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Abstract. Vibrational properties of quasi-lattices constructed by the Fibonacci substitution rule are studied using the integrated method for solving the eigenvalue problem of a tri-diagonal matrix. The 'Cantor-like' integrated density of states and its self-similar characteristics are given. The vibration eigenstates exhibit various interesting phenomena. We also examine the influence of some factors on the vibrational properties.

Following the discovery of quasi-crystals, some artificially fabricated one-dimensional quasi-crystals, such as GaAs–AlAs, Si–GeSi Fibonacci superlattices have been grown by molecular beam epitaxy (MBE) and have been investigated using x-ray and Raman scattering measurements [1, 2]. Many theoretical calculations of vibrational properties in one-dimensional Fibonacci chains have also been presented [3]. In these calculations, two force constant K_A and K_B are arranged according to the Fibonacci sequence and all masses are assumed to be identical. This system can be easily solved using transfer matrix techniques. The vibrational spectra and amplitudes show some very interesting characteristics. As is known for the periodic one-dimensional chain, vibrations with two masses m_A and m_B are different from those with two force constants K_A and K_B. In this paper, we consider a one-dimensional version of the quasi-crystal in which two masses—the masses of atom A and atom B—follow the Fibonacci sequence. Using the integrated method for solving the eigenvalue problem of a tri-diagonal matrix, which has been developed recently, we study the integrated density of states (IDOS) of quasi-crystal chains of various lengths and discuss their different eigenstates in detail. Some features are studied for the first time. We also examine the influence of various factors such as the atom number of the system, the ratio of the mass of atom A to the mass of atom B, the force constant between atoms, disorder in the usual atom arrangement, and doping by impurities on the vibrational properties.

The Fibonacci sequence ABAABAB . . . is defined by the production rules A → AB and B → A, with A as the first single element of the first generation. We find that there are certain symmetries in the Fibonacci sequence, i.e. the ith element is the same as the $(F_n - 1 - i)$th element, where F_n is the Fibonacci number of nth generation ($F_1 = 1$, $F_2 = 2, F_{n+1} = F_n + F_{n-1}$). The ratio of numbers of A elements to B elements tends to $\tau = (\sqrt{5} + 1)2$, the golden ratio. The Fibonacci sequence atomic chain has the following

vibration equation within the framework of nearest-neighbour interactions and the harmonic approximation:

$$m_n \, \mathrm{d}^2 X_n / \mathrm{d} t^2 = K_{n,n+1} X_{n+1} + K_{n,n-1} X_{n-1} - (K_{n,n+1} + K_{n,n-1}) X_n \tag{1}$$

where m_n is the mass of the nth atom, which is m_A or m_B according to its position in the Fibonacci sequence, X_n is the displacement of the nth atom from its equilibrium position and $K_{n,n\pm1}$ are the force constants coupling neighbouring atoms. Let $X_n(t) = U_n \exp(-i\omega^2 t)/m_n$ and divide both sides of equation (1) by m_n; then equation (1) becomes

$$-\omega^2 U_n = (K_{n,n+1}/\sqrt{m_{n+1} m_n}) U_{n+1} + (K_{n,n-1}/\sqrt{m_{n-1} m_n}) U_{n-1}$$
$$- [(K_{n,n+1} + K_{n,n-1})/m_n] U_n \qquad (n \neq 1, N). \tag{2}$$

Equation (2) can be rewritten in the form of a matrix as

$$\mathbf{H}|u\rangle = \omega^2 |u\rangle \tag{3}$$

where $\mathbf{H}$ is a symmetric tri-diagonal matrix, $|u\rangle$ is the eigenvector corresponding to the eigenfrequency ω^2. In the free-end boundary condition, we have

$$H_{11} = K_{12}/m_1 \qquad H_{NN} = K_{N,N-1}/m_N \tag{4}$$

where N is the total number of atoms. There are many methods of solving this kind of eigenvalue problem, but we have found that the integrated method for solving the eigenvalue problem is a simple and very efficient method. Here we only give the essential formulations for this method. The details of the derivation can be found in [4].

If $\eta(x)$ denotes the number of real negative eigenvalues of a matrix, then

$$\eta(H - \omega^2 I) = \sum_{n=1}^{N} \eta(U_n) = \sum_{n=1}^{N} \eta(V_n) \tag{5}$$

where

$$U_1 = H_{11} - \omega^2$$
$$U_n = H_{nn} - \omega^2 - H_{n,n-1} U_{n-1}^{-1} H_{n-1,n} \qquad (n = 2, \ldots, N)$$
$$V_N = H_{NN} - \omega^2$$
$$V_n = H_{nn} - \omega^2 - H_{n,n+1} V_{n+1}^{-1} H_{n+1,n} \qquad (n \leq N - 1).$$

Equation (5) states that the number of eigenvalues of the matrix $\mathbf{H}$ which are smaller than a real number ω^2 can be found from the signs of N scalar equations for $U_n(\omega^2)$ or $V_n(\omega^2)$. In our system, the IDOS is obtained easily from

$$D(\omega^2) = \eta(\mathbf{H} - \omega^2 I). \tag{6}$$

Using the trial-and-error method, the eigenvalues can be obtained in the following way. If we wish to obtain an eigenvalue which is greater than a certain value, we can choose a small interval; let

$$\Delta = \eta [\mathbf{H} - (\lambda + \Delta\lambda) I] - \eta [\mathbf{H} - \lambda I]. \tag{7}$$

If $\Delta < 1$, this means that there is no eigenvalue between λ and $\lambda + \Delta\lambda$ and, if $\Delta > 1$, there are more than one eigenvalue. So we must increase $\Delta\lambda$ or decrease $\Delta\lambda$ to make $\Delta = 1$. Thus, the eigenvalue must satisfy $\lambda < \omega^2 < \lambda + \Delta\lambda$. We can say that $\omega^2 = \lambda + \Delta\lambda/2$ so long as $\Delta\lambda$ is sufficiently small. Otherwise we make $\Delta\lambda_1 = \Delta\lambda/2$ and repeat
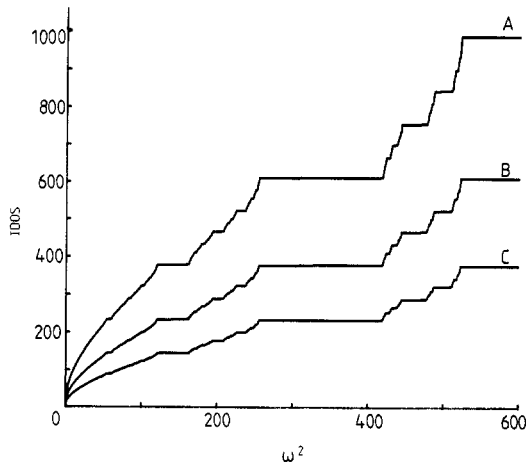
Figure 1. The IDOS for Fibonacci chains of A, 987; B, 610; and C, 377 atoms.

the above procedure. In this way we can obtain the eigenvalues of **H** to any desired degree of accuracy.

Now we calculate the eigenvectors. The eigenvector $|u\rangle$ can be expanded in terms of the basic vectors $\{|i\rangle\}$, i.e.

$$|u\rangle = \sum_{i=1}^{N} b_i |i\rangle \tag{8}$$

where $\{|i\rangle\}$ represent $(100\ldots)$, $(010\ldots)$, etc. Then the values of the functions

$$E_n = U_n + V_n - (M_{nn} - \omega^2) \qquad n = 1, \ldots, N \tag{9}$$

are calculated. We choose the minimum value E_l in a set of $\{E_n\}$; then we have

$$b_l = 1$$

$$b_{l-1} = -H_{l-1,l}/U_{l-1}$$

$$b_{l-r} = -H_{l-r,l-r+1}b_{l-r+1}/U_{l-r} \qquad r = 2 \ldots l-1 \tag{10}$$

$$b_{l+1} = -H_{l+1,l}/V_{l+1}$$

$$b_{l+r} = -H_{l+r,l+r-1}b_{l+r-1}/V_{l+r} \qquad r = 2 \ldots N-1.$$

Thus $|u\rangle$ can be found. In the process of determining the eigenvalue ω^2 of **H** the quantities $\{V_n\}$ and $\{U_n\}$ have already been calculated in equation (5). In this sense the determination of the eigenvector is almost accomplished simultaneously with the determination of the eigenvalue and they have the same accuracy. After normalising and considering the mass factor, we obtain the vibration amplitudes of the atom chain.

The IDOS for the systems with 377, 610 and 987 atoms, when $m_A = 1.0$, $m_B = 0.5$ and all $K_{n,n\pm1} = 1.0$, are displayed in figure 1. The numbers 377, 610 and 987 are the Fibonacci numbers F_n of the nth generation for $n = 12$, 13 and 14; one unit on the abscissa means $\omega^2 = 1.1$ for the convenience of calculation. These curves are essentially identical. They are 'Cantor like'. In the quasi-continuous region there are many small gaps, i.e. flat portions in spectra. It is remarkable that the gaps occur at the Fibonacci numbers
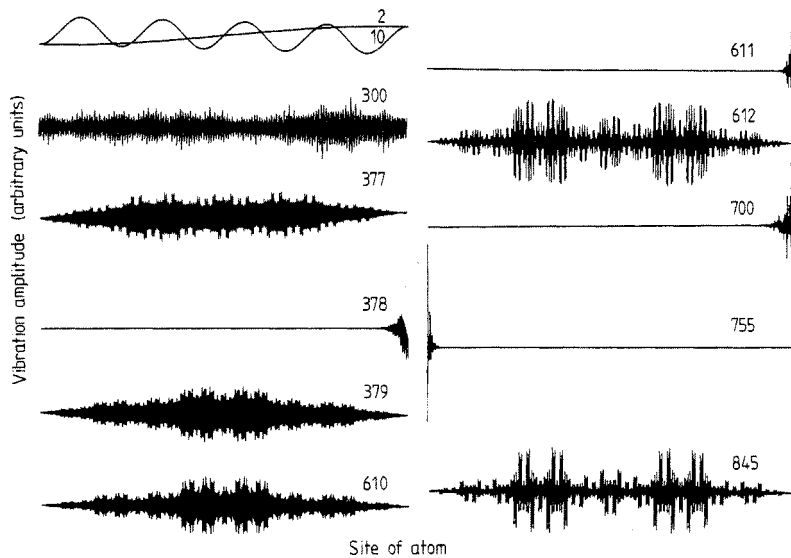
Figure 2. A few modes for a one-dimensional quasi-crystal chain of 987 atoms

and at their combinations. For example, the three larger gaps start at F_{i-2}, F_{i-1} and $F_{i-1} + F_{i-4}$ and end at $F_{i-2} + 1$, $F_{i-1} + 1$, $F_{i-1} + F_{i-4} + 1$ if the chain contains F_i atoms. There is only one state in the gap, which is called the gap state. Careful examination of our results shows that the spectra are self-similar, especially in the high-frequency region.

If the atom number in the chain is not a Fibonacci number, what is the integrated density? We examine chains which have 300, 600 and 800 atoms separately. The IDOSs have the same form as for chains with a Fibonacci number; gaps occur in some frequency regions but correspond to more complex combinations of Fibonacci numbers. So we can conclude that the characteristics of the IDOSs are determined from the former structure in chain; when the number of atoms increases, the eigenstates fill in the frequency band of spectra.

Figure 2 illustrates the amplitude of some eigenstates. The mode numbering is from the lower- to the higher-frequency mode. Modes 2 and 10 are eigenvectors corresponding to the low-frequency or long-wavelength portion of the spectra, which are affected little by the microscopic structure of the chain. They are similar to those in periodic systems. The modes 378, 611, 700 and 755, which are in the gap, are strongly localised modes. Their localised positions are different. In our case, except for mode 755 which is localised at the left-hand end of the chain, the other modes are localised at the right-hand end. As we know, the periodic lattice has an absolutely continuous spectrum and extended eigenstates, whereas the random lattice has a pure point spectrum and localised eigenstates. Now in our quasi-crystal system, besides the extended and localised states, there are also modes 300, 377, 379, 610, 612 and 845, which can be called critical states. The states at the gap edges are especially interesting. For example, the 377th, 610th and 845th states, which are located at the left-hand gap edges, are symmetric and the 379th and 612th states, which are located at the right-hand gap edges, are almost asymmetric. There is a dramatic change in the nature of the modes across the band edges. This character is similar to that in the periodic lattice to some degree. We also find that the number of local maxima in the non-gap state is equal to the mode numbering.

Now we consider some influences of various factors on eigenvectors. If $m_B = 0.7$ replaces $m_B = 0.5$ but m_A still equals 1.0, the maximum frequency decreases from 5.751 907 3 to 4.709 984 5; the eigenvalues correspondingly decrease, e.g. mode 377 eigenvalue changes from 1.336 515 4 to 1.310 237 9 and so on. If the force constant decreases, e.g. $K_{AA} = 1.0$, $K_{AB} = 0.7$, $K_{BB} = 0.5$ (here K_{AB} means the force constant between atom A and atom B), the frequency maximum changes to 4.167 319 7, 377th eigenvalue decreases to 0.942 127 4. The other characteristics of spectra do not vary in both the above conditions.

The influences of the length of the chain on the local states are also interesting. As we know, the left-hand structures of a quasi-crystal chain of different lengths are identical; so all gap states, which are localised at the left-hand end of the chain have nothing to do with the length of chain, even if the number of atoms in the chain is not a Fibonacci number, e.g. $N = 900$. Nevertheless, the local states, which are localised at the right-hand end of the chain containing F_n atoms are the same as those in the chain containing F_{n-2} atoms because the right-hand ends of the F_n and F_{n-2} sequences are the same; for example, for $\omega^2 = 1.367\,672\,839$, this state is localised in the 375th atom and nearby in the chain of 377 atoms, i.e. the vibrational amplitudes of atoms reach a maximum value in the last atom but one. If the chain contains 987 atoms, this state also localises the last atom but one and nearby.

If some disorder occurs, e.g. the 450th atom which is m_B according to the Fibonacci sequence, is replaced mistakenly by m_A, we found that eigenvalues of non-local states are slightly lower; this is because m_A is larger than m_B. The 378th and the 755th modes (the former is located at the right-hand end of chain and the latter at the left-hand end) do not vary, and the 611th mode, which is located at the right-hand end originally, is now located in the middle of the chain. The local 844th mode now becomes the 845th mode. When the disorder is at the local point of the local state, the local state can change to a non-local state. If the 449th, 450th and 451st atoms all change, the 377th mode has the same property as 378th mode in a regular Fibonacci chain. The 755th mode remains invariable. This mode can change when the second atom is changed to m_A. If the lighter atom, e.g., $m = 0.3$, replaces the 450th atom, all eigenfrequencies decrease. The IDOSS do not obviously change in all above conditions.

As we know, in the perfect diatomic chain, each unit cell has two atoms of masses m_A and m_B. Their vibration amplitudes are u_A and u_B; if $u_A/u_B > 0$ in a mode, we can say that this mode is acoustic, but otherwise it is optical. However, in the quasi-crystal chain, as there are many disordered systems, we cannot strictly distinguish between acoustic and optical branches. Each mode is a so-called mixed mode. We define $u'_n = \frac{1}{2}[u_n + \frac{1}{2}(u_{n+1} + u_{n-1})]$ and $u''_n = \frac{1}{2}[u_n - \frac{1}{2}(u_{n+1} + u_{n-1})]$. If $u'_n \simeq u_n$ and $u''_n \simeq 0$ for all n in a mode, this mode can be considered mainly acoustic; if $u'_n \simeq 0$ and $u''_n \simeq u_n$, it is mostly optical. We investigate a mode in our system and find that the mode is almost acoustic in the very-low-frequency region, e.g. in the 10th mode all $u''_n \simeq 0$. The acoustic component decreases and the optical component increases on increase in the vibration frequency. We do not consider how to determine the relative proportions of the acoustic component and the optical component in this paper.

Using the same method, we can easily study a perfect diatomic chain and a perfect monoatomic chain. We can also investigate the influences of impurities as has been studied in other work. Further comparison between the vibration spectra of periodic, disordered and quasi-crystal chains will be published elsewhere.

We also examine a large-Fibonacci-number atom chain, say $N = 17\,711$; the basic properties of vibration are the same as for $N = 987$. The CPU time for obtaining an

eigenvalue and an eigenvector is only 1 min 43 s using a Burroughs 6935. It is almost impossible to solve a 17 711 × 17 711 matrix using the standard method.

In summary, we have used a new method to study one large one-dimensional quasi-crystal which is a little more complex than a quasi-crystal only having different force constants. Because Raman scattering has been used to study longitudinal acoustic phonons propagating parallel to the growth axis in the quasi-periodic superlattices, our linear chain offers a semi-quantitative analysis model. Work is under way to study our model in more detail and to compare it directly with experimental results.

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