

Electronic Structure of One-Dimensional Quasiperiodic Materials of $AB_{1-x}C_x$

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The electronic spectrum of one-dimensional quasiperiodic materials such as $AB_{1-x}C_x$ ($0 \leq x \leq 1$) is calculated herein within the framework of the off-diagonal tight-binding model by using a simple scheme of the theory of the binary quasiperiodic lattices. The spectrum shows a Cantor-set of energy bands and consists of a center band gap for the whole range of x . We find that unless the potential difference is too strong, the magnitude of the center band gap Δ_x is interpolated by a Vegard's law type linear relation with respect to x as $\Delta_x = (1-x)\Delta_{ab} + x\Delta_{ac}$, where Δ_{ab} (Δ_{ac}) is the band gap for the regular AB (AC) alloy. This means that this system is a semiconductor for $0 \leq x \leq 1$. We also show that the center gap follows the Saxon-Hutner-Luttinger theorem.

[Cantor-set, electronic structure, quasiperiodic materials, Saxon-Hutner-Luttinger theorem, tight-binding model, Vegard's law]

§1. Introduction

One-dimensional (1D) disordered systems made by IV-IV, III-V, and II-VI compounds¹⁻⁴⁾ with being represented by a chemical formula $AB_{1-x}C_x$ have been of interest in recent years, since this type of materials became a prototype in semiconductor superlattices such as $Al_{1-x}Ga_xAs$.^{5,6)}

Consider the chemical formula, $AB_{1-x}C_x$, where we consider only the off-diagonal tight-binding Schrödinger equation for later purposes [see eq. (1) with $V_n=0$]. If $x=0$, then it represents a regular AB alloy, where within the above model there are only two types of hopping terms: AB bond (T_a) and BA bond (T_b) (i.e., long and short bonds). If $x=1$, then the system is a regular AC alloy, where similarly as before there are only two types of hopping terms, AC bond (T_a) and CA bond (T_c). Here we have assumed that the AB and AC bonds are identical to one another for the simplicity. And if $0 < x < 1$, then the system is a mixed crystal characterized by a fraction x and the local coordination is either AB or AC , where there are only three types of bonds: AB ($=AC$) bond (T_a), BA bond (T_b), and CA bond (T_c).

Apart from the physical model let us now follow the chemist's point of view. Suppose that there are two types of molecules AB and AC , each of which consists of two electrons filled in the bond. And construct an infinite molecular chain of molecules of AB and AC with a fraction x , getting closer to each other among an infinite number of AB and AC . This allows electrons to hop to nearest neighbor atoms. However, since there are two electrons in each molecule (i.e., one electron per atom), every other bond can be occupied by two electrons (Fig. 1). So, some amount of energy is

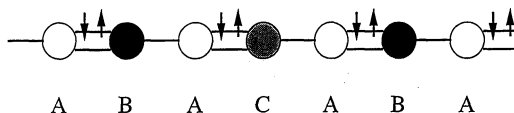


Fig. 1. The 1D quasiperiodic system of $AB_{1-x}C_x$. The local coordination in the system is assumed to be three types of bonds; $AB=AC$, CA , and BA bonds (i.e., long, short, and shorter bonds). The hopping integrals are given by T_a , T_b , and T_c , respectively, and no on-site potential $V_n=0$. To adjust with the three types of bonds we assume $0 < T_a < T_c < T_b$. In this picture the transfer matrices are progressing from the right to the left. If there are two electrons in each molecule (i.e., one electron per atom), every other bond can be occupied by two electrons.

necessary in order to remove an electron to far from the material. This means that a band gap appears at the Fermi energy. In this argument only the local coordination is taken into account and the global character of the system does not matter. Therefore, the system of $AB_{1-x}C_x$ is expected to be either an *insulator* or a *semiconductor*.

On the other hand, let us stand at the point of view of band theory. To know physical properties of these materials and to know whether the system is either metallic, semiconducting, or insulating, it is necessary to obtain the electronic spectrum explicitly. Within the off-diagonal model, if $x=0$ or 1, then there are two energy bands in the spectrum—*valence* and *conduction* bands—that are separated by a gap at the center of the spectrum. However, if x is fractional, then it becomes a very difficult problem in obtaining the spectrum. Because there is no long range order in the system, and therefore, the Bloch theorem in the band theory is not directly applicable. Thus, we would like to know the spectrum for the system of $AB_{1-x}C_x$ for $0 < x < 1$, using the standpoint of the band theory.

The above 1D system was once studied long time ago in order to prove the existence of a gap in the optical band of a disordered mixed crystal.^{4,7} And the spectrum of this type of materials was intensively investigated⁸ in relation to the *Saxon-Hutner-Luttinger (SHL) theorem*.^{9,10} However, to the best of our knowledge the spectrum for the system of $AB_{1-x}C_x$ has never been shown explicitly as globally changing the value of the fraction x for the whole range of $0 < x < 1$.

In this paper we are going to relate the above two viewpoints within the framework of the one-dimensional tight-binding Schrödinger equation with the nearest neighbor hopping terms and to show the energy spectrum changing the value of x , by using the theory of the *binary quasiperiodic lattices (BQPLs)*.^{11,12} Second, we shall discuss the physics behind the calculation such as the difference between the spectra of the $A_{1-x}B_x$ and the $AB_{1-x}C_x$ systems. Third, we clarify the relation between the theory of the BQPLs and the SHL theorem.^{4,8-10}

§2. Physical Model

We would like to solve the discrete Schrödinger equation for an electron:

$$T_{n+1}\psi_{n+1} + T_n\psi_{n-1} + V_n\psi_n = E\psi_n, \quad (1)$$

where T_n is the hopping integral between the n th and the $(n-1)$ th sites and V_n the on-site potential at site n , respectively.⁸ We convert the above equation into the transfer matrix form:

$$\Psi_{n+1} = \underline{T}(n)\Psi_n, \quad (2)$$

where the transfer matrix $\underline{T}(n)$ and the function Ψ_n are given by

$$\underline{T}(n) \equiv \begin{pmatrix} \frac{E - V_n}{T_{n+1}} & -\frac{T_n}{T_{n+1}} \\ 1 & 0 \end{pmatrix}$$

and

$$\Psi_n \equiv \begin{pmatrix} \psi_n \\ \psi_{n-1} \end{pmatrix}, \quad (3)$$

respectively.

We consider only the off-diagonal model with $V_n=0$, throughout this paper. We assume that those constants take on only three values associated with the three types of bonds [$AB(=AC)$, BA , and CA bonds] in the chain, as is discussed in the introduction. To distinguish these three bonds we assume $0 < T_a < T_c < T_b$ (Fig. 1). This is consistent with the physical situation that if the bond between two adjacent atoms is longer, the overlapping integral is smaller and so is the hopping integral.

§3. Basic Strategy

The basic idea of the present paper is the following. In the standard point of view, if a fractional value of x is taken for $AB_{1-x}C_x$, then one usually assumes that *the system is disordered or amorphous*, i.e., *random*.¹³ This assumption makes us treat the *ensemble average* over randomness in order to calculate the physical properties of the material such as in the theory of *Anderson localization*.¹⁴

This type of theory has been intensively studied numerically in higher dimensional systems

as well as in 1D one that we would like to work out in the present paper. The former is now well-known by the celebrated names of the *virtual crystal approximation (VCA)*^{2,4)} and the *coherent potential approximation (CPA)*.⁴⁾ The latter is a rigorous approach known as the *Dyson-Schmidt method*,¹⁵⁾ although the Dyson-Schmidt equation has never yet been solved analytically except numerical efforts,^{2,4)} however.

The main reason for the use of the assumption seems only because there has been no other method without using it as the starting point. However, physically speaking, it is not necessary so. But there exists another method that seems worth investigating.

Until recently, the concepts of *quasicrystals* have been fully developed¹⁶⁾ as well as the 1D modeling such as the BQPLs and others.^{11,12)} So, we may have another choice for the assumption that *the system is QP*. But it does not mean that we are using the general and broad concepts of the quasiperiodic systems. Rather, we restrict ourselves to the concept of the BQPLs within the framework discussed in the articles,^{11,12)} since we would like to investigate only the 1D model with this particular quasiperiodic structure. Thus, the theory of

the BQPLs can be the main clue for our purpose here.

In a BQPL that we are investigating, there is no translational symmetry, i.e., no long-range order in the usual sense, which means that the lattice structure is not periodic but nonperiodic with the two-distance property.^{16,17)} But it has a unique structure once x is fixed. Therefore, there is no use of the ensemble average over randomness. Thus, the BQPLs are classified in between periodic and disordered lattices.

Since there are about 10^{23} atoms in a 3D physical system, there is a sequence of about 10^7 ($\equiv N_L$) layers in one direction.^{5,6)} The density of Q 's (i.e., x) is usually some fraction with about three or four accuracy in experimental setting.¹⁻⁶⁾ So, if the total number of digits in x is less than N_L , we may expect that there are very many repetitions of the unit cell whose size is that of the digits. Thus, unless the number of digits is in the order of N_L , we may approximate the system to be periodic with using the rational approximation of x . This situation makes our 1D modeling of the BQPLs work well.

In Fig. 2, the model systems are schematically illustrated from the above two points of

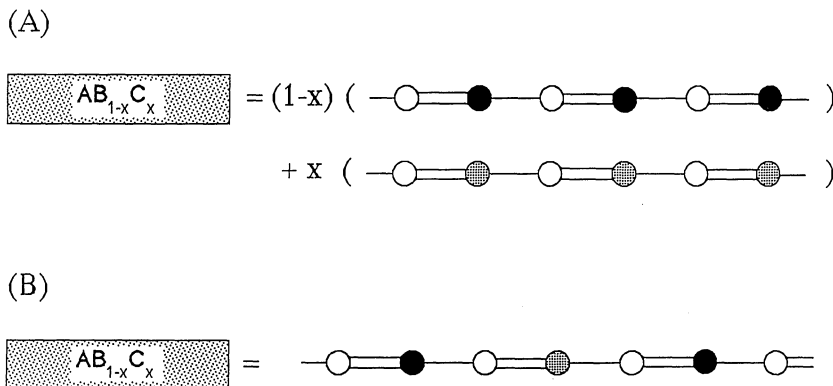


Fig. 2. The concepts of the models. (A) The concept of the VCA and the CPA: The system of $AB_{1-x}C_x$ ($0 \leq x \leq 1$) is approximated as an average over the two independent systems of the regular AB and AC alloys. To realize this situation, in the VCA the hopping integral T_n is averaged as $T_n(x) = (1-x)T_n(AB) + xT_n(AC)$, where $T_n(AB)$ [$T_n(AC)$] stands for the hopping integral of the regular AB (AC) alloy. In the CPA, the hopping integral T_n is first regarded as an external variable, and finally it is ensemble averaged over the randomness as a random variable. The same for the on-site potential, V_n . (B) The concept of the BQPLs: The system of $AB_{1-x}C_x$ ($0 \leq x \leq 1$) is approximated by a rational number x , which is the density of the unit AC in the system. To realize this situation, the hopping integral T_n (the potential V_n) is defined as a QP hopping integral (potential). Both models satisfy the same Vegard's law: $a_x \equiv (1-x)a_{AB} + xa_{AC}$ but its origin is very different.

view of the VCA (or the CPA) and the BQPL. This elucidates the difference between the two concepts.

§4. Binary Quasiperiodic Lattices

The theory of the BQPLs^{11,12)} can be summarized as follows. Consider a 1D BQPL system represented by a chemical formula $P_{1-x}Q_x$ ($0 \leq x \leq 1$). If we regard $P(Q)$ as $P \equiv A$ ($Q \equiv B$), then this expression describes the usual BQPL system of $A_{1-x}B_x$.^{11,12)} And if we regard $P(Q)$ as a molecule $P \equiv AB$ ($Q \equiv AC$), then it turns out to be the system of $AB_{1-x}C_x$ under investigation.

Let x be $x \equiv 1/(1 + \lambda_k)$. The approximant λ_k is defined as the ratio between the total numbers of P 's and Q 's in the unit cell. The continued fraction expansion of λ_k is given by

$$\lambda_k \equiv \frac{P_k}{Q_k} = n_0 + \frac{1}{n_1 + \frac{1}{n_2 + \cdots + \frac{1}{n_{k-1}}}}, \quad (4)$$

where $n_0, n_1, n_2, \dots, n_{k-1}$ are all positive integers, and P_k and Q_k are given by

$$P_{k+1} = n_{k-1}P_k + P_{k-1}, \quad Q_{k+1} = n_{k-1}Q_k + Q_{k-1} \quad (5)$$

with the initial conditions $(P_0, Q_0) = (0, 1)$ and $(P_1, Q_1) = (1, 0)$.

The array of these integers is crucially important. Because it is related to the scaling transformation for the lattice. A QP sequence is defined in terms of two symbols, P and Q . Let us denote by $L_k \equiv L_{P_k, Q_k}(P, Q)$ the unit cell of the k th generation of the sequence, which consists of P_k P 's and Q_k Q 's such that the total number N_k is given by $N_k \equiv P_k + Q_k$ with $N_1 = N_0 = 1$. If $0 \leq x \leq 1/2$ (i.e., $1 \leq \lambda_k$), then it

is constructed by a series of the scaling transformations:

$$(L_{k+1}, L_k) = S_{n_{k-1}} S_{n_{k-2}} \cdots S_{n_0}(P, Q), \quad (6)$$

and if $1/2 < x \leq 1$ (i.e., $0 \leq \lambda_k < 1$), then the role of P 's and Q 's is switched and do the same thing. Here we have started from the seed of a sequence, (P, Q) , and the scaling transformation at the k th step is given by

$$S_{n_k}(P, Q) = (QP^{n_k}, P). \quad (7)$$

In other words, we can define the sequence recursively such as

$$L_{k+1} = [L_k]^{n_{k-1}} L_{k-1} \quad (8)$$

with $L_0 = "Q"$ and $L_1 = "P"$. From this, the structure of the unit cell is explicitly given by

$$L_k \equiv (P) Q P^{d_{Qk}} Q P^{d_{Qk-1}} \cdots Q P^{d_1} \quad (9)$$

for $k = \text{even}$ (odd), where $d_n \equiv [n\lambda_k] - [(n-1)\lambda_k]$ and $[]$ denotes the Gauss symbol that takes the largest integer part in the square bracket. d_n takes only two values; either $[\lambda_k]$ or $[\lambda_k] + 1$, and is periodic with $d_{n+Q_k} = d_n$. If $k \rightarrow \infty$, then d_n becomes non periodic so that L_k defines an exact QP sequence.¹⁷⁾ If we assume the lattice constant of $P(Q)$ to be a_P (a_Q), then the total length of the unit cell is given by $|L_k| \equiv P_k a_P + Q_k a_Q$. Thus, we are naturally led to define the mean lattice constant $a_x \equiv |L_k| / N_k$, and it is exactly given by the famous *Vegard's law*:^{2,6)}

$$a_x \equiv (1-x)a_P + xa_Q. \quad (10)$$

Let us regard L_k as a matrix product of the N_k transfer matrices of P and Q . Let the initial triple of the traces be $(\bar{X}_0, Y_0, \bar{Z}_0) \equiv (\frac{1}{2} \text{Tr}(\underline{Q}), \frac{1}{2} \text{Tr}(\underline{P}), \frac{1}{2} \text{Tr}(\underline{PQ}))$. We have the generic trace map according to the series of scaling transformations:

$$(X_k, Y_k, Z_k) = T_{n_{k-1}} T_{n_{k-2}} \cdots T_{n_0}(X_0, Y_0, Z_0), \quad (11)$$

$$T_n(X, Y, Z) \equiv (Y, C_{n-1}(Y)Z - C_{n-2}(Y)X, C_n(Y)Z - C_{n-1}(Y)X) \quad (12)$$

where $C_n(Y)$ is the n th Chebyshev polynomial of the second kind, being defined by the recursion relation $C_{n+1}(Y) = 2YC_n(Y) - C_{n-1}(Y)$ with $C_{-1}(Y) = 0$, $C_0(Y) = 1$. We note that the trace map preserves an invariant surface:

$$I = X^2 + Y^2 + Z^2 - 2XYZ - 1 \quad (13)$$

at each step of the scaling transformation.

The above trace map system has been successfully applied to obtain the electronic spectrum on the 1D BQPLs¹¹⁾ and the optical property of BQP multilayers.¹²⁾

§5. Application to the 1D Ternary Quasiperiodic System of $AB_{1-x}C_x$

We now are able to apply the above method to the system of $AB_{1-x}C_x$ ($0 \leq x \leq 1$). In this case, we introduce the following initial transfer matrices of the off-diagonal model for the electronic problem:

$$\underline{P} \equiv \underline{AB} = \begin{pmatrix} \frac{E^2}{T_a T_b} - \frac{T_b}{T_a} & -\frac{E}{T_b} \\ E & -\frac{T_a}{T_b} \end{pmatrix} \quad \text{and} \quad \underline{Q} \equiv \underline{AC} = \begin{pmatrix} \frac{E^2}{T_c T_a} - \frac{T_c}{T_a} & -\frac{E}{T_c} \\ E & -\frac{T_a}{T_c} \end{pmatrix}. \quad (14)$$

From these we get the corresponding initial traces as

$$X_0 = \frac{1}{2} \text{Tr}(\underline{Q}) = \frac{1}{2} \left(\frac{E^2}{T_a T_c} - \frac{T_a}{T_c} - \frac{T_c}{T_a} \right), \quad (15a)$$

$$Y_0 = \frac{1}{2} \text{Tr}(\underline{P}) = \frac{1}{2} \left(\frac{E^2}{T_a T_b} - \frac{T_a}{T_b} - \frac{T_b}{T_a} \right), \quad (15b)$$

$$Z_0 = \frac{1}{2} \text{Tr}(\underline{PQ}) = \frac{1}{2} \left(\frac{E^4 - (2T_a^2 + T_b^2 + T_c^2)E^2}{T_a^2 T_b T_c} + \frac{T_a^2}{T_b T_c} + \frac{T_b T_c}{T_a^2} \right). \quad (15c)$$

Substitute into the invariant of eq. (13) and after a straightforward but tedious calculation we obtain

$$I = \cosh^2(u) + \cosh^2(v) + \cosh^2(u+v) - 2 \cosh(u) \cosh(v) \cosh(u+v) - 1 = 0, \quad (16)$$

where

$$\cosh(u) \equiv (T_a/T_b + T_b/T_a)/2 \quad \text{and} \quad \cosh(v) \equiv (T_a/T_c + T_c/T_a)/2. \quad (17)$$

This vanishing invariant usually means that there is no quasiperiodicity in the system such that the system is trivial.^{11,12,18)} However, this is not the case in our problem. No matter what values of T_a , T_b , and T_c are set, the invariant is always zero because of the identity in eq. (16).¹⁹⁾ It rather means that unless $T_a = T_b = T_c$ the energy level at $E=0$ always belongs to a band gap, while the energy level at $E=0$ belongs to the six-cycle in the usual setting of two types of atoms (i.e., $A_{1-x}B_x$), where $I = (T_a/T_b - T_b/T_a)^2/4$.^{11,12,18)} This implies the validity of the SHL theorem in our problem, which will be discussed later. Thus, this situation is essentially new in the theory of the BQPLs.

We now give a simple scheme to obtain the energy bands with changing the value x : Fix a value of x . Do the scaling transformation. And obtain the trace map accordingly. If $|X_k| \leq 1$, then the energy is allowed, and if

$|X_k| > 1$, then the energy is forbidden.

In Fig. 3 we show the energy bands with respect to x , where x is approximated as $x = n/702$ ($1 \leq n \leq 701$). In Figs. 3(a) and 3(b) the hopping integrals are taken as $(T_a, T_b, T_c) = (1, 1.4, 1.2)$ and $(1, 3, 2)$, respectively. These figures show that the spectrum looks like a Cantor set. This is very different from that of the VCA and the CPA,^{2,4)} where only one or two continuous bands exist in the spectrum. And the SHL theorem^{9,10)} holds for the center gap in our model.

§6. Discussions

Now, we are going to discuss the physics behind the calculation. First, it is interesting to compare the spectra of the $A_{1-x}B_x$ ^{11,12)} and the $AB_{1-x}C_x$ systems. There is a symmetry around $E=0$ in the spectrum, since the wave function satisfies the condition $\psi_n(E) = (-1)^n \psi_n(-E)$ for the off-diagonal model. Therefore, if N_k is

even, there is a gap at $E=0$. Otherwise, the state of $E=0$ lies in a center band. So, the existence of the center gap is just a reflection of the evenness of N_k in our 1D model. Since in the former system N_k can be either even or odd in the development as $k \rightarrow \infty$, the system can be either a metal, a semiconductor, or an insulator. On the other hand, in the latter system N_k is always even, since the primitive units are AB and AC . Therefore, it must be either a semiconductor or an insulator. In this way, these two systems have to show the very different physical characters. We shall show just below that the latter is related to the SHL theorem.^{9,10)}

Second, we consider the relationship between our theory and the SHL theorem. For

the regular AC alloy case, from eq. (15a) we have $X_0 = \text{Tr}(\underline{Q})/2 = [E^2/(T_a T_c) - T_a/T_c - T_c/T_a]/2$. Since if $|X_0| > 1$, the energy lies in a band gap, this gives the band gap $\Delta_{ac} \equiv 2|T_a - T_c|$. Similarly, from eq. (15b) we obtain the band gap $\Delta_{ab} \equiv 2|T_a - T_b|$ for the regular AB alloy. For the next generation of the regular $ABAC$ alloy, from eq. (15c) we have $Z_0 \equiv \text{Tr}(\underline{PQ})/2 = 2X_0 Y_0 - Z'_0$, where $Z'_0 \equiv \text{Tr}(\underline{P}^{-1}\underline{Q})/2 = (T_b/T_c + T_c/T_b)/2$. Consider an energy such that $|X_0| > 1$ and $|Y_0| > 1$. Then this energy lies in the common band gap defined by $\Delta_0 \equiv \min[\Delta_{ab}, \Delta_{ac}]$. In this case, since $|Z'_0| > 1$ unless $T_b = T_c$, we get $|Z_0| > 1$ as well. This is due to the following fact: There is an identity, $\text{Tr}(\underline{PQ}) + \text{Tr}(\underline{P}^{-1}\underline{Q}) = \text{Tr}(\underline{P})\text{Tr}(\underline{Q})$. Suppose $\text{Tr}(\underline{P})\text{Tr}(\underline{Q}) > 0$.

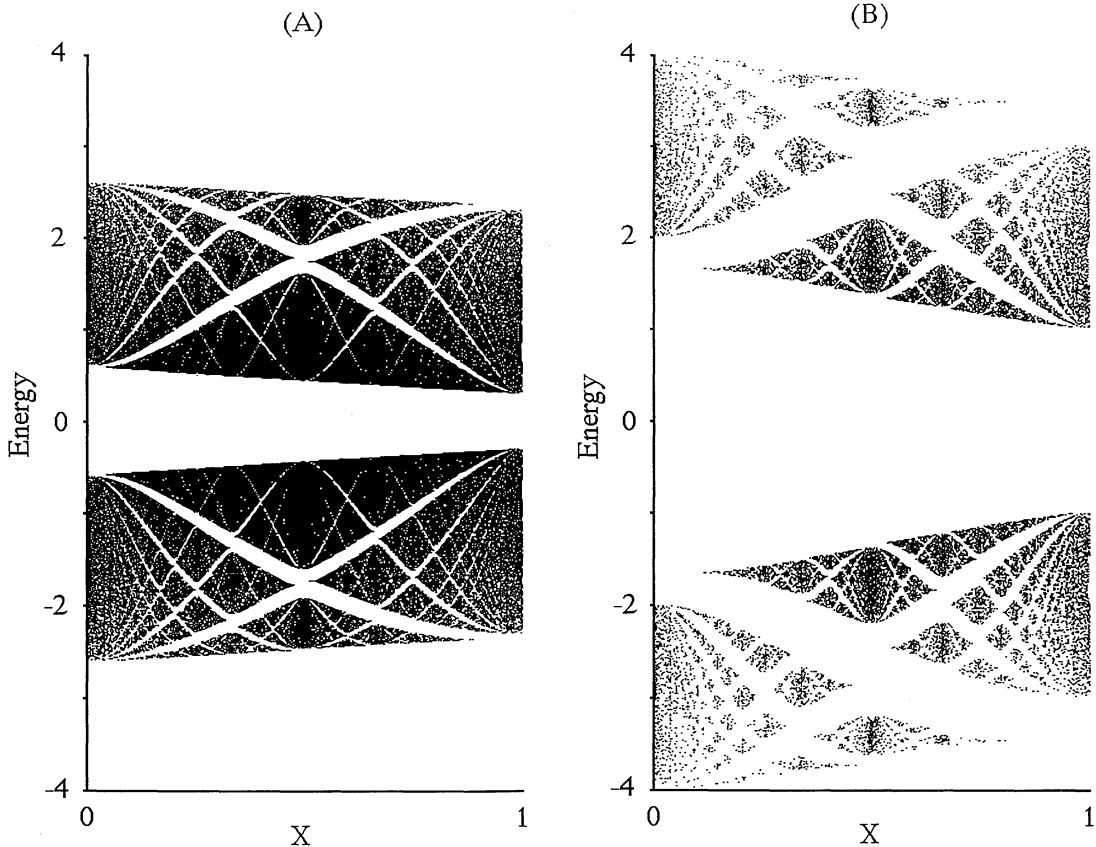


Fig. 3. The electronic energy spectrum of $AB_{1-x}C_x$ with x running from 0 to 1. The hopping integrals are taken as (A) $(T_a, T_b, T_c) = (1, 1.4, 1.2)$ and (B) $(T_a, T_b, T_c) = (1, 3, 2)$. In (A), we see that the center gap follows the SHL theorem and is fitted by a linear law, $\Delta_x = (1-x)\Delta_{ab} + x\Delta_{ac}$. But the other gaps are not the case. In (B), we see that the center gap behaves nonlinearly with respect to x when the differences in the hopping integrals become stronger.

If $|\text{Tr}(\overline{PQ})| > |\text{Tr}(\underline{P}^{-1}\underline{Q})|$, then $\text{Tr}(\overline{PQ}) = |\text{Tr}(\overline{PQ})|$ and if $|\text{Tr}(\underline{P}^{-1}\underline{Q})| > |\text{Tr}(\overline{PQ})|$, then $\text{Tr}(\underline{P}^{-1}\underline{Q}) = |\text{Tr}(\underline{P}^{-1}\underline{Q})|$. Since $Z_0 = \text{Tr}(\overline{PQ})/2$ and $Z'_0 = \text{Tr}(\underline{P}^{-1}\underline{Q})/2$, then from the above fact we can conclude $|Z_0| > |Z'_0| > 1$. Therefore, the common band gap in the spectra for the regular alloys remains as a gap for the next generation as well. In this way, by recursively using the above idea in the generic trace map the common gap remains forever as a gap for higher generations. This is the reason why we observe the validity of the SHL theorem in the calculation as in Fig. 3.

We would like to remark that the theory of Dworin^{8,20} on the 1D disordered lattices is very similar to that of the BQPLs^{11,12} in the above sense. We believe that this is the first time to argue and point out the relationship between the theory of the BQPLs and the SHL theorem in the theory of the disordered lattices.

We next consider very interesting aspects of the result. In Fig. 3 we easily observe that the magnitude of the center gap is interpolated with respect to x by a simple linear or additive form:

$$\Delta_x = (1-x)\Delta_{ab} + x\Delta_{ac}. \quad (18)$$

This is another Vegard's law type plot in experiment,¹⁻⁶ which is usually thought of as an empirical law. And this result is very similar to that from the VCA^{2,4} and the CPA.⁴ This seems very remarkable since we obtain such a result without using the ensemble average over the randomness, which is always taken into account in the VCA and the CPA. However, in our model this holds only when the differences in the hopping integrals are weak enough. If they become stronger, then there appears non-linearity with respect to x . This is seen in Fig. 3. If we look at other gaps different from the center gap, which do not seem to follow the SHL theorem, then the magnitude of the gaps cannot be represented by the simple linear form but it nonlinearly depends upon x . The nonlinearity is most seen in the gaps. In this way, the appearance of the linearity and the nonlinearity in the behavior of the gaps with respect to x is a very important aspect in this research. So, it is very interesting to investigate this point in experiment.

§7. Conclusion

In conclusion, we have shown the electronic spectrum for the 1D system of $AB_{1-x}C_x$ using the theory of the BQPLs.^{11,12} We have found numerically and proved analytically the connection between the theory of the BQPLs and the SHL theorem. We would like to point out that the present theory is straightforwardly applicable to the model of the layered materials in 3D.²¹ And its generalizations²² are applicable to the more complicated 1D systems such as the ternary alloys of $A_xB_yC_z$ and the quaternary alloys of $AB_xC_yD_z$, etc.^{5,6} These points will be very interesting for further research.

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