Incoherent exciton trapping in self-similar aperiodic lattices

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Abstract

Incoherent exciton dynamics in one-dimensional perfect lattices with traps at sites arranged according to aperiodic deterministic sequences is studied. We focus our attention on Thue-Morse and Fibonacci systems as canonical examples of self-similar aperiodic systems. Solving numerically the corresponding master equation we evaluate the survival probability and the mean square displacement of an exciton initially created at a single site. Results are compared to systems of the same size with the same concentration of traps randomly as well as periodically distributed over the whole lattice. Excitons progressively extend over the lattice on increasing time and, in this sense, they act as a probe of the particular arrangements of traps in each system considered. The analysis of the characteristic features of their time decay indicates that exciton dynamics in self-similar aperiodic arrangements of traps is quite close to that observed in periodic ones, but differs significantly from that corresponding to random lattices. We also report on characteristic features of exciton motion suggesting that Fibonacci and Thue-Morse orderings might be clearly observed by appropriate experimental measurements. In the conclusions we comment on the implications of our work on the way towards a unified theory of the orderings of matter.

PACS numbers: 71.35+z, 05.60.+w, 61.44.+p, 02.60.Cb
I. INTRODUCTION

The interest in the study of the physical properties of elementary excitations in one-dimensional (1D) self-similar aperiodic systems has considerably grown during the last years. Albeit these systems were originally considered as somewhat intermediate between the periodic (crystalline) and random (amorphous, glassy) orderings of matter, it has been progressively realized that systems containing basic units arranged according to the Fibonacci, Thue-Morse, period-doubling, or Rudin-Shapiro sequences display novel properties which are not shared by the systems usually considered in condensed matter physics. In this way, we have recently provided strong evidence supporting the idea that self-similar aperiodic systems reveal a new kind of order, rather than representing a confuse mixture of periodic order and randomness. From a theoretical point of view this line of reasoning stems from the fact that, in spite of Bloch theorem being not longer valid for arbitrary aperiodic systems lacking translational symmetry, there exist certain aperiodic systems which still endow a significative degree of structural symmetry. Between them, those systems generated from the application of a substitution sequence deserve an especial attention, for they exhibit a characteristic scale invariance symmetry directly related to their self-similar nature.

As experimental realizations of such systems become available in the fields of quasicrystalline phase research and multilayered heterostructures technology, the interest in these aperiodically ordered forms of matter goes beyond a mere conceptual interest. For the sake of illustration let us mention but the following examples. In the area of quasicrystalline matter the rapidly solidified alloys Al$_{80}$Ni$_{14}$Si$_{6}$, Al$_{65}$Cu$_{20}$Mn$_{15}$, Al$_{65}$Cu$_{20}$Co$_{15}$, which are crystalline in two orthogonal directions and quasicrystalline along the other one, are worth being mentioned. On the other side, regarding multilayered structures, after the first fabrication of Fibonacci and Thue-Morse semiconductor superlattices by Merlin and co-workers, the obtention of metallic and superconducting quasiperiodic superlattices and networks has been reported.

It is well known that self-similar aperiodic systems, described by tight-binding and Kroning-Penney models, possess singular continuous energy spectra which are Cantor sets of zero Lebesgue measure. This point has been rigorously proven for Fibonacci, period-doubling, and Thue-Morse sequences and it has recently been conjectured, on sound mathematical basis, that this spectral type may be a common characteristic of all aperiodic systems obtained by the application of a substitution sequence. In this sense, a great deal of numerical analyses have shown that this kind of spectra exhibit a highly fragmented structure, with a hierarchy of splitting subbands displaying self-similar patterns and that the associated (generalized) eigenstates behave in a very peculiar manner, referred to as critical, characterized by dramatic spatial fluctuations and becoming neither localized nor extended in the usual sense. In addition, realistic estimations on the possibility of observing characteristic quasiperiodic effects in more complex systems, like Fibonacci Si $\delta$-doped GaAs superlattices, has been recently reported.

Hence, the question as to whether the peculiar structure of the energy spectrum of self-similar aperiodic systems influences the transport properties through the sample follows in a natural way. Most studies dealing with this topic up to date have been mainly concerned in both electronic and phonon transport problems, which can be treated, in a unified and simple mathematical scheme, within the transfer-matrix approach. Nevertheless, interesting results
involving other elementary quasiparticles, including plasmons\cite{28} photons\cite{29} excitons\cite{30} and polaritons\cite{31} in aperiodic lattices have been reported, thus considerably widening the field of possible practical applications of these systems.

In this work we will investigate \textit{incoherent} exciton dynamics in 1D self-similar aperiodic systems, considering the Fibonacci and Thue-Morse sequences as canonical examples. The main aim of this study is twofold. In the first place we ascertain how self-similar order modifies exciton dynamics in comparison with the dynamics associated to the long-range disorder of random systems. To this end we adopt the following strategy: We solve numerically the master equation governing the exciton motion and, from its solution, we obtain the survival probability and the mean square displacement of excitons. These parameters are then compared to those found in random lattices of the same size with identical fraction of traps. In the second place we determine the differences between exciton propagation through periodic chains and exciton transport in aperiodic systems displaying quasiperiodic order (Fibonacci) on one hand, and non-quasiperiodic order (Thue-Morse) on the other hand. In this way we are able to report on two interesting results. First, at least regarding exciton dynamics, self-similar aperiodic lattices are more similar to periodic lattices than they are to random ones and, secondly, exciton motion in quasiperiodic lattices also differs from the corresponding motion in non-quasiperiodic chains. In view of these results the notions of both \textit{quasiperiodic order} and \textit{self-similar order}, as denoting new and different classes of matter ordering, seem completely justified\cite{32}.

Finally, along with the possible applications of our results to the physical systems indicated previously —it is worth mentioning here that for short and intermediate times 1D transport may be relevant for three dimensional systems as well\cite{32}—, one of the most appealing aspects of the present work is to show how time evolution of quasiparticles (excitons in the present case) may be usefully employed to determine structural features of lattices. In particular, we demonstrate that excitons, initially created at a single site, act as a probe of the underlying structure as time evolves and the quasiparticle interacts with larger and larger regions of the system via the combined action of diffusion and trapping.

We will report on these issues according to the following scheme. In Sec. II we describe our model and the physical magnitudes we will compute in order to properly characterize exciton dynamics. Section III contains our main results concerning survival probabilities and mean square displacements of incoherent excitons, along with the corresponding interpretation of the obtained results. Section IV concludes the paper with a brief account on practical implications of our results and comments on some general ideas concerning the notion of \textit{aperiodic order} stemming from our study.

### II. MODEL

We consider excitations in a 1D lattice whose time evolution is described by the following master equation for the probability $P_k(t)$ to find the exciton at site $k$\cite{33}

$$\frac{d}{dt}P_k = W(P_{k+1} + P_{k-1} - 2P_k) - G_k P_k,$$

(1)

where $W > 0$ is the intersite rate constant, which is assumed to be independent of $k$ hereafter and $G_k = G$ is the trapping rate at site $k$. The quantity of interest in luminescence
experiments is the survival probability \( n(t) \) defined as
\[
n(t) = \sum_k P_k(t),
\]
where the index \( k \) runs over all lattice sites. Moreover, assuming that the excitation is initially at site \( k_0 \) \((P_k(0) = \delta_{kk_0})\), we can also calculate the mean square displacement of the excitation (which is related to the diffusion coefficient\(^{[33]}\)) as follows
\[
R^2(t) = \sum_k (k - k_0)^2 P_k(t),
\]
where the lattice spacing is taken to be unity hereafter. These two functions characterize the dynamics of excitons in the lattice under the combined action of diffusion and trapping. Thus, it is known that, in infinite lattices without traps \((G_k = 0)\), the survival probability is conserved \((n(t) = 1)\) and the mean square displacement increases linearly with time\(^{[34]}\) \((R^2(t) = 2Dt, \ D \text{ being the diffusion coefficient})\).

In what follows we consider that \( G_k \) can only take on two values, \( G_A = 0 \) and \( G_B = G > 0 \), that is, only sites \( B \) are able to trap excitons. We will arrange sites \( A \) and \( B \) according to the Thue-Morse sequence, the Fibonacci sequence, at random, or periodically, depending on the particular kind of lattice we are interested in. For convenience we define \( c \) as the ratio between the number of traps and the total number of sites in the considered lattice \( N \). Deterministic aperiodic sequences can be generated by simple substitution rules. Thus, we have \( A \rightarrow AB, \ B \rightarrow BA \) for the Thue-Morse sequence and \( A \rightarrow AB, \ B \rightarrow A \) for the Fibonacci one. Finite, self-similar lattices are obtained in this way by \( l \) successive applications of the substitution rule. The \( l \)th generation lattice has \( 2^l \) elements for the Thue-Morse lattice (TML) and \( F_l \) elements for the Fibonacci lattice (FL), where \( F_l \) denotes the Fibonacci numbers. Such numbers are generated from the recurrence relationship \( F_l = F_{l-1} + F_{l-2} \) with \( F_0 = F_1 = 1 \); as \( l \) increases the ratio \( F_{l-1}/F_l \) converges toward \( \tau = (\sqrt{5} - 1)/2 = 0.618 \ldots \), which is known as the inverse golden mean. Therefore, sites are arranged according to the sequence \( ABBABAABA \ldots \) in the TML, and \( ABBABAABA \ldots \) in the FL. The value of \( c \) is strictly equal to 0.5 for any generation of the TML. On the contrary, the value of \( c \) depends on the particular generation of the FL, but for large enough systems one has \( c \sim 1 - \tau = 0.3819 \ldots \). Disordered lattices are obtained by placing traps (sites \( B \)) at random over the lattice maintaining fixed the concentration of traps \( c \). Finally, we consider in this work periodic lattices of two types. One of them is set with \( c = 0.5 \) and traps placed at sites with even index. This periodic lattice will be compared to the TML. The other type is obtained from a periodic superposition of unit cells of the form \( ABAABAABAA \), which is nothing but the fifth order approximant to the Fibonacci sequence. The concentration of traps is \( c = 0.375 \) for this periodic arrangement, a value rather close to the value \( 1 - \tau \) corresponding to infinite FLs.

**III. NUMERICAL RESULTS AND DISCUSSIONS**

We have numerically solved the master equation \((1)\) for TML, random and periodic lattices of \( N = 2^{10} = 1024 \) units and for FL, random and periodic lattices of \( N = F_{15} = 987 \) units using an implicit (Crank-Nicholson) integration scheme. To avoid free ends effects,
spatial periodic boundary conditions are introduced, so that the detailed balance required by Eq. (1) is preserved. The initial condition for the exciton motion is $P_k(0) = \delta_{kk_0}$, with $k_0 = 500$ ($k_0 = 494$) for lattices with $N = 1024$ ($N = 987$) sites, that is, we will assume that the exciton is created, for instance by a pulsed excitation, roughly at the middle of the lattice. Trapping rate $G$ will be measured in units of $W$ whereas time will be expressed in units of $W^{-1}$. The maximum integration time and the integration step are 250 and $5 \times 10^{-4}$, respectively. Smaller time steps led to similar results. Since we are mainly interested in the effects due to particular arrangements of traps rather than in a detailed description of the influence that the different parameters have in the incoherent motion of excitations, we will fix the values of $W$ and $G$ henceafter. Thus we have set $W = 1$ and $G = 0.2$ as representative values. For disordered lattices a series of random distribution of traps was generated for a given trap concentration, and ensembles comprising a number of realizations varying from 50 to 200 were averaged to check the convergence of the computed mean values. Since convergence was always satisfactory between all the ensembles, we present the results corresponding to 50 averages.

The obtained results for the mean square displacement and survival probability of excitons propagating through the TML are presented in Figs. 1(a) and 1(b), respectively, along with the corresponding results for random and periodic lattices with a trap concentration of $c = 0.5$. Analogous magnitudes describing the motion of incoherent excitons through the FL and related random and periodic lattices are shown in Figs. 2(a) and 2(b). Let us consider, in the first place, the behavior of the mean square displacement of incoherent excitons through these systems. In all cases it becomes apparent that the time evolution of $R^2(t)$ arises from the competition between two different processes, namely diffusion (the exciton is transferred from site to site, starting at $k_0$) and trapping (the exciton progressively decays in time since possible detrapping processes are not considered in our model). At short times the first mechanism dominates because of the exciton is still close to the initial position and, consequently, there exist small chances to be trapped. As time elapses, the probability of trapping also increases since the exciton can be found in a larger segment of the lattice. This competition gives rise to the occurrence of a well defined maximum in $R^2(t)$, whose position depends not only on the concentration of traps but mainly on the spatial distribution of these traps. In addition to this quite general behavior we observe significant differences between the exciton behavior in quasiperiodic (Fibonacci) and non-quasiperiodic (Thue-Morse) aperiodic lattices. In fact, the mean square displacement of an exciton propagating through a TML essentially coincides with that corresponding to the case of the periodic lattice over the entire time interval we have considered. Moreover, the $R^2(t)$ curve describing the exciton motion in the random lattice appreciably differs from both the TML and periodic corresponding curves. On the contrary, the mean square displacement of excitons in the FL cannot be easily compared with that of excitons moving in neither periodic nor random lattices at short times but, as time increases, exciton motion in FLs progressively resembles that taking place in the periodic lattice approximant.

Now, we turn our attention to the evolution of the survival probability. It is well known that, for any periodic distribution of traps, the behavior of the survival probability is simply exponential in time, and is given by the expression $n(t) = \exp(-cGt)$, whereas in random lattices presents a more complex and non-exponential dependence on time. Keeping this fact in mind, the interpretation of Figs. 1(b) and 2(b) is straightforward. The rate of trapping
of incoherent excitons in both the TML and FL is very similar to that of the corresponding periodic lattices with the same fraction of traps and quite different from that associated with the corresponding random lattices. Therefore, from this point of view, self-similar aperiodic systems behave as periodic ones in a very close manner. In particular we note that not only an exponential decay rate for both kind of aperiodic systems is observed, but the slope of the corresponding survival probabilities fits the value prescribed by the trap concentration \( c \) appearing in the general expression for periodic systems. Finally, note that the decay rate in random lattices is much slower than in the other lattices (periodic and aperiodic).

IV. CONCLUSIONS

From the comparison of the mean square displacement and survival probability plots for the lattices considered in this work, several conclusions can be drawn. In the first place we point out that excitons propagating through self-similar aperiodic lattices behave in a very similar way as they do in periodic 1D systems. We wish to stress, at this respect, that we are not merely saying that excitonic behavior resembles that corresponding to excitons moving in periodic lattices, but stating that exciton dynamics in self-similar chains exhibits an time evolution completely different from that they show in random systems. A second important result emerging from our numerical simulations is that the exciton dynamics in the TML significatively differs from that recorded in the FL at short times. This can be easily seen by comparing the corresponding mean square displacement curves. The justification for this effect can be accounted for starting from the following picture: As time evolves the exciton progressively extends over the lattice and, in this sense, it acts as a probe indicating the rate of trapping associated to the particular arrangement of traps of the underlying structure. In this way, the shape of the \( R^2(t) \) curve can be interpreted in a topological sense. As it has been explained previously the characteristic maximum of this curve indicates a cutoff between two different transport regimes in the system. At short times we have classical diffusion through the lattice, meanwhile at longer times the effects of trapping become dominant. Fig. 1(a) indicates that excitons propagate through the TML as they will do through a periodic lattice having the same trap concentration in both regimes. Hence, non-quasiperiodic order associated to the Thue-Morse sequence has no relevant effects on exciton dynamics and, as long as transport properties are concerned, Thue-Morse and binary periodic arrangements with a trap concentration \( c = 0.5 \) are completely equivalent. On the contrary, the shape of the \( R^2(t) \) curve, shown in Fig. 2(a), clearly reveals that quasiperiodic order has a profound effect on the excitonic diffusion transport regime. In fact, we see that diffusion of excitons in the FL is considerably lower than that taking place in both periodic and random lattices at the same times. This result, along with the fact that the area determined from the expression \( \int_0^\infty R^2(t) \, dt \) is also smaller than the corresponding values for the other lattices, led us to the conclusion that trapping processes are more efficient for quasiperiodic arrangements than they are for other possible orderings, including both periodic (crystalline) and random (glassy) structures. This interesting result may be of relevance from an experimental point of view, since by properly measuring the value of diffusion constants in aperiodic lattices, we should be able to estimate the kind of underlying topological order they present.

To conclude we wish to comment on some general ideas concerning the notion of aperiodic order which stem from our study. The results reported on in this work provide substan-
tial support to the view, previously put forward from our study of electronic transport in quasiperiodic systems, that aperiodic systems cannot be regarded, in general, as systems endowed with an intermediate degree of structural disorder. On the contrary, certain classes of aperiodic systems, like those arranged according to the Fibonacci and Thue-Morse sequences, are notable representatives of highly ordered systems, actually displaying, in some particular aspects, a higher level of order than usual periodic systems are able to do. In this way we realize that it might well be that the long-standing creed of periodic order as a canonical prototype of perfect order should be reconsidered.

A further step in this conceptual progress from the notion of randomness to that of perfect order comes from the progressive realization that not all self-similar aperiodic orderings can be put on the same footing. In fact, when comparing quasiperiodic and non-quasiperiodic arrangements by means of different criteria we usually find that, just depending on the adopted criteria, one kind of system seems to be more or less ordered than the other one. In this sense, it has been claimed, on the basis of the electronic wavefunctions behavior, that the TMLs might be regarded as providing a link between the FLs and the periodic lattices, and that the study of non-quasiperiodic aperiodic systems is a promising way towards a unified theory of ordered systems, able to encompass periodic as well as aperiodic orderings of matter. This earlier suggestion can be phrased in a novel fashion by introducing the concept of hierarchies of order. By this we mean that rather than thinking of different kinds of order, classified into separated categories ranging from those more periodic to those more random, as it has been the usual procedure in a number of recent works, it may be more fruitful to separate different kinds of order in a qualitative way by grading them according to a well established criterion. Since only two kinds of binary aperiodic lattices have been extensively studied to the date, it is hard to propose such a criterion on a sound basis. Nevertheless, it may be confidently conjectured that it will ultimately have to do with the self-similar properties of aperiodic deterministic lattices generated by the application of substitution sequences. Since self-similarity endows these structures with scale invariance symmetry properties, we feel that a systematic analysis of this kind of symmetry on rigorous mathematical foundations is required before a proper understanding of aperiodic order can definitively be attained.

ACKNOWLEDGMENTS

This work is partially supported by Universidad Complutense through project PR161/93-4811. A. S. is partially supported by DGICYT (Spain) grant PB92-0248, by MEC (Spain)/Fulbright, and by the European Union Network ERBCHRXT930413. Work at Los Alamos is performed under the auspices of the U.S. D.o.E.
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FIGURES

FIG. 1. (a) Mean square displacement and (b) logarithm of the survival probability of excitons as a function of time for lattices of $N = 1024$ sites with $c = 0.5$. Results correspond to Thue-Morse (solid lines), periodic (long-dashed lines), and random (short-dashed lines) arrangements of traps.

FIG. 2. (a) Mean square displacement and (b) logarithm of the survival probability of excitons as a function of time for lattices of $N = 987$ sites with $c = 0.382$. Results correspond to Fibonacci (solid lines), periodic (long-dashed lines), and random (short-dashed lines) arrangements of traps.