Solvable Hamiltonian for superlattice nanowires

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(Received 23 December 2004; published 15 April 2005)

We consider superlattice nanowires, and find an exact solution to the model-independent quantum Hamiltonian. We obtained a closed-form solution to this problem. The energy levels for general interatomic interactions were calculated in the context of the Hubbard model. We obtained an explicit formula for the function whose roots render the energy states. The corresponding energy bands can be tuned by the usual superlattices concept of pattern control but also, in the case of nanowires, by controlling the interatomic separation of the structure. We apply our results to nanowire tunneling diodes, angle-resolved photoemission spectroscopy, and Si-Ge superlattice nanowires.

DOI: 10.1103/PhysRevB.71.165313 PACS number(s): 73.21.Hb, 71.10.−w, 72.15.Nj

I. INTRODUCTION

One of the goals of nanometer-sized electronics is to develop the ability to control atomic level patterning on atomic chains. Beyond their use as connectors, nanowires can be used as active elements if structured appropriately. In particular, all the ideas learned from band control in standard superlattices can be transferred, contextually, to superlattices in nanowires (SN). Although this program is likely to require substantial future research, experimental results demonstrating the feasibility of atomically thick SN on group III-V and group IV elements already exist.[1,2] Other experiments have achieved atomically thin nanowires of carbon,[3] silicon,[4] and organic polymers.[5] A large number of experimental characterizations of these structures rely on scanning tunneling spectroscopy (STS) measurements, which provide information about density of states. In order to interpret those measurements, there is a need to develop specific theories that provide the density of states of the SN. It is the intent of this paper to develop such theory.

The impact of heterostructures in our fundamental understanding of electron systems and in technology is enormous. From a fundamental standpoint, superlattices have served as the physical support for novel behavior. For example, in magnetic-nonmagnetic superlattices, the electrical resistance is found to vary by many orders of magnitude in the presence of a magnetic field.[6] In another example, superlattices of superconductor-insulator layers have shown a crossover from two to three-dimensional melting of the vortex lattice as a function of the insulator layer thickness. From a technological standpoint, heterostrucures (quantum wells, superlattices) have served as optical and electronic detectors and actuators in optoelectronic applications since their inception in the 1970s.

On the other hand, one-dimensional electron systems are being extensively studied to understand the behavior of Luttinger liquids. In contrast to the behavior of electron system in dimensions two and higher whose properties are well established in the context of Landau's theory, the Luttinger liquid does not have stable quasiparticles, and its low energy behavior is based on separated collective spin and charge excitations.[7–9] Although some aspects of the Luttinger liquid theory have been tested in carbon nanotubes,[10] other comparisons, mainly in quantum wires, are open to further study.[11]

In addition, studies of the ground excited states of the Hubbard model have been popular in recent times. The Hubbard model is of interest due to its simplicity. In addition, there is evidence that it accounts for the prominent characteristics of electron systems in a large variety of real materials. In one dimension, these materials include TTF-TCNQ and $SrCuO_2$.[12,13] Luttinger behavior is only observed above a crossover energy range, even in strongly anisotropic materials. However, the Luttinger spectrum can be consistently mapped within the bandwidth onto separated spin and charge excitation bands of the 1D Hubbard model away from half-filling.[14]

With the recent experimental capabilities to create and control nanowires, the number of technological nanowire-based devices has grown tremendously in the area of nanoscale photonics and electronics. For a few years, people have been making "quantum wires" by cleaved-edge overgrowth in GaAs-based (and other) systems.[15] Those systems have been very useful to measure transport properties.[16,17]

More recently quantum wires (and patterns within them) have been grown on nanotubes and atomic chains[18] with applications for waveguides and photovoltaics.[19]

In Sec. II, we derive the equation that provides the energies of the system. In Sec. III, we apply the general result to specific situations. Section IV presents conclusions.

II. ENERGY SPECTRUM

There are various rigorous results regarding density of states for one-dimensional spin systems within the Hubbard model (Lieb-Mattis and Yamanaka-Oshikawa-Affleck theorems, see Ref. 20 for details). However, for the general Hubbard Hamiltonian it has not been possible to find explicit formulas for the energy spectrum. In particular, superlattice-type Hamiltonians have not been considered thus far.

We begin with the Hubbard Hamiltonian[6]

$$H = \sum_{\substack{x,y\in\Lambda \\ \sigma}} t_{xy} c_{x\sigma}^{\dagger} c_{y\sigma} + \sum_{\substack{x\in\Lambda \\ \sigma}} U_x \hat{n}_{x\downarrow} \hat{n}_{x\uparrow}, \tag{1}$$

where $c_x^{\dagger}$ is the creation operator at site x, c_y is the annihilation operator at site y, t_{xy} the hopping parameters, U_x the

on-site parameters, $\hat{n}_x$ is the number operator $c_x^\dagger c_x$, σ represents the spin degree of freedom, and Λ the atomic chain. To produce a general superlattice structure we write the nonvanishing parameters as

$$t_{nn} = \sum_{s=0}^{R-1} \left\{ \gamma_B \sum_{m=s(n_B+n_W)+1}^{s(n_B+n_W)+n_B} \delta_{nm} + \gamma_W \sum_{m=s(n_B+n_W)+n_B+1}^{(s+1)(n_B+n_W)} \delta_{nm} \right\} \tag{2}$$

and

$$t_{n,n+1} = t_{n+1,n} = \sum_{s=0}^{R-1} \left\{ \beta_B \sum_{m=s(n_B+n_W)+1}^{s(n_B+n_W)+n_B-1} \delta_{nm} + \beta_{BW}\delta_{n,s(n_B+n_W)+n_B} + \beta_{BW}\delta_{n,s(n_B+n_W)+n_B+n_W} + \beta_W \sum_{m=s(n_B+n_W)+n_B+1}^{(s+1)(n_B+n_W)-1} \delta_{nm} \right\}, \tag{3}$$

where γ are on site parameters and β are near neighbor interactions. The subindex B labels the barrier regions, the subindex W labels the well region, and the subindex BW accounts for inter barrier-well interactions. The symbol δ_{nm} stands for the standard definition of the Kronecker delta. R is an integer equal to the total number of repetitions of the barrier-well structure, n_B and n_W are the (integer) size of the barrier and well region, respectively.

Call $\Delta_N = \det(H\text{-}E)$ the corresponding eigendeterminant, where E are the sought energies, and $N = R(n_B + n_W)$ is the length of the nanowire.

To simplify the notation, we introduce the new variables

$$\eta_B = \frac{\beta_B}{\beta_{BW}}, \quad \eta_W = \frac{\beta_W}{\beta_{BW}}, \quad \xi_B = \frac{\gamma_B - E}{\beta_{BW}}, \quad \xi_W = \frac{\gamma_W - E}{\beta_{BW}},$$

and $\Delta_N = \beta_{BW}^N D_N$. We will build D_N. Call D_m $(1 \leqslant m \leqslant N)$ any upper-left subdeterminant of $H\text{-}E$. Define d_n $(0 \leqslant n \leqslant n_B)$ a generic D_m when m falls within a barrier, and $\hat{d}_n$ $(0 \leqslant n \leqslant n_W)$ a generic D_m when m falls within a well.

Consider only those determinants $F_j = D_{(n_B+n_W)j}$ and $G_j = D_{(n_B+n_W)j-1}$ $(0 \leqslant j \leqslant R)$. With this notation, our task reduces to finding F_R. These subdeterminants satisfy

$$\begin{cases} d_0 = F_j \\ d_1 = \xi_B F_j - G_j \\ d_n = \xi_B d_{n-1} - \eta_B^2 d_{n-2} \quad 2 \leqslant n \leqslant n_B \end{cases} \quad \text{and} \quad \begin{cases} \hat{d}_0 = d_{n_B} \\ \hat{d}_1 = \xi_W d_{n_B} - d_{n_B-1} \\ \hat{d}_n = \xi_W \hat{d}_{n-1} - \eta_W^2 \hat{d}_{n-2} \quad 2 \leqslant n \leqslant n_W. \end{cases} \tag{4}$$

In each set of equations, the first two equations are initial conditions for the recurrence relations given in the third line.

Noticing that, by definition, $F_{j+1} = \hat{d}_{n_W}$ and $G_{j+1} = \hat{d}_{n_W-1}$, the above relations provide an implicit link between (F_{j+1}, G_{j+1}) and (F_j, G_j). This link can be made explicit noticing that the solutions to the recurrence relations are Chebyshev polynomials.[21] Then,

$$\begin{pmatrix} F_{j+1} \\ G_{j+1} \end{pmatrix} = s\,\eta_B^{n_B}\eta_W^{n_W}\Gamma \begin{pmatrix} F_j \\ G_j \end{pmatrix}, \tag{5}$$

where

$$\Gamma = \begin{pmatrix} \omega_{n_W,n_B} & -\dfrac{\omega_{n_W,n_B-1}}{\eta_B} \\ \dfrac{\omega_{n_W-1,n_B}}{\eta_W} & -\dfrac{\omega_{n_W-1},n_B-1}{\eta_W\eta_B} \end{pmatrix},$$

$$\omega_{n_W,n_B} = \frac{1}{s}\left[U_{n_W}(z_W)U_{n_B}(z_B) - \frac{U_{n_W-1}(z_W)U_{n_B-1}(z_B)}{\eta_W\eta_B} \right],$$

$z_i = \xi_i / 2\eta_i$, $i = W, B, s$ is chosen so that the matrix above is unitary, and $U_n(z)$ is the Chebyshev polynomial of the second kind.[7]

Applying the recurrence above R times, to account for the total length of the system, and using that $F_0 = 1$ and $G_0 = 0$, this choice reproduces correctly the first two matrices as can be checked by inspection,

$$\begin{pmatrix} F_R \\ G_R \end{pmatrix} = (s\,\eta_B^{n_B}\eta_W^{n_W})^R \Gamma^R \begin{pmatrix} 1 \\ 0 \end{pmatrix}. \tag{6}$$

The matrix Γ^R can be written as $\Gamma^R = U_{R-1}(z_R)\Gamma - U_{R-1}(z_R)I$, where I is the identity matrix and

$$z_R \equiv \frac{1}{2}\,\mathrm{Tr}(\Gamma) = \frac{1}{2}\left(\omega_{n_W,n_B} - \frac{\omega_{n_W-1,n_B-1}}{\eta_W\eta_B} \right).$$

Thus, the original eigenvalue equation $\Delta_N = 0$, which is equivalent to $F_R = 0$, reduces to $U_{R-1}(z_R)\Gamma_{11} - U_{R-2}(z_R) = 0$ or, explicitly,

$$U_{R-1}(z_R)\omega_{n_W,n_B} - U_{R-2}(z_R) = 0 \tag{7}$$

which constitutes the main result of this paper.

III. APPLICATIONS

We apply our results to the calculation of properties of three systems: resonance position in a nanowire tunneling

TABLE I. The energy levels are converted into voltage via $V=2$ (E-6 meV). The 6 meV corresponds to the energy of the highest occupied level of the emitter. The factor of 2 corresponds to the fact that in an emitter-dot-collector, one must apply a voltage $V=2E$ to excite a state of energy E in the dot.

Level	Energy (meV)	Voltage (mV)	Experimental voltage (mV)[a]
1	47.6	83.2	80
2	68.3	125	~100
3	184	356	

[a]Reference 8.

diode, angle-resolved photoemission spectroscopy, and Si-Ge superlattice nanowire spectrum. The first calculation will be compared with experiments, the second with other theories, and the third is of predictive character.

A. Nanowire tunneling diode

The Nanometer Consortium has measured tunneling current through InAs-InP nanowire diodes.[22] Their system consisted of an emitter, collector and dot made out of a 19 nm InAs well, and two 9 nm barriers of InP. For the corresponding lattice parameters[23] $a_{\mathrm{InAs}}=6.05$ Å, $a_{\mathrm{InP}}=5.86$ Å, the barriers are 9 nm/0.586 nm=15 atom thick, while the well is 19 nm/0.605 nm=31 atom thick. These are n_B and n_W, respectively.

The Hubbard parameters are found from the corresponding band gaps $\Delta E_{\mathrm{InAs}}=0.42$ eV, $\Delta E_{\mathrm{InP}}=1.37$ eV, grounding the collector (that is the bottom of the band at the collector is set to zero), and the effective masses $m_{\mathrm{InAs}}=0.024$, $m_{\mathrm{InP}}=0.077$. Their values are $\gamma_W=8.70$ eV, $\gamma_B=3.49$ eV, $\beta_W=4.35$ eV, $\beta_B=1.44$ eV. In addition, $n_W=31$, $n_B=15$, and $R=2$. The last value, R, was chosen as 2 to model only one InAs between the two InP barriers.

Table I shows a comparison between our results and experiment. Although the experimental and theoretical voltages are similar, the agreement is not perfect since the energy and lattice parameters correspond to the known values for three-dimensional lattices. For one-dimensional structures, the parameters must be slightly different, but are unknown.

B. Angle-resolved photoemission spectroscopy

We use our results to calculate the angle-resolved photoemission spectrum for a system with $n_W=90$, $n_B=0$, $R=1$, $\gamma_W=4.9\beta_W$ in order to compare with extant numerical calculations performed with those parameters (it is not a superlattice, but an elemental nanowire). Specifically, we compute the photoemission spectral function, which is the imaginary part of the one-particle Green's function

$$A(k,\omega)=\frac{1}{\pi}\,\mathrm{Im}\langle\psi_0|\hat{c}^{+}_{k,\sigma}\frac{1}{H+\omega-E_0-i\eta}\hat{c}_{k,\sigma}|\psi_0\rangle.$$

Figure 1 shows both, the results of our calculations, and those of the previous work.[24] The dashed line corresponds to the numerical results, and the solid line is from this work. We find a good qualitative agreement between the two results. The agreement is not perfect since the Hamiltonian used in the two approaches differ by one term, namely in the numerical case there is an additional total number operator.

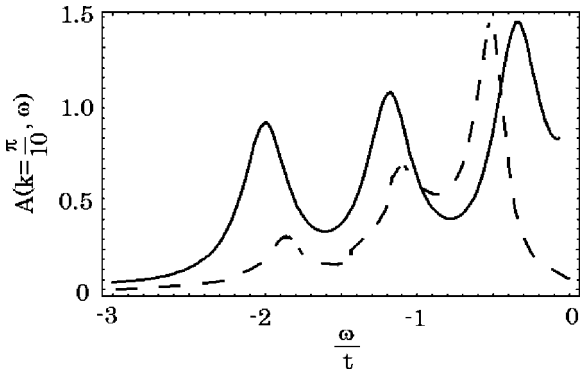

FIG. 1. Spectral function $A(k=\pi/10,\omega)$ for an elemental wire of length 90. In the horizontal axis, $t\equiv\beta_W$.

C. Si-Ge superlattice nanowire

As an illustration of application of the main formula of this paper, we consider a Si-Ge SN with 4 repetitions of a 2-atom long Si and a 5-atom long Ge. The parameters chosen for this example give the correct effective masses and electron affinities for Si and Ge.[25,26] Low-dimensional parameters are likely to differ from those of bulk, and are unknown.[27] The roots of the equation are plotted in Fig. 2 vs SN wave number. Three energy gaps are clearly seen, a consequence of the quasiperiodicity of the system.

For this example, we apply our results to model superlattices to discuss the influence of the parameters on the energy spectrum. This study is motivated by the Cyrot-Lackman theorem that relates the moments of the density of states (mean, width, etc.) with structural trends in molecules.[28,29] Specifically, we allow for β_{BW} to vary, and study its influence on the energy spectrum. This parameter is particularly important since it is the least known in practical applications due to its strong Si-Ge-distance-dependence. We parametrize

$$\beta_{BW}=\beta_{BWO}x$$

with $\beta_{BWO}=1.35$ eV, where x represents the fractional deviation from β_{BWO}.

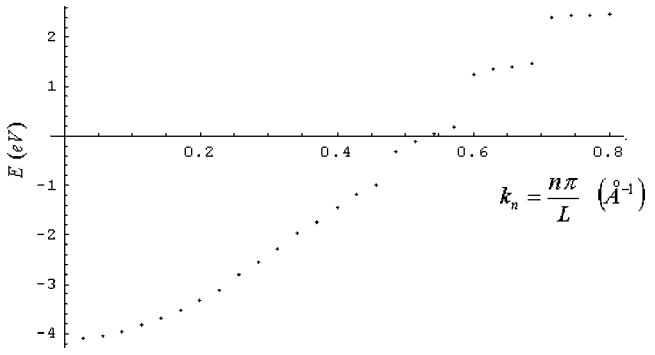

FIG. 2. Dispersion curve. Parameters used, $\gamma_B=-2.09$ eV, $\gamma_W=-0.66$ eV, $\beta_B=0.96$ eV, $\beta_W=1.74$ eV, $\beta_{BW}=1.35$ eV.

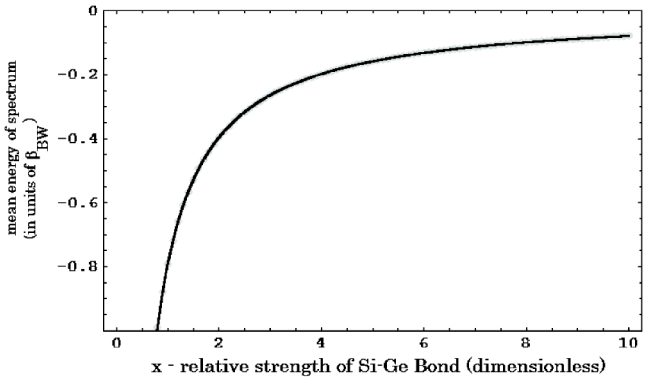

FIG. 3. Mean energy as a function of Si-Ge bond strength.

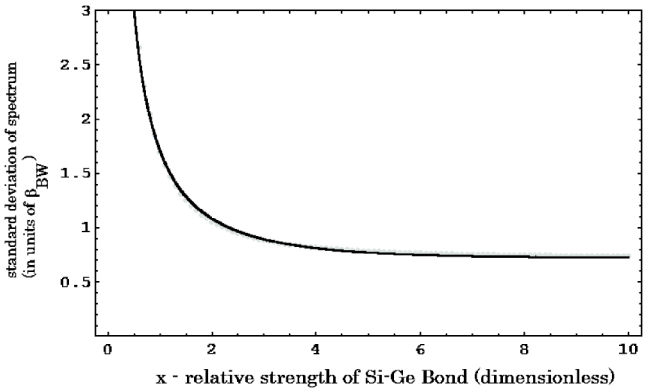

FIG. 4. Width of energy level distribution as a function of Si-Ge bond strength.

Figure 3 shows the mean value of the energy spectrum as a function of x. As x increases, the Hamiltonian becomes increasingly dominated by the Si-Ge bond. In particular, when $x \to \infty$, the Hamiltonian becomes proportional to a real symmetric matrix with null diagonal and ones located every n_B and n_W sites. This matrix has eigenvalues $(\pm 1, 0)$ centered on zero, which explains the asymptotic behavior.

Figure 4 shows the width of the energy spectrum as a function of x. When $x \to \infty$ (Si-Ge dominated Hamiltonian) and for a SN with 4 repetitions of a 2-atom long Si and a 5-atom long Ge, the standard deviation is $\sqrt{14/27} \approx 0.72$, which justifies the asymptotic behavior of the width-x curve.

IV. CONCLUSIONS

We obtained an analytical expression for a function whose roots provide the energy spectrum of a generic SN. That expression contains information of the model-independent Hamiltonian parameters, atomic periodicity of the chain, and the superimposed superlattice periodicity. In calculations involving standard superlattices, it is common practice to substitute atomic potentials for average values of band edges. In nanowires that approximation can typically break down as there might not be enough atoms in a given region to produce an electron band. Our results show a solution that overcomes that problem. We applied our results to three model systems: nanowire tunneling diodes, angle-resolved photoemission spectroscopy, and Si-Ge superlattice nanowires. In the first two cases we compared with extant experimental and theoretical results. In the last case we studied the dependence of DOS moments with the bond parameters in the spirit of Cyrot-Lackman theorem. Finally, our results can serve as the base to interpret STM spectroscopy data from nanowires since it is known that the STM current-voltage curve is a measure of the system's density of states.[30,31]

ACKNOWLEDGMENTS

Work supported by Research Corporation through Grant No. CC5786.

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