Resonant scattering in random-polymer chains with inversely symmetric impurities

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In the frame of the tight-binding approximation, we study the electronic transport and delocalization in one-dimensional random-polymer chains, where the impurity clusters are randomly distributed in the host monatomic chain and the impurity atoms possess inverse symmetry in each cluster. As an example, the asymmetric trimer case is presented first. The resonant energy is obtained analytically, which has been demonstrated by the numerical calculation on the electronic transmission. According to the zero Lyapunov exponent at the resonant energy, we confirm that electronic delocalization, indeed, exists in the model. Moreover, the undecayed electronic wave functions are shown near the resonant energy. Thereafter, we generalize the asymmetric trimer model to a polymer one with inversely symmetric impurities. The electronic delocalization and resonant scattering have been found in this generalized polymer model. The result is beyond the common viewpoint that the internal mirror symmetry in the defects is the necessary condition for the presence of delocaliztion in such correlated systems. The polymer model presented here may provide a way to make different types of polymers with high electronic conductivity.

I. INTRODUCTION

In 1958, Anderson presented the electronic localization theorem based on the model with site-diagonal disorder.[1] The exponentially localized eigenstates in one-dimensional (1D) disordered systems have been demonstrated by the exponential decay of the electronic transmission coefficients.[2,3] In the last decade, many experimental and theoretical studies have provided deeper insights into the problem of electronic localization. It is found that extended states can exist in the system with correlated disorder. One typical case is the random-dimer model (RDM) introduced by Dunlap, Wu, and Phillips in 1990.[4] In the RDM, one of the two site energies ϵ_a and ϵ_b is randomly assigned to pairs in the lattice, and an invariable nearest-hopping integral V connects the sites. Provided that $|\epsilon_a - \epsilon_b| \lesssim 2V$, there are $\sqrt{N}$ extended electronic states in the RDM with length N; otherwise localization occurs. Recently, much attention has been paid to the random-dimer model. For example, the $\sqrt{N}$ rule of extended states has been extensively discussed by the perturbation calculation on the density of states, the Lyapunov exponent, the Laudauer theory on the conductance, and also the electronic wave function.[5–8] The experimental evidence of the delocalization has been found in the random-dimer semiconductor supperlattice,[9] and the RDM has been successfully applied to explain the high conductivity of some polymers.[10,11] In 1993, Phillips proposed an ultimate generalization of the RDM, i.e., the twined disordered system.[12] Up to now, the RDM has been generalized to the random-trimer, random-dimer-trimer, and even the random-polymer model.[13–15] The widely accepted understanding of the electronic delocalization in RDM-like systems is that the impurity possesses internal mirror symmetry, and this short-range correlated disorder can make the localization length comparable to the length of the system at some specific energies, which are called ''resonant energies.''[4,10,11] However, the investigation on the ''asymmetric dimer'' has shown that the internal mirror symmetry is not a necessary condition for the presence of the electronic delocalization in

RDM,[16] instead, the short-range spatial correlation plays an important role in such kind of systems.[17–20]

In this paper, the electronic resonant scattering is investigated in the 1D random-polymer chain, where the internal structure of the impurity cluster is considered to possess an inverse symmetry. It is found that electronic delocalization can indeed take place in this system. This paper is organized as follows. In Sec. II, we analytically discuss the resonant energy of a host monatomic chain inserted by a single asymmetric trimer (AST). In Sec. III, the transmission coefficient of the AST model is presented numerically. In Sec. IV, the electronic delocalization in the AST model is discussed based on the Lyapunov exponent and the electronic wavefunction behavior. In Sec. V, we generalize the AST model to the random-polymer chain with impurities possessing an inverse symmetry, i.e., random antisymmetric polymer (RAS polymer), and we rigorously prove the presence of electronic delocalization in this generalized model. Finally, a summary is given in Sec. VI.

II. THE THEORETICAL ANALYSIS

Under the tight-binding approximation, the Schrödinger equation for a one-dimensional chain is

$$(E-\epsilon_j)C_j - V_{j,j+1}C_{j+1} - V_{j,j-1}C_{j-1}=0, \tag{1}$$

where ϵ_j is the site energy, C_j is the amplitude of the electronic wave function on the jth site, and $V_{j,j\pm1}$ is the nearest-hopping integral. We restrict ourselves to the on-site model, that is, $V_{j,j\pm1}$ is taken as the same constant V, whereas ϵ_j depends on the atom that occupies the jth site. It is assumed that the host chain is only composed of atom a. From Eq. (1), energy E of the monatomic host chain satisfies

$$E-\epsilon_a=2V\cos k, \tag{2}$$

where k is the wave vector. That is, the allowed energy must locate in the range $[-2V+\epsilon_a, 2V+\epsilon_a]$.

Equation (1) is easily rewritten in the matrix form

$$\begin{pmatrix} C_{j+1} \\ C_j \end{pmatrix} = \bar{M}_j \begin{pmatrix} C_j \\ C_{j-1} \end{pmatrix}, \qquad (3)$$

with

$$\bar{M}_j = \begin{pmatrix} (E-\epsilon_j)/V & -1 \\ 1 & 0 \end{pmatrix}, \qquad (4)$$

where $\bar{M}_j$ is the transfer matrix that correlates the adjacent site amplitudes C_j and $C_{j\pm1}$.

Now we consider the electronic transmission in the case that only a single impurity is inserted into the host monatomic chain. Provided that a defect occupies the sites from $n+1$ to $n+m$, the amplitude of the Bloch wave with a specific wave vector k at both sides of the defect follows as

$$C_j = \begin{cases} e^{ikj} + r e^{-ikj} & (\text{if } j \leqslant n+1) \\ t e^{ikj} & (\text{if } j \geqslant n+m). \end{cases} \qquad (5)$$

Here, r and t are the reflection and transmission amplitudes, respectively. The correlation between the wave amplitudes at both ends of the defect can be written in the matrix form

$$\begin{pmatrix} C_{n+m} \\ C_{n+m-1} \end{pmatrix} = \bar{P}_m \begin{pmatrix} C_n \\ C_{n-1} \end{pmatrix}, \qquad (6)$$

where $\bar{P}_m$ is the total transfer matrix across the impurity. It satisfies

$$\bar{P}_m = \prod_{j=n}^{n+m-1} \bar{M}_j. \qquad (7)$$

For a given $\bar{P}_m$, the reflection amplitude r can be written as[11]

$$r = -\xi^{2n} \frac{\alpha^T \Gamma \bar{P}_m \alpha}{\alpha^T \Gamma \bar{P}_m \alpha^*}, \qquad (8)$$

where $\xi = e^{ik}$, $\Gamma = \begin{pmatrix} 0 & 1 \\ -1 & 0 \end{pmatrix}$, $\alpha = \begin{pmatrix} \xi \\ 1 \end{pmatrix}$, and α^T is the transpose of α.

In the AST model, the impurity has the form as $\epsilon_b \epsilon_a \epsilon_c$. Note that the middle atom of the defect is the element of the host chain. The total transfer matrix across a single AST $\epsilon_b \epsilon_a \epsilon_c$ is

$$\begin{aligned} \bar{P}_m &= \begin{pmatrix} C & -1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} A & -1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} B & -1 \\ 1 & 0 \end{pmatrix} \\ &= \begin{pmatrix} ABC-B-C & -AC+1 \\ AB-1 & -A \end{pmatrix} \equiv \begin{pmatrix} u & v \\ w & x \end{pmatrix}, \end{aligned} \qquad (9)$$

where $A=(E-\epsilon_a)/V$, $B=(E-\epsilon_b)/V$, and $C=(E-\epsilon_c)/V$. Thus, the reflection coefficient can be given by

$$R = |r|^2 = \frac{-4vw\cos^2 k + 2(u-x)(v-w)\cos k + (u-x)^2 + (v+w)^2}{-4ux\cos^2 k + 2(u-x)(v-w)\cos k + (u+x)^2 + (v-w)^2}. \qquad (10)$$

It is not difficult to deduce that when

$$\epsilon_a = \frac{\epsilon_b + \epsilon_c}{2} \qquad (11)$$

and

$$E = \epsilon_a, \qquad (12)$$

we can obtain $R=0$. Therefore, Eqs. (11) and (12) are the necessary conditions for the unit transmission coefficient in the case of the AST model. It is important to note that Eqs. (11) and (12) are independent of the hopping integral V. Moreover, if a definite concentration of such asymmetric trimers $\epsilon_b \epsilon_a \epsilon_c$ has been added into the host chain, a remarkable property is that the resonant energy always exists at $E = \epsilon_a$ for random values of ϵ_b and ϵ_c in each trimer $\epsilon_b \epsilon_a \epsilon_c$, as long as Eq. (11) is satisfied. For example, if $\epsilon_a = 1.0$, we can choose $\epsilon_b = 0.5$ and $\epsilon_c = 1.5$ for the first defect, then choose $\epsilon_b = -0.2$ and $\epsilon_c = 2.2$ for the second defect, . . . , and so on. If Eq. (11) is always observed in each defect, the reflection coefficient certainly vanishes at $E = \epsilon_a = 1.0$.

III. THE NUMERICAL CALCULATION

If a series of impurities is randomly inserted into the ordered host chain, the electronic-transport property can be studied by the numerical calculation of the transmission (or reflection) coefficient. Generally, the sample chain of length N is embedded in an infinite monatomic chain composed of atom a.[21] Then, in the conduction region to the left and right of the sample chain, the normalized wave functions can be written as

$$C_j = \begin{cases} e^{ikj} + f_r e^{-ikj} & (\text{when } -\infty < j \leqslant 1) \\ f_t e^{ikj} & (\text{when } N+1 \leqslant j < \infty). \end{cases} \qquad (13)$$

Here, we define a new transfer matrix $\bar{Q}(N)$ by

$$\begin{pmatrix} f_t \\ if_t \end{pmatrix} = \bar{Q}(N) \begin{pmatrix} 1+f_r \\ i(1-f_r) \end{pmatrix}, \qquad (14)$$

where
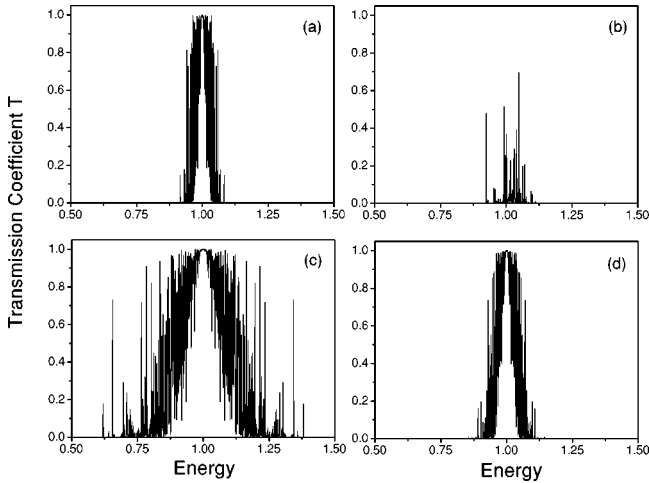
$$\bar{Q}(N) = \sigma^{-1} \bar{S}(N)\sigma, \qquad (15)$$

FIG. 1. The transmission coefficient T as a function of energy for the AST $\epsilon_b\epsilon_a\epsilon_c$ with $N=12\,000$. (a) $\epsilon_a=1.0$, $\epsilon_b=0.0$, $\epsilon_c=2.0$, and $V=1.0$. Around $E=1.0$, there is a symmetric peak in the transmission spectrum. (b) Electronic transmission greatly decreases if $\epsilon_b=0.1$, while other parameters are the same as those in (a). (c) The width of the peak increases when $V=2.0$ with other parameters are the same as in (a). (d) $\epsilon_a=1.0$, $V=1.0$, ϵ_b and ϵ_c randomly distribute in the range $[0,2]$, respectively. Note that Eq. (11) is always satisfied in each trimer.

$$\sigma=\begin{pmatrix} \cos k & \sin k \\ 1 & 0 \end{pmatrix}, \qquad (16)$$

and $\bar{S}(N)$ is the total transfer matrix across the sample chain of length N, i.e., $\bar{S}(N)=\Pi_{j=1}^{N}\bar{M}_j$. Because $\bar{Q}(N)$ is unimodular, the transmission coefficient satisfies[21]

$$T=\frac{4}{2+\sum\limits_{i,j=1}^{2}|\bar{Q}(N)_{i,j}|^2}. \qquad (17)$$

Based on Eqs. (15)–(17), the transmission coefficient as a function of energy can be calculated numerically. We take the length of the sample chain $N=12\,000$ and the concentration of defects $p=0.5$ (the strength of disorder is maximum under this condition) in all following calculations. The transmission coefficients of the AST model are shown in Fig. 1. In Fig. 1(a), an obvious transmission peak occurs at $E=\epsilon_a=1.0$. Here, the parameters are set as $\epsilon_a=1.0$, $\epsilon_b=0.0$, $\epsilon_c=2.0$, and $V=1.0$; therefore, the constraint condition, Eq. (11), is satisfied. If Eq. (11) is not satisfied, there is indeed no resonant transmission, as shown in Fig. 1(b), where the parameters are the same as those in Fig. 1(a), except $\epsilon_b=0.1$. Actually, in the case of resonant transmission, the width of the transmission peak can be enlarged if the hopping integral V is increased. The reason is that when V increases, the overlap between site Wannier orbitals is enhanced. Consequently, the width of the transmission peak will increase, but the resonant energy does not change. Figure 1(c) shows a wider peak around $E=\epsilon_a=1.0$ when V is increased to be $V=2.0$ and other parameters are kept same as in Fig. 1(a). Moreover, if we increase the strength of the disorder in the AST system by
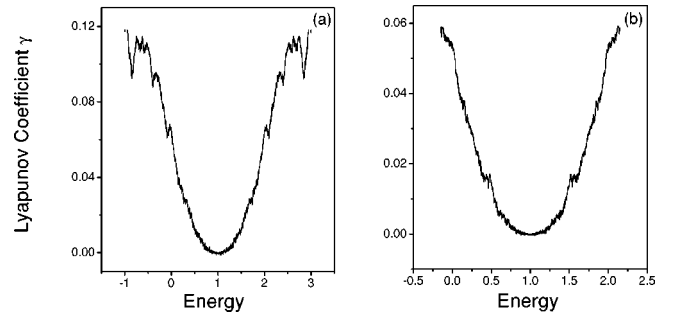

FIG. 2. The Lyapunov coefficient γ as a funtion of the energy for the AST model with $N=12\,000$. (a) $\epsilon_a=1.0$, $\epsilon_b=0.0$, $\epsilon_c=2.0$, and $V=1.0$. (b) $\epsilon_a=1.0$, $V=1.0$, ϵ_b and ϵ_c randomly distribute in the range $[0,2]$, respectively. Note that Eq. (11) is always satisfied in each trimer.

setting both ϵ_b and ϵ_c randomly distributed in the interval $[0,2]$, but letting Eq. (11) be always satisfied in every defect $\epsilon_b\epsilon_a\epsilon_c$, it is found that the transmission spectrum still possesses the resonant peak around $E=\epsilon_a=1.0$ [as shown in Fig. 1(d)].

IV. THE LYAPUNOV COEFFICIENT AND THE WAVE FUNCTION

It is known that in the disordered system, the involved matrices possess random elements, but statistical properties are available. One important quantity to reveal the physical problem in the theory of random matrices is the Lyapunov coefficient γ, which has the following expression:

$$\gamma=\lim_{N\to\infty}\frac{1}{N}\ln\frac{|\bar{S}(N)z(0)|}{|z(0)|}, \qquad (18)$$

where $\bar{S}(N)$ is the total transfer matrix across the sample chain and the initial vector is $z(0)=\binom{C_1}{C_0}$. According to the Furstenberg theorem,[22] the limit in Eq. (18) exists and converges to its mean value for long chains. In fact, the Lyapunov coefficient γ is directly related to some physical quantities, such as the localization length and the density of states in electronic and vibrational systems, and the free energy in random Ising models, etc. The localization length is inverse to the Lyapunov coefficient. When the length of the sample tends to infinity, the zero Lyapunov coefficient corresponds to delocalized states with infinite localized length. Therefore, dependent on the Lyapunov coefficient, we can obtain the overall behavior of the electrons, i.e., we can know whether they are localized or delocalized at specific energies in the system.

The Lyapunov coefficients γ of the AST chains are plotted in Fig. 2. It can be seen from Fig. 2 that around the corresponding resonant energy in each chain, γ is very close to zero though the fluctuation exists. Actually, the fluctuation can be eliminated when N increases, and the Lyapunov coefficient tends to a finite value for every eigenenergy of the system. Therefore, the zero Lyapunov coefficient appears at the resonant energy where the localized length is infinite.
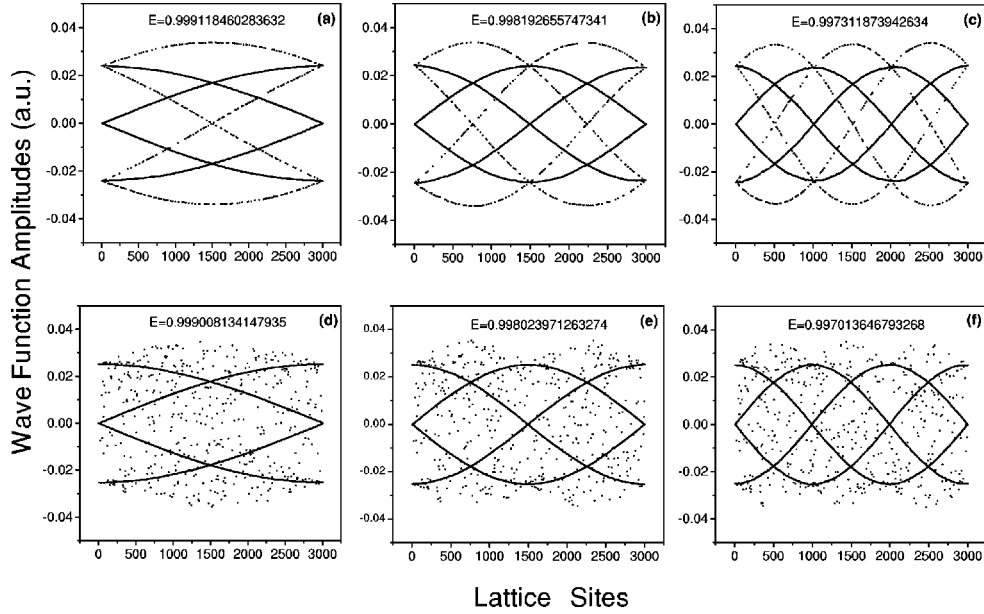
FIG. 3. The periodiclike wave function near the resonant energy for the AST model with $N=3000$. (a)–(c) In the vicinity of the resonant energy, the electronic wave function consists of two Bloch-like waves. Here, $\epsilon_a=1.0$, $\epsilon_b=0.0$, $\epsilon_c=2.0$, and $V=1.0$. (d)–(f) One of the Bloch-like waves persists, while the other turns to be chaotic. Here, $\epsilon_a=1.0$, $V=1.0$, ϵ_b and ϵ_c randomly distribute in the range $[0,2]$, respectively.

Under this condition, the electronic wave function cannot "feel" the existence of the defects, and the wave funtion can propagate through the chain without decay. It is different from the exponentially localized states occurring in the general Anderson model.

It is worthwhile to give a descriptive picture of the delocalized state at resonant energy. The spatial distribution of electronic wave functions is considered to be a direct and solid evidence of localized or delocalized states in low-dimensional systems. The wave function near the resonant energy is shown in Fig. 3. It is found that when the eigenenergy is in the vicinity of the resonant energy $E=\epsilon_a=1.0$, the electronic wave function consists of two Bloch-like waves [as shown in Figs. 3(a)–3(c)], and the periodicity of the wave function changes in different eigenvalues. More interestingly, if ϵ_b and ϵ_c are randomly distributed in the interval $[0,2]$, respectively, one of the Bloch-like waves persists, while the other turns to be chaotic [as shown in Figs. 3(d)–3(f)]. The chaos originates from the randomness of the site energy in the trimer impurity.

V. RANDOM-POLYMER CHAINS WITH IMPURITIES POSSESSING AN INVERSE SYMMETRY

Equation (11) indicates that the site energy antisymmetrically distributes around the center atom a in the trimer $\epsilon_b \epsilon_a \epsilon_c$. Actually, the above discussion can be generalized to a more common case, i.e., the random-polymer chain with inversely symmetric impurity (RAS polymer). The impurity cluster contains three parts: the central part consists of $2k-1$ ($k\geq1$) host atoms a; the left part consists of j ($j\geq1$) atoms with site energies distributed randomly as "$\epsilon_1\epsilon_2\cdots\epsilon_j$"; and the right part "$\epsilon_j'\cdots\epsilon_2'\epsilon_1'$" is the inver-

sion of the left one, that is, $\epsilon_a=(\epsilon_1+\epsilon_1')/2=(\epsilon_2+\epsilon_2')/2=\cdots=(\epsilon_j+\epsilon_j')/2$ is satisfied. Thus, the total transfer matrix across the impurity cluster in the RAS polymer model follows as

$$\bar{P}_m=\bar{P}_L\cdot\bar{P}_C\cdot\bar{P}_R, \tag{19}$$

where $\bar{P}_R$ is the transfer matrix across the left j sites of the polymer, $\bar{P}_C$ is the transfer matrix across the center $2k-1$ atoms a, and $\bar{P}_L$ is the transfer matrix across the right j sites of the polymer. It is obvious that

$$\bar{P}_C=\begin{pmatrix} A & -1 \\ 1 & 0 \end{pmatrix}^{2k-1}, \tag{20}$$

where $A=(E-\epsilon_a)/V$. Because the site energy within the impurity has an antisymmetric distribution around the center atom, it is ready to derive that when $E=\epsilon_a$, $\bar{P}_R$ and $\bar{P}_L$ can be written, respectively, as

$$\bar{P}_R=\begin{pmatrix} \Delta_j & -1 \\ 1 & 0 \end{pmatrix}\cdots\begin{pmatrix} \Delta_2 & -1 \\ 1 & 0 \end{pmatrix}\begin{pmatrix} \Delta_1 & -1 \\ 1 & 0 \end{pmatrix}\equiv\begin{pmatrix} X & Y \\ G & H \end{pmatrix} \tag{21}$$

and

$$\bar{P}_L=\begin{pmatrix} -\Delta_1 & -1 \\ 1 & 0 \end{pmatrix}\begin{pmatrix} -\Delta_2 & -1 \\ 1 & 0 \end{pmatrix}\cdots\begin{pmatrix} -\Delta_j & -1 \\ 1 & 0 \end{pmatrix}$$

$$\equiv(-1)^j\begin{pmatrix} X & G \\ Y & H \end{pmatrix}. \tag{22}$$

After substituting Eq. (19) by Eqs. (20)–(22), the total transfer matrix across the RAS polymer can be rewritten as

$$\bar{P}_m = (-1)^j \begin{pmatrix} X & G \\ Y & H \end{pmatrix} (-1)^{k-1} \begin{pmatrix} A & -1 \\ 1 & 0 \end{pmatrix} \begin{pmatrix} X & Y \\ G & H \end{pmatrix}$$

$$= (-1)^{j+k-1} (HX - GY) \begin{pmatrix} A & -1 \\ 1 & 0 \end{pmatrix}. \tag{23}$$

Using the property of unimodular matrix, finally we obtain

$$\bar{P}_m = (-1)^{j+k-1} \begin{pmatrix} (E-\epsilon_a)/V & -1 \\ 1 & 0 \end{pmatrix}. \tag{24}$$

That is, the matrix $\bar{P}_m$ turns to be proportional to that of the ordered system.

As we know, the reflection coefficient will vanish only when the matrix $\bar{P}_m$ is proportional to the unit matrix, or the transfer matrix of the ordered system, or some linear combinations of both.[11] In the case that the site energy within the impurity has an inverse symmetry around the central host element atoms, the total transfer matrix $\bar{P}_m$ across the impurity can be simplified to the transfer matrix of the ordered system at $E = \epsilon_a$. Consequently, the corresponding reflection coefficient is zero. Therefore, we reach a general conclusion that if the site energy within the impurity cluster has an antisymmetric distribution around the host element atoms, the resonant transport and electronic delocalization will definitely take place at the electronic energy that equals the site energy of the host atom. This result is beyond the general understanding that the internal mirror symmetry in the defects is the necessary requirement for the presence of delocalization in RDM-like systems. Moreover, when the number of central host atoms is large enough, the impurity cluster seems to be a cluster of two isolated random defects that are separated by a long periodic chain. However, the resonant scattering still exists due to the intrinsic correlation determined by the structure of the impurity. In this case, the electronic delocalization in the RAS polymer may be similar to the delocalization in the 1D Anderson model with a long-range correlated disorder.[23]

The RAS polymer model presented here is useful in understanding the high electronic conductivity of some polymers. Generally speaking, polymers are nonconductors. However, chemically doped polymers have shown a drastic enhancement on electronic conductivity, which shows the presence of the insulator-metal transition in some sense. As we know, the random-dimer model has succeeded in explaining the conductivity of polyaniline.[10,11] Our model of random polymer with an inverse symmetry provides wider perspectives on the conducting mechanism of polymers. According to the electronic-transport properties of this model, the resonant electronic scattering can take place at a specific energy, which originates from the spatial correlation, but not from the internal mirror symmetry of the impurities or bond defects as the RDM requires. Technically, it is possible to select some clusters with the inverse symmetry of the impurity distributions, and attach them to certain sites of the polymeric chain. Following this, we may synthesize different conducting polymers. Though we do not choose site and hopping energies of our Hamiltonians with specific reference to a realistic polymer, we have indeed presented the resonant scattering in the RAS polymer, and we can expect the presence of the metal-insulator transition in the polymers constructed based on this model.

VI. SUMMARY

Based on the tight-binding approximation and the on-site model, we have investigated the electronic transport and delocalization in a random-polymer model, where the impurity clusters are randomly distributed in the host monatomic chain, and the impurity atoms possess an inverse symmetry around the central host atoms in each cluster. The asymmetrical trimer $\epsilon_b \epsilon_a \epsilon_c$ is one of the examples. It is found that the resonant transport and electronic delocalization can take place in such systems. This result does not agree with the general viewpoint that the internal symmetrical structure in the defect is the necessary condition for the electronic delocalization in the RDM-like system. Furthermore, the RAS polymer model is valuable to understand the conducting mechanism of some polymeric materials, and it may provide a method to produce different types of polymers with high electronic conductivity.

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