Density of States for a Two-Dimensional Penrose Lattice: Evidence of a Strong Van Hove Singularity

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The density of states for a tight-binding Hamiltonian on a two-dimensional Penrose lattice is computed numerically by the continued fraction recursion method. The result shows evidence of a strong Van Hove–type singularity which is remarkable for a system possessing *no* long-range periodic translational order. By a method of finite-size scaling extrapolation the exponent α and amplitude C, where $\rho(E) \sim CE^{-\alpha}$, are estimated to be $\alpha = 0.90 \pm 0.05$ and $C = \exp(3.8 \pm 0.2)$.

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Considerable excitement has been generated in recent months as a result of the experiments of Shechtman *et al.*[1] who gave the first crystallographic data for the existence in nature of quasicrystals ($Al_{86}Mn_{14}$).[2] These systems are characterized by a lack of long-range periodic translational order (and hence no Bravais lattice) but have long-range bond orientational order, a canonical example of which is perhaps the pattern of Penrose tilings in two dimensions (2D) (Fig. 1). Motivated by this evidence, several authors have begun to study their structural and electronic properties.[2,3] Of particular importance is the presence of sharp Bragg spots in the diffraction pattern in the absence of long-range periodic translational symmetry as demonstrated by Levine and Steinhardt.[2] In this Letter I wish to report on the first results for the density of states of a tight-binding Hamiltonian on such a lattice in 2D. My viewpoint is more closely related to that adopted in earlier work on topologically disordered random networks, e.g., in studies of amorphous silicon,[4] in contrast to the viewpoint of other authors who have made use of stereographic projections of curved-space ideal crystals.[3]

In particular, I wish to address the question of possible true Van Hove–type singularities in the distribution for this lattice, since it is perhaps common folklore that disorder tends to round off any Van Hove singularities of a pure crystal as is indeed indicated by various numerical calculations.[5] There is, however, a high degree of orientational order on the Penrose lattice which may lead to features in the density of states arising from its peculiar self-similarity properties. It is well known that (at least with s electrons) the properties of the Hamiltonian are given entirely by the connectivity matrix,[6] on which we shall focus our main attention. In this context the lattice is especially interesting as it contains $n = 3$-, 4-, 5-, 6-, and 7-fold coordinated sites, and hence differs somewhat from fixed coordinated random networks,[4] whose densities are not random, and is probably connected by factors of the golden ratio.[7] Here we will be particularly interested in the local density of states (LDOS)

$$\rho_i(E) = \sum_n \psi_n(i)\psi_n^*(i)\delta(E-\epsilon_n),$$

at a particular n-fold coordinated site as well as its thermodynamic average

$$\rho(E) = (1/N)\sum_i \rho_i(E),$$

the global density of states (GDOS).

The absence of odd-membered rings, however, allows division into two sublattices, and by well-known arguments[4,8] we deduce that the LDOS and GDOS are even functions of E. Unfortunately, in spite of the high degree of symmetry, the problem is probably not amenable to an exact analytic solution on the infinite lattice. After examining various procedures I have adopted the widely used recursion method of Haydock, Heine, and Kelly[9,10] which offers fast computa-

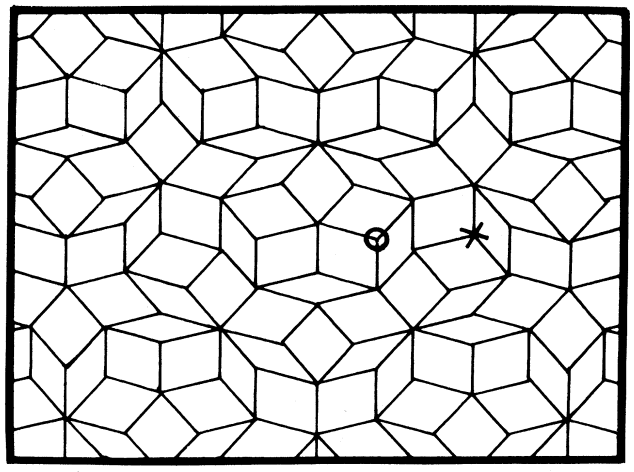

FIG. 1. A section in the bulk of a computer-generated cluster of a 2D Penrose lattice (3806 atoms) using the algorithm of Mackay (Ref. 12). The algorithm can deflate indefinitely to any number of generations. LDOS for the atoms marked with a circle and a cross are shown in Fig. 3.

tional speed and, above all, reliability. Technical details concerning the implementation of the recursion procedure to this problem are best deferred to a later publication.[11] Briefly, the continued-fraction expansion of the Green's function, which is exact for a finite system, is truncated in the infinite lattice. Information on the connectivity of the lattice is given by a nearest-neighbor table generated by the algorithm of Mackay[12] (Fig. 1). A small imaginary part γ is added to the energy to smooth out the distribution, but I have avoided using terminator functions, which are unreliable for this problem when we have *no* prior knowledge of the shape of the distribution and band edges. To assess the quality of this procedure we first check the recursion algorithm on the three typical 2D lattices, i.e., for the square, triangular, and honeycomb nets (Fig. 2), for which exact results are well known.[11,13,14] Their distributions each have individual characteristics which are useful tests and can be expressed in closed form in terms of complete elliptic integrals of the first kind $K(k)$.[15]

The numerical calculations of Fig. 2 were done for over 3000 atomic sites taking 32 sec of central-processing-unit time on a Cyber machine but only 6 sec on the Cray *without* special efforts for vectorization. However, on the Penrose tiles most of the computing time is employed in generating the atomic cluster, which takes over 200 sec for about 3800 atoms, the main limitation here coming from memory size rather than computing speed. Looking at Fig. 2 we can see that the finite-size effects reduce and round off the logarithmically divergent Van Hove singularities[16] and sharp band edges, the usual feature of a numerical calculation. Although the logarithmic singularity is often more difficult to observe in a finite system, we see, nevertheless, that the various peaks are well resolved in these calculations. These points are to be noted in the interpretation of the data (Fig. 3) which are the main results of this Letter. The choice of initial vector is crucial in all of these calculations. For the LDOS we place an orbital at the chosen site well in the interior of the cluster and ensure that the continued fraction is not extended beyond $2L$ or $3L$, where L equals the square root of the number of atoms. For the GDOS we choose an initial vector with all elements as random variables chosen from a Gaussian distribution.

However, in this method of averaging for the GDOS care has to be taken in the interpretation of the data. The effect of open boundaries with dangling bonds, an unavoidable feature of the deflation algorithm, might introduce spurious small peaks near the band edges that are not genuine features of the bulk. The alternative of directly averaging over many atoms by repeatedly computing a LDOS is computationally inconvenient and will still not remove the effect of boundaries. In Fig. 3(a) we see two sharp peaks for the LDOS on the threefold coordinated site marked with a circle in Fig. 1. Such sites are very few in number. On a typical network of 1500 atoms fewer than ten such atoms have been located (by random selection) in the bulk. The majority of threefold coordinated sites, like most of such sites shown in the cluster of Fig. 1, pro-

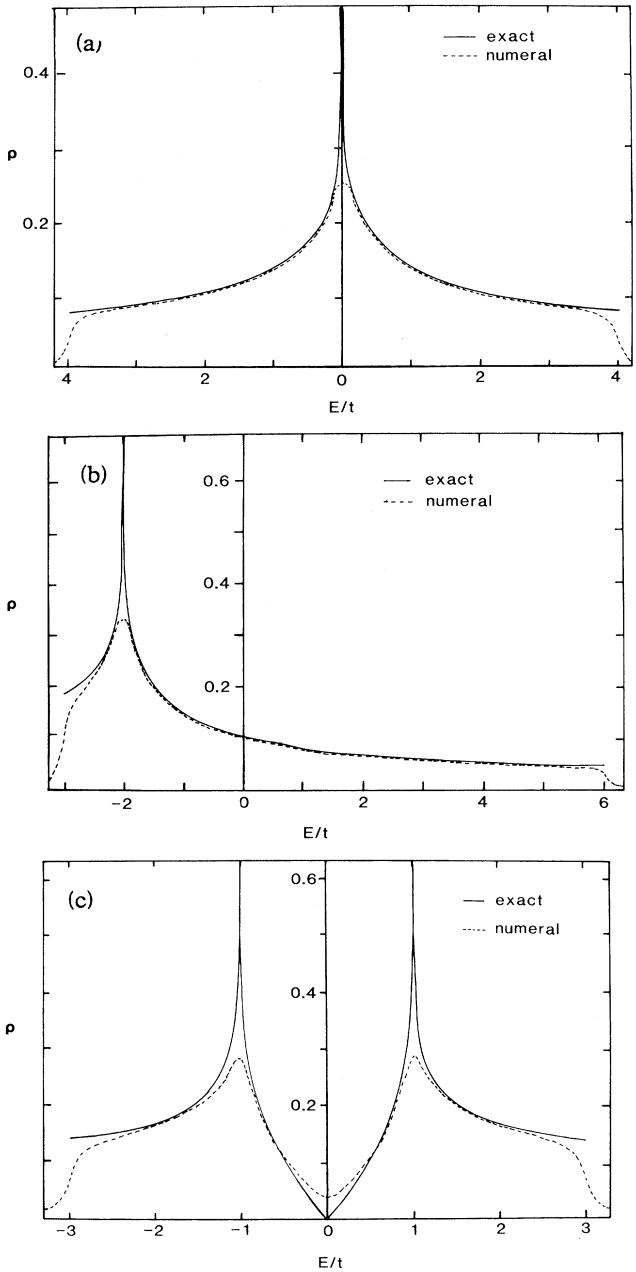

FIG. 2. Comparison of numerical and exact results for the LDOS on the (a) square, (b) triangular, and (c) honeycomb lattices. Numerical calculations were done for 3271 atoms with free boundary conditions and a truncation of the continued fraction at 200 levels with $\gamma = 0.1$.

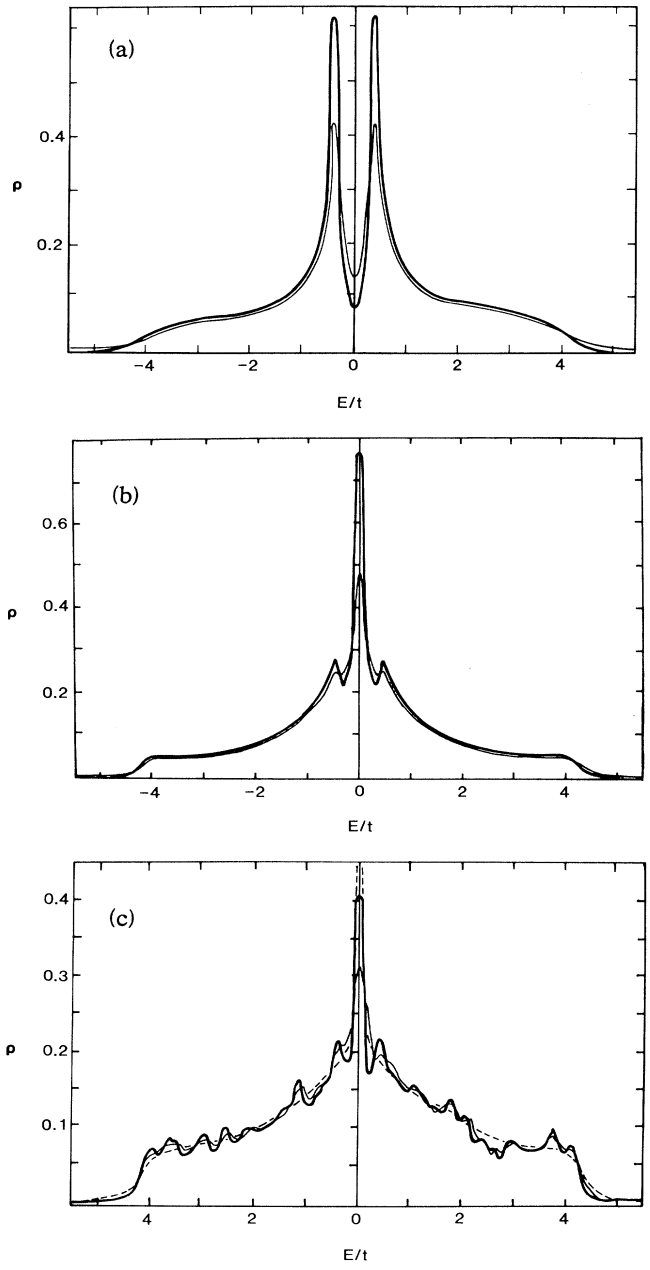

FIG. 3. (a) LDOS at the threefold coordinated site marked with a circle in Fig. 1, which shows two peaks. (b) LDOS at the threefold coordinated site marked with a cross in Fig. 1, which shows one peak. (c) Solid curves are raw data for the GDOS using the average technique discussed in the text. The dotted curve is the predicted GDOS without boundary effects and noise. Heavy curves are for 200 levels with $\gamma = 0.1$ and light curves are for 100 levels with $\gamma = 0.15$.

vide only a single sharp peak. I have also examined the LDOS on other n-fold ($n > 3$) coordinated sites. The scarcity of such sites on the finite lattice again introduces boundary difficulties that must be carefully handled. I found no evidence of sharp peaks comparable to those above. Finally, using the averaging technique discussed above, I show the GDOS of a cluster of 3806 atoms [Fig. 3(c)]. The full curves are the raw data while the dotted plot is my prediction. On the basis of the points mentioned above we do not expect that the smaller peaks near the band edges are genuine features of the bulk. The center peak, however, appears to persist unrenormalized. This evidence strongly suggests that the two peaks of Fig. 3(a) do not survive the thermodynamic averaging process over the whole system. By comparison with previous test results (Fig. 2) we clearly see evidence that the singularities of Figs. 3(a)–3(c) are more strongly divergent, a remarkable feature. Furthermore, I have explicitly diagonalized numerically the connectivity matrix of the first-generation deflationary pattern of 29 atoms. Only three states belong to zero energy, adding to the evidence that the singularity observed is a true topological effect of the infinite cluster. The interplay between a finite γ and system size make the process of obtaining a finite-size scaling estimate of the exponent tricky. The following method which I used, although crude, has been tested on the 1D chain and yields the correct result to within 1% for a 1000-atom system. This is the first time a finite-size scaling extrapolation has been used with the recursion method. The procedure is to plot the peak height of the global density of states versus $\ln\gamma$ over a range that does not make the distribution noisy. The slope of this plot gives the exponent $\alpha(N)$ and the intercept gives $\ln C(N)$ where C is the amplitude. This procedure is repeated against $1/N$ and extrapolating to zero gives an estimate: $\alpha = 0.90 \pm 0.05$, $\ln C = -3.8 \pm 0.2$. No attempt will be made at this stage to assess the consequences of these results. Clearly, they suggest strongly the need for an extension of current concepts of solid-state physics to quasiperiodic structures. Extensions to more dimensions and an investigation of transport properties like the conductivity will be published elsewhere.

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