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Continued-fraction technique for tight-binding systems. A generalized-moments method

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The tight-binding scheme has been largely used in the description of the electronic properties of disordered systems. Several direct-space techniques of calculation have been developed in order to compute the density of electronic states; among those the moments method and the recursion scheme have become popular. We discuss both of them and we propose a new method that interpolates between these two techniques. It is based on the calculations of generalized moments. We show that the generalized-moments method shares the advantages of both methods and avoids their difficulties.

I. INTRODUCTION

Disordered systems (amorphous semiconductors, transition-metal alloys, etc.) have been widely studied in a one-electron description using parametrized tight-binding Hamiltonians. The calculation of the density of electronic states in noncrystalline systems requires special techniques that do not make use of translational invariance. Most of them have been developed in the last decade. The most important are the moments method,[1,2] the recursion method,[3,4] the cluster-Bethe-lattice method,[5,6] and the equation-of-motion method.[7] All these methods have been proven useful when the Bloch theorem is not applicable: Nearly ten years of experience has shown that all these techniques are comparable with characteristic advantages and limitations that we briefly discuss here before presenting the generalized-moments method.

The moments method consists of the calculation of the power moments (1) of the density of states

$$\mu_k=\int_{-\infty}^{+\infty} E^k n(E)dE \tag{1}$$

that are nothing but the coefficients of the expansion of the diagonal elements of the resolvent $R(z)$ in inverse powers of the complex energy variable z

$$R_0(z)\equiv\langle 0|(z-H)^{-1}|0\rangle=\sum_{k=0}^{\infty}\frac{\mu_k}{z^{k+1}} , \tag{2}$$

where $|0\rangle$ denotes the orbital on which the density of states is calculated. In a tight-binding Hamiltonian, the moments are easily computed: They are associated with closed walks on the structure.[1] As shown by Eq. (1) the moments are linear on the density of states so that they can be configurationally averaged. The calculation of the density of states from the moments is the famous "inverse-moments problem" solved by Stieltjes[8] using a continued fraction expansion of the resolvent $R_0(z)$.[4] Numerically this problem is ill-conditioned and consequently a high accuracy in the calculation of the moments is required. Indeed, the moments do not store properly the information: In a moment of given order a larger contribution comes from lower-order moments as shown in Sec. II. The same problem arises in the related Gaussian quadrature where the poles and residues are determined from the moments of the weight function.[9]

The so-called "recursion method"[3,4] is an adaptation of Lanczos matrix tridiagonalization algorithm[10]: The coefficients of the continued fraction are directly calculated by an algorithm optimized from the point of view of numerical stability, as demonstrated in Sec. III. However, the continued fraction coefficients cannot be averaged as they are strongly nonlinear on the density of states.

Apart from these two differences (numerical stability and linearity), the moments method and the recursion method are essentially identical. In both methods the spectral resolution depends on the size of the cluster. Outside the cluster, an unstructured external mean field is assumed; practically this is realized by the continuation of the fraction with asymptotic constant coefficients.

We present in this paper a new method that has the advantages of both the moments method (linearity) and the recursion method (numerical stability). The basic quantities of the method are the "generalized moments" defined by

$$\nu_{2k(-1)}=\int_{-\infty}^{+\infty} P_k(E)P_{k(-1)}(E)n(E)dE , \tag{3}$$

where P_k and P_{k-1} are, respectively, polynomials of degree k and $k-1$. As we show in the following sections in detail, the power moments corresponds to $P_k(E)=E^k$ and the recursion algorithm corresponds to $P_k(E)$ which are orthogonal polynomials with respect to $n(E)$. Having chosen a polynomial sequence $P_k(E)$, $k=0,1,2,\ldots$, the generalized moments are linear upon the density of states, by construction. A careful choice of the polynomial $P_k(E)$ eliminates most of the redividant contributions present in the power moments.

This paper is organized as follows: Sec. II recalls the main aspects of the power-moments method with a special attention to the formulas that are subject to generalization in the generalized-moments method which is developed in Sec. III. Some examples are treated in Sec. IV.

II. THE POWER-MOMENTS METHOD

In this section we briefly recall the principles and main formulas of the power-moments method. In order to obtain the local density of states $n(E)$, a two-step procedure is developed. The power moments $(\mu_0,\mu_1,\ldots,\mu_{2n})$ of the density of states are computed as shown below. In the second step, $n(E)$ is determined from its moments: It is the famous "inverse-moments problem." The calculation of the moments is particularly simple in the case of one-electron Hamiltonians. We consider a one-electron tight-binding Hamiltonian H,

$$H=\sum_i \epsilon_i \,|\,i\,\rangle\langle\, i\,| + \sum_{\substack{i,j\\ j\neq i}} \beta_{ij}\,|\,i\,\rangle\langle\, j\,| \ . \tag{4}$$

For the sake of simplicity we neglect the orbital degeneracy so that $|\,i\,\rangle$ labels the atomic site. ϵ_i is the atomic level and β_{ij} is the hopping (or resonance) matrix element. The extension of the method, as well as that of Sec. III, to Hamiltonians with orbital degeneracy is straightforward. However, the extension of the moments method to many-body Hamiltonians is difficult,[11] essentially because the calculation of the moments (i.e., the powers of H) is hard to perform.

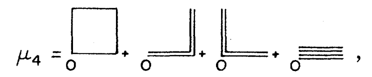
The inspection of the right-hand side of Eq. (2) shows that the moments are nothing but the diagonal elements of the powers of the Hamiltonian. If we label 0 the site on which the local density of states is calculated, we get

$$\begin{aligned}\mu_l&=\langle 0\,|\,H^l\,|\,0\rangle\\ &=\sum_{i_1,\ldots,i_{l-1}} \langle 0\,|\,H\,|\,i_1\rangle\\ &\qquad\times\langle i_1\,|\,H\,|\,i_2\rangle\cdots\langle i_{l-1}\,|\,H\,|\,0\rangle \ .\end{aligned} \tag{5}$$

In practice, formula (5) is written as

$$\begin{aligned}\mu_{2k}&=\sum_i \langle 0\,|\,H^k\,|\,i\,\rangle\langle\, i\,|\,H^k\,|\,0\rangle \ ,\\ \mu_{2k+1}&=\sum_i \langle 0\,|\,H^k\,|\,i\,\rangle\langle\, i\,|\,H^{k+1}\,|\,0\rangle \ ,\end{aligned} \tag{5'}$$

from which it is obvious that the even-order moments are positive. Practically the computer calculation requires the simultaneous storage of the two vectors $\langle 0\,|\,H^k\,|\,i\,\rangle$ and $\langle 0\,|\,H^{k+1}\,|\,i\,\rangle$, a total of $2\times N$ words where N is the numbers of orbitals of the structure. As it is obvious from (1) the moments are linear upon the density of states; consequently, the moments of the average density of states are the corresponding averages of the moments of the individual densities of states. This property is largely used in disordered systems (alloys, amorphous and liquid structures, etc.) in which a configurational average usually has to be taken. The geometrical meaning of the moments is obvious by (5): The moment of order k is the sum of the contributions of all the closed walks of length k that can be done on the structure by successive jumps from site to site connected by a nonvanishing element of the Hamiltonian β_{ij}. For example, the moment of order 4 on the square lattice is written pictorially,[12]



(6)

$$\mu_4=8\beta^4+12\beta^4+12\beta^4+4\beta^4=36\beta^4 \ .$$

The first two terms involve atoms from the first and the second coordination shells; the last two terms involve only atoms from the first coordination shell and so they correspond to information already contained in the lower-order moments (μ_2).

For high-order moments μ_k, the kth-order proper contribution is dominated by the contribution of the walks confined in the near neighborhood of the central atom. The proper contribution μ_{2k}^P to the even-order moments for a symmetric band has been estimated[13] to be

$$\mu_{2k}^P \sim (Z\beta/2)^{2k}$$

while the value of the moment is of the order of $(Z\beta)^{2k}$ so that the ratio is asymptotically given by

$$\frac{\mu_{2k}^{p}}{\mu_{2k}} \sim \frac{1}{2^{2k}} . \tag{7}$$

It becomes negligible when the order of the moments increases. This is the main drawback of the moments method: It has awkward consequences on the numerical stability of the inverse-moments problem. The new method presented in Sec. III avoids most of this difficulty. The second step consists in calculating the diagonal element $R_0(z)$ of the resolvent operator (2) under the form of a continued fraction[8]

$$R_0(z)=\cfrac{1}{z-a_1-\cfrac{b_1}{z-a_2-\cfrac{b_2}{z-a_3-\cdots}}} . \tag{8}$$

The coefficients b_k are positive. The coefficients a_k and b_k are determined from the moments by different algorithms[14,15] that are numerically ill conditioned: they are of PD (product difference) or QD (quotient difference) type. In a typical calculation on a 32 bit word computer, b_k has no more significant digit when $n \gtrsim 10$.

The relation between the moments and the coefficients a_k and b_k is formally given by ratios of the Hankel determinants Δ_k and $\Delta_k^{(1)}$ defined by

$$\Delta_k=\det(H_k)=\det\begin{bmatrix} \mu_0 & \mu_1 & \mu_2 & \cdots & \mu_k \\ \mu_1 & \mu_2 & \mu_3 & \cdots & \mu_{k+1} \\ \mu_2 & \mu_3 & \mu_4 & \cdots & \mu_{k+2} \\ \vdots & \vdots & \vdots & & \vdots \\ \mu_k & \mu_{k+1} & \mu_{k+2} & \cdots & \mu_{2k} \end{bmatrix} , \tag{9}$$

and

$$\Delta_k^{(1)}=\det(H_k^{(1)})=\det\begin{bmatrix} \mu_1 & \mu_2 & \mu_3 & \cdots & \mu_{k+1} \\ \mu_2 & \mu_3 & \mu_4 & \cdots & \mu_{k+2} \\ \mu_3 & \mu_4 & \mu_5 & \cdots & \mu_{k+3} \\ \vdots & \vdots & \vdots & & \vdots \\ \mu_{k+1} & \mu_{k+2} & \mu_{k+3} & \cdots & \mu_{2k+1} \end{bmatrix} . \tag{10}$$

Then

$$a_k=\frac{1}{\Delta_{k-2}^{(1)}}\left[\frac{\Delta_{k-1}\Delta_{k-3}^{(1)}}{\Delta_{k-2}}+\frac{\Delta_{k-1}^{(1)}\Delta_{k-2}}{\Delta_{k-1}}\right] \tag{11}$$

and

$$b_k=\frac{\Delta_k\Delta_{k-2}}{(\Delta_{k-1})^2} . \tag{12}$$

We keep this formulation because it is easily generalized for our purpose. But in practice, because the calculation of the determinant requires too many operations and is unstable, the a_k and b_k coefficients are calculated recursively from the moments.

III. THE GENERALIZED-MOMENTS METHOD

The generalized-moments method consists of defining a new type of moments that are linear on $n(E)$ like the power moments, but that are better numerically conditioned than the latter. The idea is to use a particular type of modified moments m_l generally defined by[16,17]

$$m_l=\int_{-\infty}^{+\infty} Q_l(E)n(E)dE , \tag{13}$$

that are obviously linear on $n(E)$. The interesting situation corresponds to alternate signs of the coefficients of $Q_l(E)$, i.e., at the order l, lower-moments contributions are subtracted. However, it is, in general, impossible to compute *directly* the modified moments by formulas analogous to (5). It is why we define a more restricted family of modified moments that we call the generalized moments ν_l:

$$\nu_{2k-1}=\int_{-\infty}^{+\infty} P_k(E)P_{k-1}(E)n(E)dE \tag{14a}$$

and

$$\nu_{2k}=\int_{-\infty}^{+\infty}P_k(E)P_k(E)n(E)dE\ , \tag{14b}$$

where $P_k(E)$ is a polynomial of degree k.

The generalized moments of even order are positive by construction, like the power moments to which they reduce when $P_k(E)=E^k$. Moreover, they can be calculated by formulas analogous to (5′):

$$\nu_{2k-1}=\sum_i\langle 0\,|\,P_k(H)\,|\,i\,\rangle\langle i\,|\,P_{k-1}(H)\,|\,0\rangle\ . \tag{15}$$

In order to limit the memory storage to two vectors $\langle 0\,|\,P_{k-1}(H)\,|\,i\,\rangle$ and $\langle 0\,|\,P_k(H)\,|\,i\,\rangle$, as in the case of the power moments, we assume a three-term recursion relation on the P_k's, i.e.,

$$P_{k+1}(H)=(H-c_{k+1})P_k(H)-d_kP_{k-1}(H),\quad k=0,1,2,\ldots \tag{16}$$

with the initial conditions

$$P_{-1}(H)=0,\quad P_0(H)=I\ , \tag{16′}$$

where I is the identity operator. The polynomials $P_k(H)$ are in addition monic, i.e., the coefficient of H^k is unity.

Let us recall that most of the known families of polynomials satisfy a three-term recursion relation. For example, the Chebyshev polynomials of second kind are defined by $c_k=0$ and $d_k=\frac{1}{4}$, the Hermite polynomials by $c_k=0$ and $d_k=k/2$, the Legendre polynomials by $c_k=0$ and

$$d_k=k^2(2k-1)^{-1}(2k+1)^{-1}\ .$$

An extensive list can be found in Ref. 18. The vectors $\langle 0\,|\,P_l(H)\,|\,i\,\rangle$ are generated by a recursion relation deduced from (16):

$$\begin{aligned}\langle 0\,|\,P_{k+1}(H)\,|\,i\,\rangle=&\sum_j\langle 0\,|\,P_k(H)\,|\,j\,\rangle\langle j\,|\,H\,|\,i\,\rangle\\&-c_{k+1}\langle 0\,|\,P_k(H)\,|\,i\,\rangle\\&-d_k\langle 0\,|\,P_{k-1}(H)\,|\,i\,\rangle\ .\end{aligned} \tag{17}$$

The coefficients c_k and d_k can be chosen arbitrarily but their choice is crucial for the numerical stability of the method. In practice, as we shall see below, one takes a polynomial sequence which is close to the polynomial sequence orthogonal with respect to the density of states $n(E)$. This idea will be made clearer in the section devoted to the illustrations of the method.

The second step of the calculation is the determination of the continued fraction coefficients from the generalized moments ν_k. This step follows exactly the same procedure as that used for the power-moments method. Defining the Gram matrix G_k by

$$(G_k)_{ij}=\int_{-\infty}^{+\infty}P_i(E)P_j(E)n(E)dE\ ,\quad 0\le i\text{ and }j\le k \tag{18}$$

and employing the expansion

$$P_i(E)=\sum_{l=0}^{i}C_{il}E^l,\ \ l\le i,\ C_{ii}=1 \tag{19}$$

yields

$$G_k=CH_k\widetilde{C}\ , \tag{20}$$

where H_k is the Hankel matrix given by Eq. (9). Since C is a lower triangular matrix with diagonal elements (and hence a determinant) equal to unity, we obtain

$$\det(G_k)=\det(H_k)=\Delta_k\ . \tag{21}$$

Similar arguments applied to the Gram matrix $G_k^{(1)}$ defined by

$$(G_k^{(1)})_{ij}=\int_{-\infty}^{+\infty}EP_i(E)P_j(E)n(E)dE\ ,\quad 0\le i\text{ and }j\le k \tag{22}$$

yield

$$\det(G_k^{(1)})=\Delta_k^{(1)}\ . \tag{23}$$

Hence, in (9) and (10) the replacement $\mu_k\rightarrow\nu_k$ may be employed.

If the polynomials $P_i(E)$ are orthogonal with respect to $n(E)$ (in the case $c_k=a_k$ and $d_k=b_k$), the Gram matrix is obviously diagonal:

$$G_k=\mathrm{diag}(1,b_1,b_1b_2,\ldots,b_1b_2\cdots b_k)\ , \tag{24}$$

and (12) is an identity. In addition, the matrix $G_k^{(1)}$ is tridiagonal and symmetric

$$G_k^{(1)}=\begin{bmatrix} a_1 & b_1 & & & \\ b_1 & a_2b_1 & b_1b_2 & & \\ & b_1b_2 & a_3b_1b_2 & b_1b_2b_3 & \\ & & \ddots & & \ddots \\ & & & b_1b_2\cdots b_k & a_kb_1b_2\cdots b_k \end{bmatrix} . \tag{25}$$

The determinant of $G_k^{(1)}$ is the product of a Hankel determinant and an orthogonal polynomial calculated at $E=0$, i.e.,

$$\det(G_k^{(1)})=\Delta_{k-1}P_k(0) .$$

The calculation of the determinants of G_k and $G_k^{(1)}$ given by (24) and (25) is obviously well conditioned in opposition to the calculation of Δ_k and $\Delta_k^{(1)}$. In practice, formulas (11) and (12) are not useful. The a_k and b_k coefficient are computed recursively from the generalized moments.

The closer one is to orthogonal polynomials, the better conditioned is the algorithm of calculation of a_k and b_k. This stability of the calculation of a_k and b_k from the generalized moments is identical to the stability of the calculation from the modified moments that have been discussed previously in the literature.[16,19–22]

Another way to express that consists of looking for the minimum value of the generalized moments (the nonredundant contribution to the moments[13]): If we formally differentiate ν_{2k} relatively to the polynomial $P_k(E)$ orthogonal with respect to $n(E)$ we obtain

$$\begin{aligned}\frac{\delta\nu_{2k}}{\delta P_k}&=\frac{\delta}{\delta P_k}\int_{-\infty}^{+\infty}P_k^2(E)n(E)dE\\ &=2\int_{-\infty}^{+\infty}P_k(E)n(E)dE=0 ,\end{aligned} \tag{26}$$

and

$$\nu_{2k}=b_1b_2\cdots b_k . \tag{27}$$

Consequently, the minimum value of the generalized moment of even order corresponds to the orthogonal polynomials (*a priori* unknown). This situation corresponds to the recursion scheme of Haydock *et al.*,[3] which is consequently the most stable algorithm, but as the coefficients c_k and d_k are self-consistently determined in the course of the calculations, the corresponding generalized moments are no longer linear on the density of states (in fact $c_k=a_k$ and $d_k=b_k$ which are themselves functions of the density of states).

IV. EXAMPLES OF GENERALIZED MOMENTS AND APPLICATIONS OF THE METHOD

In order to shed some light on the properties of the generalized moments, let us first consider the problem of the electronic states of an ordered, one-dimensional crystal described in the tight-binding approximation as a linear chain of atoms with nearest-neighbor interactions only. The electronic density of states of this system is given by

$$n(E)=\frac{1}{\pi}(1-E^2)^{-1/2} , \tag{28}$$

by assuming that the origin of the energies coincides with the diagonal elements ϵ_i of the Hamiltonian (4) and the unit of energy is equal to half the bandwidth (in these units, the hopping integrals β_{ij} are equal to $\frac{1}{2}$). The coefficients a_k and b_k of the continued fraction (8) related to the density of states (28) are

$$\begin{aligned}&a_k=0,\ \ k=1,2,\ldots\\ &b_1=\tfrac{1}{2},\ \ b_k=\tfrac{1}{4},\ \ k=2,3,\ldots .\end{aligned} \tag{29}$$

In order to show how the generalized moments depend on the polynomial sequence $P_k(E)$ chosen for their calculation, let us consider the recursion equation (16) with coefficients c_k and d_k proportional to the a_k and b_k coefficients given by Eq. (29). More precisely, we take

$$c_k=0,\ \ d_k=\delta^2b_k,\ \ k=1,2,\ldots \tag{30}$$

where δ is a real parameter. When $\delta=0$, the generalized moments are the classical power moments, while when $\delta=1$, the moments are minimal.

The polynomials $P_k(E)$ defined in this way are

$$P_k(E)=2\left[\frac{\delta}{2}\right]^kT_k\left[\frac{E}{\delta}\right],\ \ k=1,2,\ldots \tag{31}$$

where $T_k(x)$ are the Chebyshev polynomials of the first kind. The generalized moments of the density of states (28), computed from their definition (14) using the polynomials (31), vanish for odd order

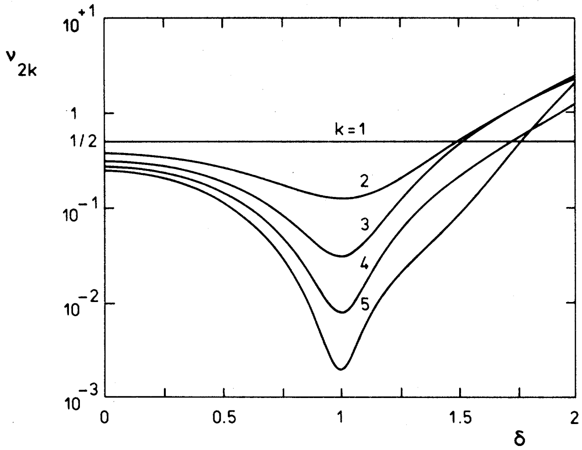


FIG. 1. Variation of the first generalized moments of a perfect linear chain of atoms computed from the polynomial sequence defined from Eq. (31). The generalized moments of even order are minimal for $\delta=1$, when the polynomials $P_k(E)$ are orthogonal with respect to the density of states.

and are positive for even order. When $\delta=1$, the polynomials (31) are orthogonal with respect to the density of states (28) and the corresponding generalized moments of even order ν_{2k} are simply given by the product of the b_k coefficients [Eq. (27)], i.e.,

$$\nu_{2k}=(\tfrac{1}{2})^{2k-1} . \tag{32}$$

The first generalized moments are plotted in Fig. 1 as functions of the parameter δ. The curves clearly have a minimum at $\delta=1$, the minimum values being given by Eq. (32). When $\delta\neq 1$, analytical expressions are difficult to give in a closed form, except when $\delta=0$ where we have the classical power moment of the linear chain

$$\nu_{2k}=\frac{(2k)!}{(2^k k!)^2} . \tag{33}$$

The redundant information that characterizes the power moments may be evaluated from the ratio of the minimum moments, given by Eq. (32), and the μ_{2k}, i.e.,

$$2\frac{(k!)^2}{(2k)!}\sim\frac{\sqrt{4\pi k}}{2^{2k}} \quad \text{as } k\to\infty , \tag{34}$$

a result which has to be compared with Eq. (7). As Fig. 1 shows, the use of carefully chosen generalized moments instead of power moments reduce the redundant information: In this example this happens in the neighborhood of $\delta=1$.

Secondly, we apply the generalized-moments method to the case of perfect fcc crystal. The hopping integrals β_{ij} of the tight-binding Hamiltonian (4) are set equal to -1 when i and j are nearest neighbors and 0 in other cases. In these units, the bandwidth W is equal to 16. In other words, the coefficients b_k of the continued fraction (8) converge towards $(W/4)^2=16$ as k tends to infinity. If the zero of energy coincides with the atomic levels ϵ_i, the electronic band is centered at the energy -4 and the asymptotic value of the a_k coefficients is equal to that value. In order to compute the a_k and b_k coefficients of the continued fraction (8), we have applied the generalized-moments method, taking as "trial" coefficients the values

$$\begin{aligned} c_1&=a_1=\langle 0|H|0\rangle=0 ,\\ d_1&=b_1=\langle 0|H^2|0\rangle-a_1^2=12 , \end{aligned} \tag{35}$$

and

$$c_k=a_\infty=0,\quad d_k=b_\infty=16,\quad k=2,3,\ldots . \tag{36}$$

An alternative choice, comparatively successful, would have been the Bethe-lattice coefficients [with coordination (12)]. The results we have obtained are presented in Table I together with the values of the a_k and b_k coefficients we have computed using the standard recursion scheme.[3] All these calculations have been performed on an IBM 370-158 computer using a single-precision arithmetics (32 bits). Table I shows that the generalized-moments method is numerically well conditioned, which gives a numerical precision as good as the recursion method. The computation time of the recursion algorithm is 10% higher than for the calculation of the generalized moments. This is essentially owing to the normalization of the basis in the recursion method (let us say that the normalization is not essential in the recursion; without this operation, both the recursion and the generalized-moments methods are characterized by the same computation time).

As a third application of the generalized-moments method, we have considered a disordered system known as the bond percolation problem in a simple cubic lattice. The tight-binding Hamiltonian (4) of this system is characterized by diagonal elements ϵ_i equal to zero, while the hopping integral β_{ij} between two nearest-neighbor lattice sites are randomly set equal to 1 or 0 with probabilities p and $1-p$. It is known from percolation theory

TABLE I. Coefficients a_k and b_k of the continued fraction expansion of the diagonal elements of the resolvent operator in a perfect fcc crystal. The coefficients have been computed using the recursion scheme (Ref. 3) and the generalized-moments method as described in the text. The exact results are given for comparison. The computation time (in seconds) is also given.

a_k	Recursion	Generalized moments	Exact results
1	0	0	0
2	−4	−4	−4
3	−3.764 71	−3.764 71	−3.764 705 8
4	−4.039 82	−4.039 82	−4.039 821 3
5	−4.015 19	−4.015 19	−4.015 191 2
6	−3.932 69	−3.932 69	−3.932 687 5
7	−4.015 84	−4.015 85	−4.015 845 1
8	−4.008 95	−4.008 96	−4.008 958 3
9	−3.965 70	−3.965 70	−3.965 696 7
10	−4.009 36	−4.009 36	−4.009 361 5
11	−4.006 06	−4.006 07	−4.006 064 3
12	−3.978 47	−3.978 47	−3.978 474 4
13	−4.006 42	−4.006 43	−4.006 428 3
14	−4.004 46	−4.004 46	−4.004 463 7
15	−3.984 93	−3.984 94	−3.984 931 6
b_k			
1	12	12	12
2	16.999 98	17.000 00	17
3	16.356 38	16.356 40	16.356 401 3
4	15.725 07	15.725 06	15.725 065 1
5	16.201 83	16.201 84	16.201 853 3
6	16.124 48	16.124 50	16.124 506 7
7	15.875 10	15.875 12	15.875 120 1
8	16.093 89	16.093 93	16.093 939 8
9	16.066 44	16.066 47	16.066 481 6
10	15.924 22	15.924 26	15.924 259 2
11	16.056 27	16.056 34	16.056 340 0
12	16.042 42	16.042 47	16.042 476 0
13	15.947 59	15.947 64	15.947 637 9
14	16.038 28	16.038 35	16.038 351 0
15	16.029 92	16.029 95	16.029 981 5
Time	8.9 s	8.1 s	

that the critical probability for the bond percolation problem in a simple cubic lattice is equal[23] to 0.247 (i.e., for $p > 0.247$, a cluster of infinite size has a nonvanishing probability). We determine the averaged density of states of this system. For this purpose, a large number N of cluster configurations have been simulated on the computer using a standard pseudo-random-numbers generator. For each cluster, the first generalized moments have been computed. The generalized moments have been averaged over the N configurations and the averaged density of states has been computed from the averaged moment using the continued fraction expansion given by Eq. (8). In that continued fraction, the a_k coefficients are equal to zero. The first b_k coefficients may be computed analytically to give

$$b_1 = 6p \, , \tag{37}$$

$$b_2 = 1 + 4p + 4p^3 \, , \tag{38}$$

$$b_3=\frac{10p+15p^2+16p^3+16p^4+44p^5-16p^6}{1+4p+4p^3}\ . \tag{39}$$

When localization occurs at the center of the band ($E=0$), it is not difficult to show that the weight of the localized state is given by

$$w=\left[1+\frac{b_1}{b_2}+\frac{b_1b_3}{b_2b_4}+\frac{b_1b_3b_5}{b_2b_4b_6}+\cdots\right]^{-1}\ . \tag{40}$$

Then, at the limit of small p, Eqs. (37)–(40) yield

$$w\sim 1-6p \quad \text{as } p\to 0\ . \tag{41}$$

We do not dwell any longer on the interesting but difficult problem of localization[24,25] in this system but just present the result we have obtained for $p=0.25$, a value very close to the critical probability. For each of the N configurations, 34 generalized moments have been computed by taking $c_k=0$ for $k=1,2,\ldots,17$, d_k equal to the corresponding b_k coefficients of the averaged density of state for $k=1,2,3$ [Eqs. (37)–(39)] and $d_k=2$ for $k\geq 4$. The density of state is presented in Fig. 2 when averaged over $N=2000$ configurations. It shows a strong peak (possibly a localized state) at $E=0$ and two satellite peaks at $E=\pm 1$. The density of states then decreases smoothly when the absolute value of the energy increases and vanishes for $|E|$ greater than 3.

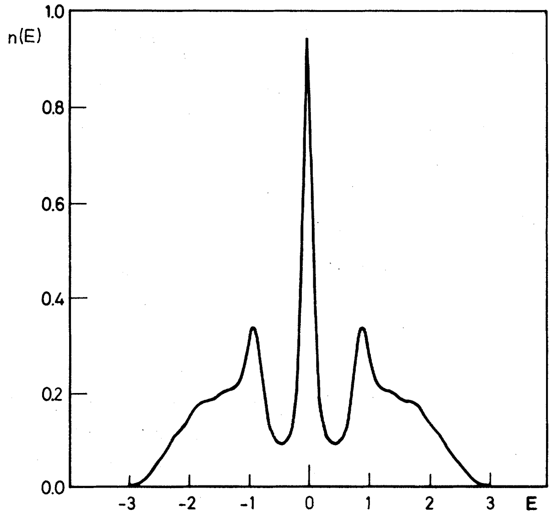


FIG. 2. Density of states of a simple cubic lattice with 75% of broken bonds. The density of states has been averaged over 2000 configurations using the linearity of the generalized moment with respect to the density of states.

V. CONCLUSION

The generalized-moments method improves the stability of the computation of the coefficients a_k and b_k of the continued fraction expansion (8) of the resolvent operator. This has already been shown by numerous authors in the framework of the so-called modified moments defined by Eq. (13). Our generalized moments are special modified moments which may be computed directly from the Hamiltonian of tight-binding systems. This is a crucial improvement of the modified-moments method of Modrak and Jozwiak.[22] The generalized-moments method gives rise to a continuous interpolation between two classical techniques, namely the recursion and the (power) moments methods. Consequently, it keeps the linearity of the moments method (which allows us to average the moments and then calculate the continued fraction coefficients associated with the averaged density of states) and gives a stability which may be comparable with that of the recursion method.

There are no general rules for the choice of the polynomial sequence $P_k(E)$ to which generalized moments are related. As we have said above, this polynomial sequence must be taken "close enough" to the polynomial sequence orthogonal with respect to the density of states which has to be computed, in order to ensure the numerical stability of the method. Let us recall that the polynomials $P_k(E)$ are given by the set of recursion coefficients c_k and d_k [Eq. (16)] which are input coefficients of the method. For the low-order input coefficients one can take the first continued fraction coefficients a_k and b_k of the density of states (in general, it is easy to calculate at least the a_1 and b_1 coefficients). On the other hand, some information may be obtained on the band limits and, consequently, on the asymptotic values of the a_k and b_k coefficients. These values may be used as high-order coefficients c_k and d_k as we have done in Sec. IV. It should be outlined that a perturbative aspect of the generalized-moments method has not been investigated along the present paper. Indeed a tight-binding Hamiltonian is very often used—because of its simplicity—for the determination of the electronic properties of crystalline defects (impurity, surface, etc.). For this kind of application, a natural choice for the input coefficients seems to be the continued-fraction coefficients related to the nonperturbed crystal.

Note added in proof. Recently Magnus pointed out to us that the asymptotic behavior of the coefficients a_k and b_k may also be related to the internal singularities of the band (if any) in addition to the band-edge singularities. An interesting feature of the internal singularities is that they give rise to an additional term in the k^{-1} expansion of Eqs. (A10) and (A14), which is an oscillating function of k multiplied by k^{-1}. This phenomenon is clearly shown in Fig. 3 for the bcc density of states which has a logarithmic peak at the center of the band: An oscillation of period two is found. The same kind of oscillations exists for the quantities ν_{2k} related to the honeycomb or the diamond s-band density of states (not analyzed in the present paper) which is characterized by a zero gap at the center of the band.

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APPENDIX

This appendix is devoted to a study of the asymptotic behavior of the coefficients a_k and b_k of the continued fraction (8). This may serve as a guide for the choice of the input coefficients c_k and d_k giving rise to the generalized moments. We investigate the case where the a_k and b_k coefficients have bounded and single limiting values a_∞ and b_∞ as k tends to infinity. In these conditions, the allowed energy band extends continuously between finite limits E_l and E_u related to a_∞ and b_∞ by the expressions

$$a_\infty = C = (E_u + E_l)/2 \qquad \text{(A1)}$$

and

$$b_\infty = \left[\frac{W}{4} \right]^2 = \left[\frac{E_u - E_l}{4} \right]^2 , \qquad \text{(A2)}$$

where C is the (geometrical) center of the band and W is the bandwidth. We shall try to relate the asymptotic behavior of the continued-fraction coefficients to the band-edge exponents assuming the density of states to be like

$$n(E) \propto (E - E_l)^\alpha \qquad \text{(A3)}$$

for E close to the lower band limit E_l, and

$$n(E) \propto (E_u - E)^\beta \qquad \text{(A4)}$$

when E is close to the upper band limit E_u. In that framework, reference will be made to the analytical model density of states

$$n(E) = \frac{\Gamma(\alpha+\beta+2)}{\Gamma(\alpha+1)\Gamma(\beta+1)} W^{-1} \left[\frac{E - E_l}{W} \right]^\alpha \times \left[\frac{E_u - E}{W} \right]^\beta , \qquad \text{(A5)}$$

where α and β are real exponents greater than -1 for integrability reasons.

There are numerous tight-binding systems which belong to the class of single and bounded limiting values for the a_k and b_k coefficients. The following tables give the continued-fraction coefficients related to the density of states of the most simple tight-binding Hamiltonian (4) when the atomic levels ϵ_i are equal to zero and the hopping integrals β_{ij} are set equal to 1 when i and j label nearest-neighbor lattice sites and equal to 0 in the other cases. Table II is devoted to some loose-packed bulk lattices. There are no triangular paths on these lattices so that the a_k coefficients vanish. The related densities of states are even functions of the energy and extend from $-Z$ to $+Z$, where Z is the coordination number. In Table III, the b_k coefficients are given for the local density of states on two kinds of surface planes in a cleaved simple cubic lattice. Two- and three-dimensional closed-packed lattices have been considered for Table IV; the band extends from -3 to $+6$ for the triangular (or hexagonal) lattice and from -4 to $+12$ for the fcc and hcp structures.

Let us now consider the minimum generalized moments

$$\nu_{2k} = b_1 b_2 \cdots b_k = \int_{E_l}^{E_u} P_k(E) P_k(E) n(E)\, dE , \qquad \text{(A6)}$$

where $P_k(E)$, $k = 0,1,2\ldots$ is the monic polynomial sequence orthogonal with respect to the density of states $n(E)$. For the particular model density of states (A5), the $P_k(E)$ are related to the Jacobi polynomials and the minimum generalized moments of even order are given by

$$\nu_{2k} = \frac{k!(\alpha+1)_k(\beta+1)_k(\alpha+\beta+2)_{k-1}}{[(\alpha+\beta+2)_{2k-1}]^2(\alpha+\beta+2k+1)} W^{2k} \qquad \text{(A7)}$$

TABLE II. Coefficient b_k of the continued fraction related to the tight-binding Hamiltonian (4) for quadratic, sc, and bcc lattices. Corresponding a_k coefficients are equal to zero.

k	Quadratic	sc	bcc
1	4	6	8
2	5	9	19
3	3.8	9.444 444 444 4	14.263 157 894 7
4	4.305 263 157 9	8.555 555 555 6	17.316 177 898 6
5	3.872 731 952 1	9.297 020 626 4	15.008 264 166 3
6	4.169 205 421 6	8.899 991 649 9	16.834 387 813 7
7	3.908 537 161 1	8.942 267 870 9	15.312 116 847 8
8	4.114 165 178 8	9.140 569 385 3	16.606 154 962 0
9	3.929 248 280 6	8.857 375 016 7	15.476 604 461 0
10	4.085 034 411 9	9.089 609 073 3	16.473 570 243 2
11	3.942 616 943 3	8.986 120 431 5	15.579 337 086 6
12	4.067 218 458 7	8.950 324 641 2	16.387 218 225 3
13	3.951 910 047 8	9.079 828 227 6	15.649 389 241 7
14	4.055 285 772 3	8.928 753 130 7	16.326 664 620 7
15	3.958 721 038 9	9.035 674 450 8	15.700 099 899 5
16	4.046 777 732 4	9.008 318 252 4	16.281 940 629 3
17	3.963 914 245 8	8.958 485 338 7	15.738 439 630 4
18	4.040 427 828 3	9.052 494 870 4	16.247 611 057 3
19	3.967 997 182 6	8.960 066 263 2	15.768 400 694 5
20	4.035 520 523 4	9.012 719 947 7	16.220 464 137 6
∞	4.000 000 000 0	9.000 000 000 0	16.000 000 000 0

TABLE III. Continued-fraction coefficients b_k related to the surface density of states for (110) and (100) cleaved simple cubic lattices.

k	(110)	(100)
1	4	5
2	6.5	7.4
3	8.423 076 923 1	8.005 405 405 4
4	7.926 238 145 4	8.318 429 841 0
5	8.619 443 240 5	8.579 263 530 5
6	8.705 903 025 5	8.632 475 684 9
7	8.551 570 159 6	8.749 832 434 5
8	8.947 513 214 2	8.790 760 359 9
9	8.719 888 245 3	8.821 944 473 4
10	8.861 542 143 7	8.868 270 201 7
11	8.939 106 240 9	8.873 018 699 4
12	8.798 511 007 0	8.901 258 316 9
13	8.990 484 174 8	8.912 657 795 8
14	8.882 910 388 3	8.919 314 575 4
15	8.915 071 925 8	8.936 294 158 8
16	8.989 947 216 2	8.936 718 964 3
17	8.882 851 746 2	8.947 105 651 8
18	8.994 730 288 2	8.952 590 518 3
19	8.944 065 478 8	8.954 121 755 4
20	8.936 817 723 5	8.962 439 825 0
∞	9.000 000 000 0	9.000 000 000 0

TABLE IV. Coefficients a_k and b_k of the continued fraction related to the tight-binding Hamiltonian (4) for triangular and fcc (and hcp) lattices.

	Triangular		fcc and hcp	
k	a_k	b_k	a_k	b_k
1	0	6	0	12
2	2	5	4	17
3	1.6	5.24	3.764 705 882 4	16.356 401 384 1
4	1.438 167 938 9	5.342 054 658 8	4.039 821 301 9	15.725 065 182 6
5	1.437 970 593 9	5.170 674 153 7	4.015 191 207 4	16.201 853 359 5
6	1.517 849 703 6	5.005 336 085 7	3.932 687 525 3	16.124 506 722 1
7	1.546 977 349 7	5.017 715 897 2	4.015 845 169 8	15.875 120 174 3
8	1.507 691 403 0	5.121 249 871 0	4.008 958 340 2	16.093 939 876 2
9	1.469 467 997 6	5.146 433 411 1	3.965 696 705 8	16.066 481 682 3
10	1.483 714 440 4	5.070 470 962 1	4.009 361 586 6	15.924 259 236 9
11	1.518 011 278 4	5.018 575 145 9	4.006 064 376 8	16.056 340 056 0
12	1.520 260 162 9	5.053 534 671 2	3.978 474 452 1	16.042 476 041 4
13	1.493 658 381 9	5.108 254 724 4	4.006 428 368 1	15.947 637 944 4
14	1.480 873 032 8	5.098 510 019 7	4.004 463 704 4	16.038 351 080 8
15	1.498 260 972 9	5.047 323 667 6	3.984 931 691 8	16.029 981 576 2
16	1.515 636 439 3	5.034 202 796 6	4.004 788 508 5	15.960 952 751 1
17	1.507 893 416 9	5.071 489 187 8	4.003 468 836 0	16.028 175 476 4
18	1.489 637 709 6	5.097 472 604 5	3.988 713 338 8	16.022 548 714 2
19	1.489 105 640 7	5.074 012 178 3	4.003 755 931 4	15.969 400 772 9
20	1.504 946 353 5	5.041 536 418 0	4.002 799 830 9	16.021 784 147 3
∞	1.500 000 000 0	5.062 500 000 0	4.000 000 000 0	16.000 000 000 0

with the use of the Pochhammer's symbol. A simple calculation shows that the asymptotic behavior of this last expression is

$$\nu_{2k} \sim \frac{2\pi}{4^{\alpha+\beta+1}} \frac{\Gamma(\alpha+\beta+2)}{\Gamma(\alpha+1)\Gamma(\beta+1)} \times \left[1+\frac{\alpha^2+\beta^2-\frac{1}{2}}{2k}\right] \left[\frac{W}{4}\right]^{2k} \quad \text{as } k \to \infty \; . \tag{A8}$$

Let us come back to the general case. Because of the orthogonal properties of the polynomial $P_k(E)$, it is obvious that Eq. (A6) may be rewritten as

$$\nu_{2k} = \int_{E_l}^{E_u} (E-C)^k P_k(E) n(E)\, dE \; , \tag{A9}$$

where C is the center of the band. We explicitly assume that the a_k and b_k coefficients of the continued fraction related to $n(E)$ have single and bounded limiting values so that the set of all zeros of all polynomials $P_k(E)$ is dense in the interval (E_l, E_u). In these conditions, when k increases infinitely, the integral (A9) is only sensitive to the values taken by the density of states $n(E)$ in the vicinity of the band limits E_l and E_u for the following reasons. First, the absolute value of the factor $(E-C)^k$ is larger near the band limits than

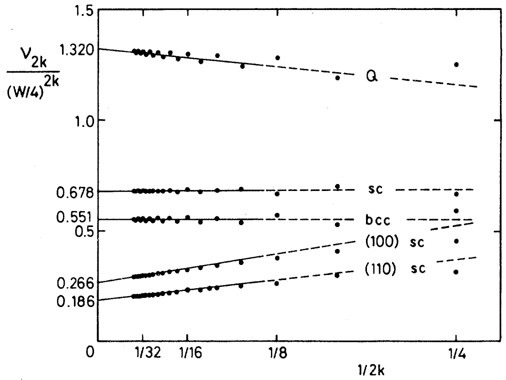

FIG. 3. Plot of $\nu_{2k}/(W/4)^{2k}$ vs $(2k)^{-1}$ for tight-binding densities of states related to the quadratic lattice (Q), the simple cubic (sc), and body-centered-cubic (bcc) lattices and at the (100) and (110) surface planes of the simple cubic lattice. Straight lines give the asymptotic behavior of the plots for $k \to \infty$ and are related to the band-edge components of the corresponding densities of states.

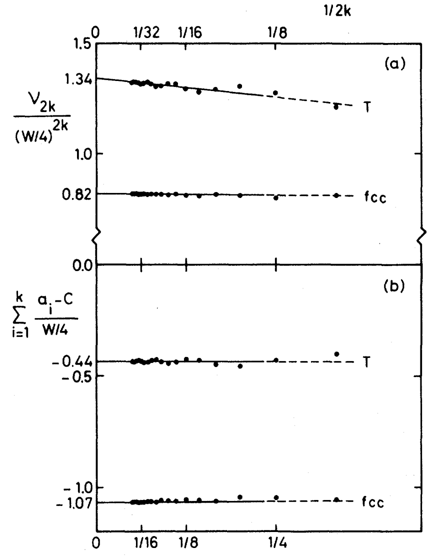

FIG. 4. (a) Plot of $\nu_{2k}/(W/4)^{2k}$ vs $(2k)^{-1}$ for tight-binding densities of states corresponding to the triangular (T) and face-centered-cubic (fcc) lattices. (b) Plot of $\sum_{i=1}^{k}(a_i - C)/(W/4)$ vs k^{-1} for the triangular (T) and fcc densities of states.

near the center of the band. Second, $P_k(E)$ has k real roots in the interval (E_l, E_u) so that $(E-C)^k P_k(E)$ is a rapid oscillating function of E between the lower and upper roots of $P_k(E)$, and that expression is positive at $E = E_l$ and $E = E_u$. From these arguments ν_{2k} asymptotically depends only on the comportment of $n(E)$ near the band edges. Assuming this comportment to be given by Eqs. (A3) and (A4), one may conclude from Eq. (A8) that

$$\nu_{2k} = b_1 b_2 \cdots b_k = B\left[1 + \frac{\alpha^2 + \beta^2 - \frac{1}{2}}{2k} + O(k^{-2})\right] \times \left[\frac{W}{4}\right]^{2k} \quad \text{as } k \to \infty , \qquad \text{(A10)}$$

where B is a positive constant. This equation shows that the first term in the k^{-1} expansion of $\nu_{2k}/(W/4)^{2k}$ vanishes when $|\alpha| = |\beta| = \frac{1}{2}$. This is clearly shown in Figs. 3 and 4(a) for three-dimensional densities of states which are characterized by square-root singularities at the band edges ($\alpha = \beta = \frac{1}{2}$). For surface densities of states, the asymptotic behavior of $\nu_{2k}/(W/4)^{2k}$ plotted in Fig. 3 has been found in complete agreement with the theoretical band-edge exponents $\alpha = \beta = \frac{3}{2}$. In the case of two-dimensional lattices too, agreement between Figs. 3, 4(a), and the theoretical asymptotic behavior given by Eq. (A10) has been found, taking $\alpha = \beta = 0$.

Equations (A9) and (A10) were related to the b_k coefficients only. In order to obtain another expression depending on the a_k coefficients, let us take the factor $(E-C)^{k+1}$ in the right-hand side of Eq. (A9) instead of the actual $(E-C)^k$ one. A simple recurrence demonstration shows that

$$\int_{E_l}^{E_u} (E-C)^{k+1} P_k(E) n(E)\, dE = \nu_{2k} \sum_{i=1}^{k} (a_i - C) . \qquad \text{(A11)}$$

For the model density of states (A5), one obtains

$$\sum_{i=1}^{k} (a_i - C) = \left[\alpha - \beta - \frac{\alpha^2 - \beta^2}{\alpha + \beta + 2k}\right] \frac{W}{4} \qquad \text{(A12)}$$

$$\sim \left[\alpha - \beta - \frac{\alpha^2 - \beta^2}{2k}\right] \frac{W}{4} \quad \text{as } k \to \infty . \qquad \text{(A13)}$$

In the general case, from the same arguments than that used above, one may conclude that expression (A11) is only sensitive to the values taken by $n(E)$ in the vicinity of the band limits and one gets from Eq. (A13),

$$\sum_{i=1}^{k} (a_i - C) = \left[A - \frac{\alpha^2 - \beta^2}{2k} + O(k^{-2})\right] \frac{W}{4} \quad \text{as } k \to \infty . \qquad \text{(A14)}$$

where A is a constant, α and β being the band-edge exponents of the density of states [Eqs. (A3) and (A4)]. That last equation is illustrated in Fig. 4(b) for closed-packed structures. The asymptotic behavior of the quantities $\sum_{i=1}^{k}(a_i - C)/(W/4)$ plotted in Fig. 4(b) as a function of $1/k$ are in agreement with Eq. (A14) taking $\alpha = \beta$.

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