Triple-, double-, and fractionally-spaced Wannier–Stark ladders

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Abstract

The concept of Wannier–Stark (WS) quantization literally refers to infinite crystals or bulk electron states. In real finite crystals, the bulk and edge or surface states always coexist. Moreover, when the surface states are already considerably localized due to the presence of a constant electric field, the WS ladder (WSL) and hence the WS localization may not yet come into play, at least, in the canonical form of WS quantization $E_j = \text{const} \pm j\epsilon$, $j = 0, 1, 2, ...$ (E_j is the one-electron energy, and the parameter ϵ is associated with the electric field strength). We show that at certain voltages V_m, $m = 3, 4, ...$, which are lower than V_∞ needed for the WS band opening (the sub-WS regime), the mid-spectrum levels can form triple-, double-, and fractional-spaced WSLs, where $E_l = \pm l[1 + 2/(m - 2)]\epsilon_m$. It is also found that in the WS regime, the quantization of surface localized states (sls) smoothly changes from the Airy type (at the spectrum edges) to the WS type with a pronounced energy interval in between, where the level spacing doubles that of canonical WSL. Possible experimental manifestations of predicted effects are also outlined. © 2001 Elsevier Science Ltd. All rights reserved.

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1. Introduction

Due to periodic atomic structure, band states in finite crystals are quantized regularly. For an ideal crystal lattice the electron states (Bloch electrons) are labeled by discrete values of quasi-momentum, which are determined by the boundary conditions. Both factors, periodicity and confinement are of importance for the regular quantization of band states. However, as was predicted by Wannier in 1960 [1,2], the electron band spectrum of a crystal subjected to a uniform electric field, which breaks down periodicity, consists of discrete, evenly spaced levels forming a ladder. This kind of band spectrum has come to be known in literature as the Wannier–Stark ladder (WSL).

Literally, the Wannier–Stark (WS) quantization refers to infinite crystals or bulk electron states which have been studied extensively in various aspects, [3–7] see also review articles [8,9,10] and references therein. The electric field effect on the edge or surface states has received much less attention and is mainly explored numerically [11–15]. Few

related analytical results have been obtained so far for finite systems; all of them in the one band nearest-neighbor approximation [16–19]. In particular, Fukuyama et al. [19] and in a more recent work Yakovenko and Goan [20] have shown explicitly that WSL always coexists with the Airy spectrum. The derivation of this result implies, however, that it is valid only for the high voltage limit, when the total electrostatic energy is larger, than the electron bandwidth. In fact, in the semi-infinite crystal discussed in Refs. [19,20], the electrostatic energy is infinite. For this reason, the model does leave out of consideration, the case of low voltages (the sub-WS regime). With regard to the latter, general trends of the band levels rearrangement in response to the increase of applied voltage are not well understood. Numerical modeling in this direction, [14] though useful, is insufficient, because the results are always restricted to the given choice of system parameters.

The WS effect may have specific signatures in a number of phenomena in crystalline and molecular systems. In particular, it is relevant to the through empty band tunneling, Bloch oscillations [8,9,10], Zener tunneling [3,21,22], and Franz–Keldysh absorption [4,23,24]. Combined with the exact results obtained earlier, the present analysis provides

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a firm basis of further exploration of the above mentioned and related effects. It also suggests reliable tests for more sophisticated models.

In the current letter, we prove the existence of specifically quantized series at sub-WS voltages and show a new type of quantization (distinct from Airy and WS quantization) in the case of intermediate and high electric fields.

2. Problem statement

Let us 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 (shown 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
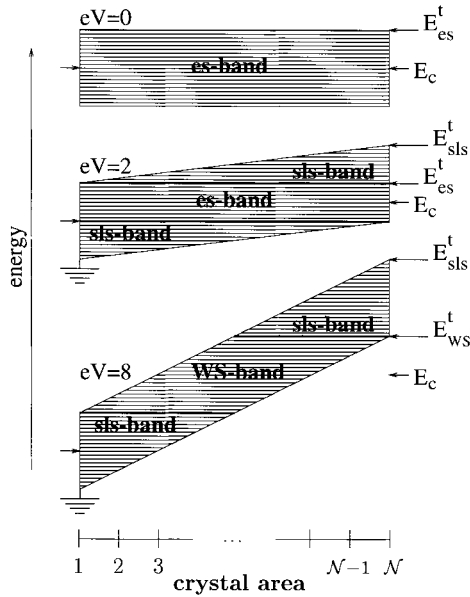

Fig. 1. Untilted (upper) and tilted (middle and lower) bands of electronic levels in a crystal of finite thickness. The crystal area in the direction of electric field is occupied by $\mathcal{N}$ atomic layers. In the division of the band spectrum at a particular voltage V, the center of the spectrum E_c serves as a reference point. At $V = 0$ all states within the band are extended from the left hand side to the right hand side of the crystal. These states form an *extended state* band or es-band which is indicated by a shaded rectangle. The zero-field bandwidth is equal to 4 in units of the electron resonance transfer energy. At $0 < eV < 4$ in addition to the (narrowed) es-band, two bands of surface localized states appear (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 of a respective band: $E^t_{es} = 2 - eV/2$, $E^t_{WS} = eV/2 - 2$, $E^t_{sls} = 2 + eV/2$.

the form

$$H_{mn} = -eFa(N + 1 - n - \theta)\delta_{m,n} + \beta\delta_{|m-n|,1}, \tag{1}$$

where $n, m = \overline{1, \mathcal{N}}$, $\theta = 0.5$ for $\mathcal{N} = 2N$, and $\theta = 0$ for $\mathcal{N} = 2N + 1$; 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 $\epsilon = eFa/\beta$ stands for the field parameter, and $eV = \epsilon(\mathcal{N} - 1)$ is the energy of electrostatic potential applied to the bounding surfaces which are perpendicular to one of the crystal principal axes.

The eigenvalues of matrix H_{mn} are given by zeros of the determinant

$$D_{\mathcal{N}}(E) = |E\delta_{m,n} - \beta^{-1}H_{mn}| = 0. \tag{2}$$

As it has been shown by many authors [17–19,25], the Hamiltonian matrix determinant can be expressed in terms of Bessel functions. For the particular choice of the energy reference point and lattice sites numbering, as is shown in Fig. 1, we have [25]

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

where $\mu \equiv E/\epsilon$, $z \equiv 2/\epsilon$, and $J_\mu(z)$ and $Y_\mu(z)$ are the Bessel functions of the first and second kind, respectively.

We discuss first particular solutions to Eq. (2) for an odd number of atomic layers $\mathcal{N} = 2N + 1$. The case of an even $\mathcal{N}$ can be examined similarly. Because of the spectrum symmetry only positive energies, i.e., the upper half of the spectrum will be considered.

3. Fractional quantization of the mid spectrum

For the purpose it is convenient to transform $D_{\mathcal{N}}(E)$ to the form of a finite order polynomial. For odd values of $\mathcal{N}$ Eq. (3) is transformed into [25]

$$D_{2N+1}(E) = (-1)^N \frac{\epsilon}{\pi} \sum_{l=0}^{N} \epsilon^{2l+1} \binom{N + l + 1}{N - l}$$
$$\times (\mu)_{l+1}(-\mu + 1)_l, \tag{4}$$

where notation $(...)_l$ stands for the Pohgammer symbol $(a)_l = a(a + 1)...(a + l - 1)$. The above expression is equivalent to the Lommel polynomial form of Eq. (2) which was first derived by Stay and Gusman [17], and by Saitoh [18].

Let us show that at certain values of eV equation $D_{2N+1}(E) = 0$ does have solutions $E = E_n = n\epsilon$, where n takes positive integer values to be specified. For $E = E_n$ all terms in Eq. (4) with $l \geq n$ are cancelled. Hence we

can write

$$D_{2N+1}(E_n) = \frac{\epsilon}{\pi} \sum_{l=0}^{n-1} (-1)^{l+N}$$

$$\times \epsilon^{2l+1} \frac{(N+l+1)!}{(N-l)!} \frac{(n+l)!}{(n-l-1)!(2l+1)!}. \tag{5}$$

For $n \ll N$ we have $(N+l+1)!/(N-l)! \approx N^{2l+1}$ (the Stirling formula). And performing the summation, we obtain

$$D_{2N+1}(E_n) \approx (-1)^{N+n+1} \frac{\epsilon}{\pi} \frac{\sin\{2n \arccos(\epsilon N/2)\}}{\sin\{\arccos(\epsilon N/2)\}}, \tag{6}$$

if $\epsilon N < 2$, and

$$D_{2N+1}(E_n) \approx (-1)^{N+n+1} \frac{\epsilon}{\pi} \frac{\sinh\{2n \cosh^{-1}(\epsilon N/2)\}}{\sinh\{\cosh^{-1}(\epsilon N/2)\}}, \tag{7}$$

if $\epsilon N > 2$.

It immediately follows from Eq. (6) that if $eV < 4$, *some* (or none) of electronic states have energies which are exactly equal to $n\epsilon$. These are the nth states for which $2n \arccos(N\epsilon/2) = \pi k$, and k is a positive integer. In particular, if the applied voltage satisfies the condition $V = V_m = (N-1)\epsilon_m$, where

$$\arccos\left(\frac{eV_m}{4}\right) = \frac{\pi}{m}, \qquad m = 3, 4, ..., \tag{8}$$

there exists a sequence of eigenvalues $E_n = n\epsilon_m$, where $n = 0, m, 2m, ...$, if m is odd ($E = E_j = jm\epsilon_m, j = 0, 1, 2, ...$), and $n = 0, m/2, m, 3m/2, ...$, if m is even [$E = E_j = j(m/2)\epsilon_m$]. It is seen from Eq. (8) that the first 'odd' series ($m = 3$) appears at $eV_3 = 2$ ($\epsilon_3 = 1/N$). The level spacing within this series is equal to $3\epsilon_3 = 3/N$. On the other hand, the es-band width at this voltage is a half of zero-field band width, showing that the level spacing must be near the value of $\pi/(N+1)$, so that $E = E_j \approx \pi j/(N+1)$. From the closeness of the latter estimate to the *nearly exact* solution $E_j = 3j\epsilon_3 = 3j/N$ (if N is large and we are not far from the spectrum center), it follows that the first *odd series* lists *all* levels in the mid part of es-band. The same is true about the first even series, which appears at $eV_4 = 2\sqrt{2} > eV_3(\epsilon_4 = \sqrt{2}/N > \epsilon_3)$. The second, third, and so on odd series, which in accordance with Eq. (8) appear at ever higher voltage V_5, V_7, and so on, coincide with each third, fifth and so on spectrum level. The energies of the second, third, and so on even series coincide with each second, third and so on spectrum level. Further analysis shows that the mid part of the spectrum consists of nearly equidistant levels.

Combining all these results yields the following structure of the mid part of one-electron spectrum at $2 < eV = eV_m < 4$

$$E_l = l\left(1 + \frac{2}{m-2}\right)\epsilon_m. \tag{9}$$

This equation implies that the WSL with ϵ-spaced levels, which appears at $eV > 4$, [17–19] is preceded by a $3\epsilon_3$-spaced WSL appeared at $eV_3 = 2$, then by a $2\epsilon_4$-spaced WSL at $eV_4 = 2\sqrt{2}$, and then by a $[1 + 2/(m-2)]\epsilon_m$-spaced WSL (a sort of fractionally quantized spectrum) at higher voltages V_m determined by Eq. (8). Thus it is possible to change one regular level spacing to the other by proper tuning of the applied voltage. For instance, by increasing the applied voltage from the value of V_5 to V_6, the WSL with $\frac{5}{3}\epsilon$ level spacing will be replaced by one where $E_{j+1} - E_j = \frac{3}{2}\epsilon$.

Finally we notice, as can be concluded from Eq. (7), for $eV > 4$ the spectrum *is never described exactly* by equation $E = E_n = n\epsilon$. By no means does this result exclude the existence of WSL in finite systems. It just shows that the WSL must always be understood in an asymptotic sense [17,26].

4. Surface localized states (sls) spectrum. Double-ϵ quantization in the mid of sls-band

From the analysis of $D_{\mathcal{N}}(E)$ as a function of energy it follows that in the energy intervals, which are shown in Fig. 1 as *extended states* (es), *surface localized states* (sls), and WS bands, the corresponding solutions to Eq. (2) have distinctive properties. Here we focus on the eigenvalues within the sls-band. These have been examined by Saitoh [18], Fukuyama at al. [19], and Yakovenko and Goan [20]. However, their results refer to the band edges, where the spectrum is nearly the same as in an infinite triangle well (the Airy spectrum). It will be shown soon that in the mid of sls-band, as well as in its part verging on the es-band, the electron state quantization is different.

The energy interval of sls-band is defined as $E_{es}^t \equiv 2 - eV/2 < E < 2 + eV/2 \equiv E_{sls}^t$, if $eV < 4$ (in such a case, the sls-band width is equal to eV); and $E_{WS}^t \equiv eV/2 - 2 < E < E_{sls}^t$, if $eV > 4$. In the latter case, the sls-band width has the maximal value which is equal to the width of untilted band. For $V \neq 0$, the crystal areas, which are classically accessible (shaded in Fig. 1) and forbidden (unshaded), are divided by sloped lines. As is seen in the figure, while the es-band states are extended over the entire length $\mathcal{N}$, the crystal thickness, the sls-band states tend to be preferably localized near one of the crystal boundary surfaces; for positive energies this is the right hand side surface.

Under the condition $\epsilon \ll 1$ and $\mathcal{N} \gg 1$, which covers most cases of interest[1], the secular equation determining the sls-band spectrum is expressible in a more simple form. This can be done by making use of standard approximate expressions of Bessel functions with large arguments and small or large orders [27]. Skipping lengthy calculations

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[1] The opposite case of large field parameter $\epsilon \gtrsim 1$ is easily treated by perturbation theory in the small parameter ϵ^{-1}; it may require, however, unrealistically high applied voltages.

we write only the result

$$D_{\mathcal{N}}(E) \approx \frac{\epsilon}{\pi} \sqrt{\frac{1}{\sin \xi \sinh \delta}} \cos \left(\frac{2\Phi_\xi}{\epsilon} + \xi - \frac{\pi}{4} \right) \times$$

$$\exp \left(\frac{2\Phi_\delta}{\epsilon} + \delta \right), \tag{10}$$

where

$$2\cosh \delta = E + \frac{eV}{2}, \qquad 2\cos \xi = E - \frac{eV}{2}, \tag{11}$$

$\Phi_\delta = \delta \cosh \delta - \sinh \delta$, and $\Phi_\xi = \sin \xi - \xi \cos \xi$. To derive Eq. (10), small energy intervals ϵ, which contain one or no levels at all, are excluded. Namely, $E_{es}^t + \epsilon < E < E_{sls}^t - \epsilon$, if $eV < 4$; and $E_{WS}^t + \epsilon < E < E_{sls}^t - \epsilon$, if $eV > 4$.

Using Eq. (10) in Eq. (2), one obtains

$$\sin \left(\frac{2}{\epsilon} \Phi_\xi + \xi + \frac{\pi}{4} \right) = 0, \tag{12}$$

so that

$$\xi^{sls} \equiv \frac{2}{\epsilon} \Phi_\xi + \xi + \frac{\pi}{4} = \pi n. \tag{13}$$

The latter equation labels the sls-band states by a single quantum number $\xi_n^{sls} = \pi n, n = 1, 2, ..., n^{sls}$. Smaller values of ξ_n^{sls} correspond to energies closer to the top of sls-band E_{sls}^t.

The number of states in the sls-band is given (up to unity) by

$$n^{sls} = \left[\frac{2}{\pi \epsilon} \left(\sqrt{eV \left(1 - \frac{eV}{4} \right)} - \left(1 - \frac{eV}{2} \right) \times \right. \right.$$

$$\left. \left. \arccos \left(1 - \frac{eV}{2} \right) \right) + \frac{1}{\pi} \arccos \left(1 - \frac{eV}{2} \right) + \frac{1}{4} \right], \tag{14}$$

if $eV < 4$, and by

$$n^{sls} = \left[\frac{2}{\epsilon} + \frac{5}{4} \right], \tag{15}$$

if $eV > 4$. In Eqs. (14) and (15), and henceforth, square brackets indicate the integer part of the argument. Eq. (15) up to the term 5/4 coincides with that obtained by Yakovenko and Goan [20] for a semi-infinite crystal. As is readily seen, at high voltages the number of sls-band states is fully controlled by the field strength only. However, this is not true at arbitrary voltages. For instance, at very low voltages $eV \ll 1$, Eq. (14) to the first approximation can be rewritten as

$$n^{sls} = \left[\frac{2\mathcal{N}}{3\pi} \sqrt{eV} \right], \tag{16}$$

i.e., n^{sls} depends on the crystal thickness $\mathcal{N}$ more strongly than on the field parameter ϵ. These results clearly indicate that the manifestation of electric field effects in thin crystals

may be very much different from those that can be obtained from semi-infinite crystal models.

In certain energy intervals (1) $E_{sls}^t - E \ll 1$, (2) $|E - eV/2| \ll 1 (eV > 2)$, and (3) $E - E_{WS}^t \ll 1 (eV > 4)$; Eq. (12) can be solved analytically. Let us consider the corresponding solutions.

(1) $E_{sls}^t - E \ll 1$. Under the condition $\xi \approx \sqrt{E_{sls}^t - E} \ll 1$, Φ_ξ can be expanded in powers of ξ. Using the expansion in Eqs. (11) and (12) yields

$$E_{sls}^t - E_m = \left\{ \frac{3}{2} \pi \left(m - \frac{1}{4} \right) \epsilon \right\}^{2/3}, \tag{17}$$

which is nothing but the Airy spectrum see, e.g., Ref. [28]. This equation is quite familiar for the free electron or effective mass approximation [19,29]. To obtain a more common representation of Eq. (17), one has to replace the energy scale β by its equivalent in the continuous limit: $\beta \rightarrow \hbar^2/(2m^*a^2)$, 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

$$|E_{m+1} - E_m| \approx 2\epsilon, \tag{18}$$

with $m \ll 1/(2\epsilon)$ and the energy reference point appropriately defined [26], gives the eigenvalues of matrix (1) with a good accuracy. Thus for energy levels lying above ($E_{m+1} - E_m$ is positive) and below ($E_{m+1} - E_m$ is negative) $E = eV/2$, the level spacing is equal to the *doubled* spacing in the corresponding canonical WSL.

(3) $E - E_{WS}^t \ll 1 (eV > 4)$. In this case, it is easy to see from Eq. (13) that $E_m - E_{WS}^t = 2 - \epsilon(m - 5/4)$, where m is of the order of n^{sls}. This gives the Wannier quantization rule

$$E_{m+1} - E_m = \epsilon, \tag{19}$$

which, however, refers not to the WS-band energy interval, but to the bottom of the upper sls-band having a triangular shape, see Fig. 1. We stress that the validity of Eq. (19) near the edge of WS-band is not at all obvious.

As mentioned above, for a semi-infinite crystal Eq. (17) was derived earlier by different techniques [17–19,29]. At the same time, Eqs. (18) and (19) have never been reported in the given context, at least, to the best of our knowledge.

5. Conclusions

The dependence of the one-electron spectrum on the applied voltage may have reflections in a variety of experiments. The most straightforward manifestation of our results is seen in the field dependence of peaks of the tunneling probability through tilted bands. 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 resonant tunneling. The position of peaks in the transmission spectrum (i.e., the dependence of resonant tunneling probability on energy $T(E)$) can be controlled by the applied voltage as is explained in the above discussion. It can be

stated, therefore, that in the case of sub-WS voltages and weak coupling of the tunnel region (modeled by the tilted band) with the source and drain of electrons, the mid part of transmission spectrum will have peaks at the voltages and energies determined by Eqs. (8) and (9). The shape of these peaks depends on a number of factors. However, their integral intensity is likely to differ little, since all of them correspond to *extended-state* assisted tunneling. In contrast, the peaks of $T(E)$ in the sls-band energy intervals correspond to *localized-state* assisted tunneling. As a result, they will have exponentially decaying intensities the closer are peaks to the tilted band edges. Depending on the part of transmission spectrum, the peak spacing can be of the Airy type (predicted earlier), doubled ϵ, and ϵ spacing, see Eqs. (17)–(19). The method used above is applicable with some modifications to the analysis of the transmission spectrum dependence on the field, energy, and tunneling region thickness, to be discussed elsewhere [30].

The Franz–Keldysh effect reflects the electric-field-induced changes of the band spectrum. The through gap Zener tunneling also associates with field affected electron states. The WSL signatures in these processes, which involve at least two bands, are far more complicated [3,4]. Even relevant to these effects, the zero-approximation model, which is two non-interacting bands influenced by a constant electric field, has not yet received an adequate theoretical treatment. In this context, the results presented are of immediate and future use.

Summarizing, electric field effects on the bulk and surface states spectra are described in one band nearest-neighbor approximation. The new results obtained are addressed, first of all, to thin crystal layers and/or superlattices in a few-tens nanometer range, whose band spectrum contains an empty symmetric band well separated from the others. We predict triple-, double-, and fractional quantization of bulk states at sub-WS voltages, and the existence of a part of surface state spectrum, which has regular doubled Wannier level spacing. These predictions are of importance for reliable experimental identification of electric field effects on the electron spectrum and dynamics.

Acknowledgements

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