{E}$, i.e., the lower half of the sls band. For these energies

$$\begin{aligned}J_{-\mu-1}(2/\mathcal{E})&=J_{|E+\mathcal{E}|/\mathcal{E}}(2/\mathcal{E})\\&\approx\sqrt{\frac{\mathcal{E}}{\pi\sin\xi}}\cos\left[\frac{2}{\mathcal{E}}(\sin\xi-\xi\cos\xi)-\frac{\pi}{4}\right],\end{aligned} \tag{32}$$

where $\cos\xi=|E+\mathcal{E}|/2$. Note that Eq. (32) coincides with Eq. (2.8) from Ref. 15 to within the multiplier and the energy reference point. The substitution of Eq. (32) in Eq. (31) yields

$$\frac{2}{\mathcal{E}}(\sin\xi-\xi\cos\xi)+\frac{\pi}{4}=\pi n,\quad n=1,2,\ldots, \tag{33}$$

which, if compared with Eq. (23), lacks a linear term in ξ. Such a term would be present, if the $n=1$ on-site energy were equal not to zero but $\mathcal{E}$.

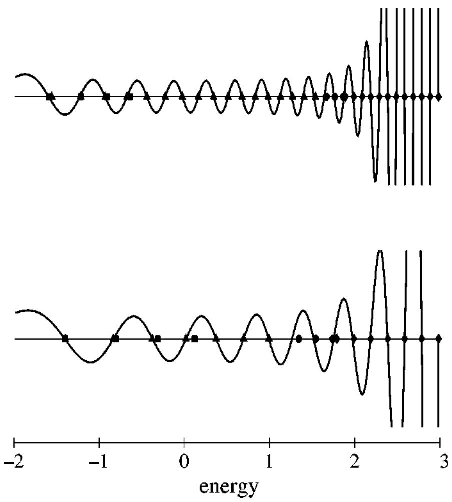

FIG. 4. Exact dependence of $J_{-E/\mathcal{E}-1}(2/\mathcal{E})$ on E (solid lines) for $\mathcal{E}=0.1$ (upper curve) and $\mathcal{E}=0.2$ (lower curve). Approximate values of eigenenergies given by Eqs. (34), (35), (37) and $E_n=\mathcal{E}n$ are marked by filled squares, triangles, circles, and diamonds, respectively.

Near the bottom of the sls band ($E+2\ll 1$) we have $\xi^2=E+2+\mathcal{E}\ll 1$, under which condition the electronic levels can be approximated by the expression

$$2+E_n=\left[\frac{3\mathcal{E}}{2}\pi\left(n-\frac{1}{4}\right)\right]^{2/3}-\mathcal{E},\quad n=1,2,\ldots. \tag{34}$$

The above reconfirms Eq. (27). The accuracy of the approximation is illustrated in Fig. 4, where the values of E_n are indicated by filled squares.

In the middle of the sls band $\xi=\pi/2-\alpha$ with $\alpha\ll 1$. As a result, for $|E|\ll 1$ the solution to Eq. (31) may be represented as

$$E_n=-\pi\left\{1-\sqrt{1-\frac{4\mathcal{E}}{\pi}\left(\frac{2}{\pi\mathcal{E}}+\frac{\pi}{4}-n\right)}\right\}-\mathcal{E}, \tag{35}$$

where $n=n_1$, $n_1\pm 1$, $n_1\pm 2,\ldots$, and $n_1=[2/(\pi\mathcal{E})+\frac{1}{4}]$. The respective energies are indicated as filled triangles in Fig. 4.

By the condition of the above derivation, the square root in Eq. (35) must be close to unit. One can see therefore, that except the energy reference point E_{n_1}, the obtained relation is essentially the same as that given in Eq. (28). It turns out that approximation (35) is surprisingly accurate far beyond the limit of its applicability, see Fig. 4.

Consider now the energy interval $-\mathcal{E}<E<2-\mathcal{E}$, which represents the positive half of the sls band. Under the restriction indicated, and $\mathcal{E}\ll 1$, the Bessel function in Eq. (31) can be expressed in the form (32) but with the summand $(E+\mathcal{E})\pi/\mathcal{E}$ added to the cosine argument. Equation (33) is thus replaced by

$$\frac{2}{\mathcal{E}}(\sin\xi-\xi\cos\xi)+\frac{\pi}{4}+\frac{(E+\mathcal{E})\pi}{\mathcal{E}}=\pi n. \tag{36}$$

At the top of the sls band $2-E-\mathcal{E}\ll 1$, Eq. (36) simplifies to

$$E_n\approx 2-\mathcal{E}-\xi^2=(n-1)\mathcal{E}-\frac{\mathcal{E}}{4}, \tag{37}$$

which is similar to Eq. (29).

Finally, for the semi-infinite interval $E>2-\mathcal{E}$ (WS band)

$$J_{-\mu-1}(2/\mathcal{E})\approx\sqrt{\frac{\mathcal{E}}{\pi\sinh\delta}}\left(\frac{1}{2}e^{-(2/\mathcal{E})(\delta\cosh\delta-\sinh\delta)}\times\cos\frac{\pi(E+\mathcal{E})}{\mathcal{E}}+e^{(2/\mathcal{E})(\delta\cosh\delta-\sinh\delta)}\sin\frac{\pi(E+\mathcal{E})}{\mathcal{E}}\right), \tag{38}$$

where $2\cosh\delta=E+\mathcal{E}$. As a result, Eq. (31) takes the form

$$\tan\left(\frac{\pi E}{\mathcal{E}}\right)=-\frac{1}{2}\exp\left\{-\frac{4}{\mathcal{E}}(\delta\cosh\delta-\sinh\delta)\right\}, \tag{39}$$

which is to be compared with Eq. (18). Under the condition $4(\delta\cosh\delta-\sinh\delta)\gg\mathcal{E}$, and $\mathcal{E}n\gg 1$, the solution to (39) can be approximated by

$$E=E_n=n\mathcal{E}-\frac{\mathcal{E}}{2\pi}\left\{\frac{e}{\mathcal{E}(n+1)}\right\}^{2(n+1)}, \tag{40}$$

showing that for energies of the order or larger than e, edge effects on the WSL are negligible.

VII. SYNOPSIS

This work is addressed, first of all, to dielectric crystalline layers (and superlattices), whose thickness (length) reaches the edge of the direct tunneling. On the other hand, from below ten to fifteen atomic layers (wells) are required, to meet the condition $\mathcal{N}\gg 1$. One-particle, single band approach implies that in a full scale, the obtained results are applicable only to empty bands which are weakly mixing with the others in the actual voltage region. Furthermore, the condition of a symmetric zero-field band is likely to be crucial for the existence of canonical, as well as noncanonical WSLs. In the most of relevant real systems such symmetry is rather not the case. However, the band asymmetry is often small and, in the first approximation, can be neglected. Even though the model assumptions used explicitly or implicitly in the above derivations are quite demanding, there is a certain Heuristic value of obtained results highlighting the main trends of electric field effects on the surface and bulk electronic states which always coexist in real systems.

The results of the above analysis of field effects on one-electron spectrum are summarized in Fig. 5 which exposes quantum mechanical content of classical diagrams in Fig. 1. A constant electric field increases an average interlevel separation in a nontrivial way and redistributes electronic levels over the band. In zero field, the density of states is minimal in the band center, while it is maximal at the band edges. At high voltages, the level distribution is just reversed. In an obvious limit $eV\gg E^0_{\rm bw}$ (not considered here), the $\mathcal{E}$-spaced

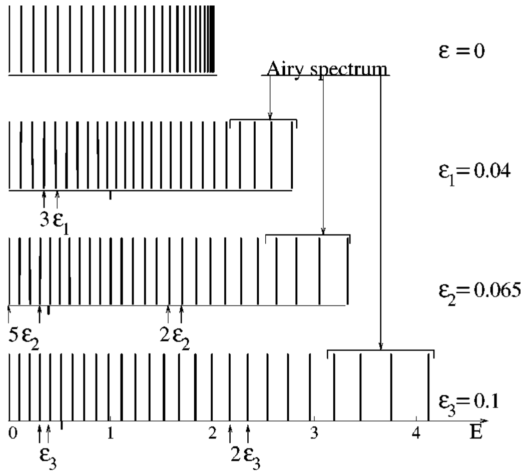

FIG. 5. Band levels (vertical lines) calculated for $\mathcal{N}=51$ and electrostatic energy difference (from top to bottom) $eV=0$, $eV=2$, $eV=4\cos\pi/5\approx3.24$, and $eV=5$. Arrows mark levels within specific series; accented ticks indicate the top of WS (lower diagram) and es band. Airy spectrum refers to levels given by Eq. (27).

levels are distributed homogeneously throughout the spectrum.

The following lists main transformations of the band spectrum in response to an increase of the applied voltage. (i) At $eV=2.3\mathcal{E}^{2/3}-\mathcal{E}$ the first two surface localized states appear, above and below the es band. At higher voltages, two sls bands emerge. (ii) At certain voltages, which satisfy Eq. (12), the middle part of the spectrum consists of evenly spaced levels forming a noncanonical WSL with the level spacing equal to $\mathcal{E}$ times integer or fractional number (>1 and dependent on the applied voltage). For experimental observations, however, the WSLs $E_l=l\{1+2m'/(m-2m')\}\mathcal{E}$, where the second summand in brackets is not too small, may be most meaningful. The level spacing $3\mathcal{E}$ is expected at $eV=E^0_{\rm bw}/2$. The increase of the applied voltage by $\sqrt{2}$ should result in a new level spacing equal to the doubled dimensionless field parameter. Equations (12) and (13) suggest a number of other tests for validating noncanonical WSLs experimentally. (iii) When $eV=E^0_{\rm bw}$, the spectrum consists of two sls bands, and at higher voltages the WS-band emerges. At $eV>E^0_{\rm bw}$ the band spectrum gradually changes its character from the canonical WSL in the middle to the Airy type at the edge parts of the spectrum. This changing is such that in the middle of the sls bands, the level spacing equals to the doubled Wannier spacing, see Fig. 5. It is worth stressing that the related (and often used) semi-infinite model is just unable to describe the electric field effects, when the electrostatic energy is smaller, than the zero-field band width. The model of finite and tilted tight-binding band is thus shown to be much more rich and fertile physically, than is commonly thought.

From the experimental point of view, the most straightforward application of our results is seen in the field dependence of peaks of through tilted band tunneling probability. Due to the band states, an electron with properly tuned kinetic energy can be transmitted across a thin crystal layer or superlattice by means of resonance tunneling. The position of peaks in the transmission spectrum [i.e., the dependence of resonance tunneling probability on energy $T(E)$] can be controlled by the applied voltage, as is explained in the above discussion. Precisely, noncanonical WSLs can be observed in the case of low voltages and weak coupling of the tunnel region (modeled by the tilted band) with the source and drain of electrons. Such WSLs will manifest themselves as evenly spaced central peaks in the transmission spectrum. Similarly, in the region of sls-assisted tunneling, peaks with $\mathcal{E}$-, double-$\mathcal{E}$-, and Airy-type spacing can be observed. Probably, ballistic-electron-emission microscopy that can probe heterostructure transmission spectra in a nondestructive manner[23] is the most suitable but not the only technique for the purpose.

Methodologically, the use of the above approach can be straightforwardly extended to the analysis of the transmission spectrum dependence on the field, energy, and length of tunneling region. Further usage is envisioned in a description of the Franz-Keldysh effect which reflects changes of the band spectrum induced by a constant electric field. The through gap Zener tunneling also associates with field affected electron states. Signatures of the Wannier-Stark effect in Zener tunneling and Franz-Keldysh absorption, which involve at least two bands, are expected but not well understood yet. Even within the relevant zero-approximation model, which is two noninteracting bands influenced by a constant electric field, the above mentioned processes have not received an adequate theoretical treatment. Some preliminary results have been reported[24] and work in this direction is in progress.

ACKNOWLEDGMENTS

This research was supported by the Swedish Research Council (former NFR and TFR). Partial support from INTAS under Grant No. 99-864 is also gratefully acknowledged.

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