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On the nature of vibrations in diatomic lattices

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Abstract. The lattice dynamics of a one-dimensional diatomic lattice with nearest-neighbour interactions is examined with mathematical techniques involving the setting up and solving difference equations. The method gives the exact sets of analytical solutions for the normal modes of vibration. The theory of localised vibrational modes due to local imperfection is developed as a characteristic value problem. The approach seems to have advantages over previous methods used to solve local defect problems in lattices.

1. Introduction

The subject of the vibrations in both one-dimensional monotomic and diatomic lattices with nearest-neighbour interactions and with local imperfection has received considerable attention throughout the past few years. An enormous literature exists, which contains the different methods propounded to solve this lattice vibrational problem [1–25].

Recently a simple and effective method to examine the influence of defects in monatomic and diatomic lattices with nearest-neighbour, next-nearest-neighbour and more complex interactions, based on the solution of the difference equations, has been proposed [26]. This method shows how the lattice dynamics study draws naturally upon the operational method of solution for the difference equations, which gives the exact sets of solutions for the vibrational normal modes. Also an analytic method has been introduced to analyse the localised vibrational modes due to local imperfections as a characteristic value problem, which includes following the piecewise method and the matching conditions of the solution in the imperfection region. In this treatment simple formulae are obtained for the localised mode frequencies in an elementary and straightforward way.

The potential of a diatomic linear chain model with nearest-neighbour interactions to explain the optical absorption data in real crystals has been demonstrated by Lukovsky, Brodsky and Burstein ([27]; see also [23, 25] and others [28]; see also [16, 18]), when they investigated local and gap mode frequencies in alkali halides and semiconductors as different atomic substituents were incorporated in these crystals. Lukovsky *et al* used the local mode analysis given by Mazur *et al* [17] and their results are in excellent agreement with those obtained from the full three-dimensional lattice dynamics calculations. So the linear chain model of point masses with nearest-neighbour coupling and with local imperfection is a surprisingly good model for theoretically predicting the

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frequencies of the localised modes in real crystals. Thus, it is required to obtain the solution of this vibrational problem in a different form that is analytically closed following the finite difference method mentioned above. It is also necessary because this solution provides the mathematical basis to obtain the frequencies of the localised modes in a diatomic linear chain with different models of local imperfection. These findings can be compared with experimental results and are important because attempts to give an adequate and general theory meet great difficulties with other mathematical methods and crystal models [29]. The purpose of this paper is to present a more direct and intuitively obvious method to examine the lattice dynamics for a one-dimensional diatomic lattice with nearest-neighbour interactions via the standard mathematical techniques of solving finite difference equations [26, 30]. Section 2 starts by giving an explanation of the operational method of solving difference equations for a perfect diatomic chain, and by performance of the transfer operator the sets of the general analytical solution are obtained. In § 3 a theory for the localised vibrational modes due to local imperfection is developed as a characteristic value problem. In order to illustrate the method it is applied to two cases: first, the very well known problem of the effect of an isotopic mass; and secondly, the single anomalous force constant as local imperfections in the lattice are solved in an easy manner. The conclusions are given in § 4.

2. General solution for the diatomic linear lattice

In this section the operational method to solve the problem of a one-dimensional perfect diatomic lattice is proposed. It will be shown that the approach can give the exact solution, making it simpler to find the vibrational normal modes for the system. The time-independent equations of motion for this perfect diatomic lattice formed by equidistant atoms of alternating masses m and M and nearest-neighbour interactions are

$$(\mathscr{E} + 1)y_n + (x - 2)x_n = 0 \tag{1}$$

for particles of mass m, and

$$(2 - \varepsilon x)\mathscr{E}y_n - (\mathscr{E} + 1)x_n = 0 \tag{2}$$

for atoms of mass M. In equations (1) and (2), $x = m\omega^2/K$ and $\varepsilon = M/m$, and y_n and x_n denote the maximum displacements in the lattice of the particles of mass M and m, respectively. Also K is the force constant associated with central force interaction of nearest neighbours and ω indicates the angular frequency of the normal modes of vibration. It also was necessary to make use of the transfer operator $\mathscr{E}$ [26, 30–32]. The system of equations (1) and (2) form a set of two homogeneous, simultaneous, linear, difference equations of the second order with constant coefficients and will be solved by using operational methods. Expressions for y_n and x_n must be found in terms of ε and x. From (1) and (2) by elimination a final equation involving only one of the variables with its differences and successive values will be obtained. Integration will give the general value of that variable and the equations employed in the process of elimination will allow the expression of the other dependent variable by means of it. If the coefficients are constants then the symbols can simply be separated and one can effect the eliminations as if those symbols were algebraic. Thus, the equation derived for the variable y_n is

$$(\mathscr{E}^2 - \Omega\mathscr{E} + 1)y_n = 0 \tag{3}$$

where $\Omega = \varepsilon x^2 - 2(\varepsilon + 1)x + 2$. To integrate equation (3) the auxiliary (or charac-

teristic) equation of this difference equation is needed. The auxiliary equation is a quadratic algebraic equation. For present purposes, it is noteworthy that the form for the general solution of the pair (1) and (2) depends on the roots of the auxiliary algebraic equation. There are three cases: (*a*) the roots E_1 and E_2 are real numbers and unequal; (*b*) the two roots are real and equal; and (*c*) the two roots are complex numbers. These exhaust the possibilities for a quadratic equation.

In these cases the general solution of the couple (1) and (2) can be written for (*a*)

$$y_n = C_1E_1^n + C_2E_2^n \qquad x_n = [(E_{1,2} + 1)/(2 - x)]y_n \tag{4a}$$

where C_1 and C_2 are arbitrary constants. From the auxiliary algebraic equation it is clear that in this case $E_1E_2 = 1$. Since the difference equations (1) and (2) are defined over sets of consecutive integers, then the solution (4*a*) is a linear combination of sequences. Only some of the more important aspects of these solutions in terms of the roots E_i are mentioned: (i) if $|E_i| \leqslant 1$, then the sequence converges; (ii) if $|E_i| > 1$ then it diverges; (iii) if $-1 < E_i < 0$, it is a damped oscillation; and (iv) if $E_i < -1$ it oscillates infinitely. These types of solutions could be associated with localised vibrations in lattices with imperfections. Note from the auxiliary equation that E_1 and E_2 are real and unequal when $\Omega^2 > 4$ and hence we obtain two cases: $\omega > \omega_0$, where $\omega_0 = [2K(1/M + 1/m)]^{1/2}$ the top of the optical branch, and $\omega > (2K/M)^{1/2}$ and $\omega < (2K/m)^{1/2}$ when $M > m$, or $\omega > (2K/m)^{1/2}$ and $\omega < (2K/M)^{1/2}$ when $M < m$, the frequencies for unequal real roots are situated in the gap between the branches. From the derivation, it is obvious that the frequencies of the localised vibrations in a diatomic linear chain, i.e. the solution (4*a*) always lies above the top of the optical branch or in the forbidden gap between the branches of the perfect lattice.

The general solution in the case (*b*) when there is a repeated root is

$$y_n = (C_1 + C_2n)E_1^n \qquad x_n = [(E_1 + 1)/(2 - x)]y_n \tag{4b}$$

where C_1 and C_2 also are arbitrary constants. The behaviour of this solution is divergent if $|E_1| > 1$ (unless both C_1 and C_2 are 0) or if $|E_1| = 1$ (unless $C_2 = 0$). Now consider the case $|E_1| < 1$. Then it can easily be proved that the sequence nE_1^n converges to 0. Thus, if $|E_1| < 1$, the sequence solution also converges to 0. It is oscillatory if E_1 is negative. These solutions could be connected with vibrations whose frequencies are the edges of the branches.

The case (*c*) has two complex roots that appear as a pair of complex conjugates. To write the general solution, observe that the maximum atomic displacements of the normal modes of vibration are real numbers and hence require solutions which have real number values for all n values for which they are defined. Then it is possible to show that if C_1 and C_2 are complex, y_n and x_n are always real numbers. To prove this, all complex numbers E_1, E_2, C_1 and C_2 must be written in polar form. Thus using equation (4*a*) the following expression is obtained for the general solution:

$$y_n = C\cos(n\theta + D) \qquad x_n = -C[(\varepsilon x - 2)/(x - 2)]^{1/2}\cos[(n + \tfrac{1}{2})\theta + D] \tag{4c}$$

where C and D are arbitrary constants. The number θ in equation (4*c*) is determined in terms of x and ε and it is given by

$$mM\omega^4 - 2K(M + m)\omega^2 + 2K^2(1 - \cos\theta) = 0. \tag{5}$$

Equation (5) is the well known dispersion relation for the frequency in a perfect diatomic linear lattice with nearest-neighbour interactions. The functions $\cos(n\theta + D)$ and $\cos[(n + \frac{1}{2})\theta + D]$ in (4*c*) oscillate finitely, with the result that the first part of the

equation (4*c*) represents a wave propagating only through the particles of mass M, while the second represents a wave propagating only through those of mass m. The wavelengths and frequencies for a given disturbance are equal. The amplitudes of the two waves on the other hand are not equal. These particular solutions could be associated with vibrations in a perfect lattice or with the perturbed branch modes [18] (quasi-localised resonance behaviour). The shapes of the vibrational normal modes can be shown in the diatomic linear lattice using equation (4*c*), but this work for the perfect lattice is well known and a careful study of the atomic motion in this system has been given by many authors. The results are in accordance with those obtained by them and the reader is referred to the works of Brillouin [33], Hori [7], Dean [4] and Barker and Sievers [22] to complete this description.

Another point of interest is the frequency distribution function of the normal vibrations. This can be calculated from (5) for a diatomic lattice consisting of $2N$ particles for which $\theta(\theta = \pi j/N;\ j = \pm 1, \pm 2, \ldots, \pm N)$ is regarded as a continuous parameter and can be written in the form

$$\frac{\mathrm{d}j}{\mathrm{d}\nu} = \frac{N}{\pi\nu_0\varepsilon^{1/2}}\left(\frac{\pm(1-2\nu^2/\nu_0^2)}{(1-\nu^2/\nu_0^2)^{1/2}\{1-[(\varepsilon+1)^2/\varepsilon](\nu^2/\nu_0^2)(1-\nu^2/\nu_0^2)\}^{1/2}}\right). \tag{6}$$

In (6) ν is the frequency of the normal modes of vibration ($\omega = 2\pi\nu$). For $\nu \ll \nu_0$ this density of modes reduces to

$$\frac{\mathrm{d}j}{\mathrm{d}\nu} = \frac{N(M+m)}{\pi\nu_0(mM)^{1/2}}.$$

This frequency distribution function had already been obtained by Dean [4] and Blackman [34].

The general solution, equations (4*a*), (4*b*) and (4*c*), contains the exact sets of analytic solutions for the vibrational normal modes in a diatomic linear lattice with nearest-neighbour interactions. It is striking that this approach does not involve heuristic solutions to obtain the general solution of the couple of equations (1) and (2) as had been done in all of the calculations mentioned previously [1–25].

3. Theory for localised vibrational modes

To illustrate the difference equations method when local imperfection is introduced in the lattice, two simple models will be considered, in which the effect of a single localised defect on the ordered diatomic lattice can be easily analysed. It will be proposed then to obtain the solution of this problem as a characteristic value problem.

3.1. Mass defect

The first model of a local defect is a mass defect in the lattice. Some literature exists which contains discussions concerning the effect of a single impurity mass on the vibrations in a diatomic linear lattice with nearest-neighbour interactions [2, 6, 17, 22, 35–38]. It is quite significant that in all the calculations mentioned above the mathematical methods contain unnecessary complications, and therefore a more accessible and naive method is presented to solve this localised vibrational mode problem.

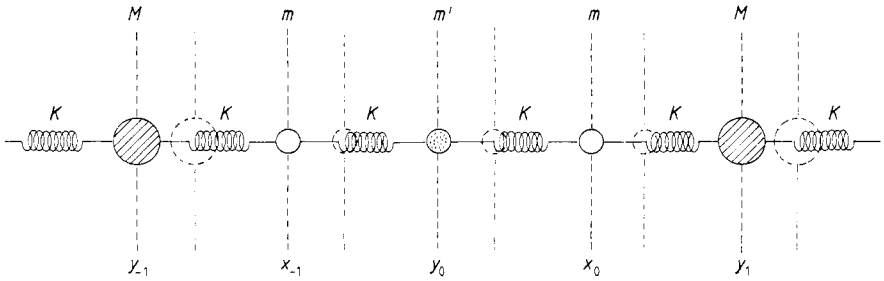

Figure 1. Diatomic linear lattice with a local imperfection, a mass defect m' situated at site y_0.

In order to obtain the solution for the problem of the effect of a single impurity mass on the vibrational normal modes, briefly here as a boundary value problem, the atom of mass M is replaced at site y_0 by one of mass m'. The diatomic lattice with local imperfection is sketched in figure 1.

The condition of localisation for each part of the solution around defect $y_n \to 0$ and $x_n \to 0$ as $n \to \pm\infty$ prescribes the boundary conditions and the boundary value problem is posed. All solutions of the couple of equations (1) and (2) that simultaneously satisfy the boundary conditions must be found. One solution is given by the trivial solution but the interest is on the possibility of finding non-trivial solutions of the characteristic value problem.

Now, a piecewise solution will be introduced and the interval in which the pair of equations (1) and (2) of the perfect lattice are valid will be split into two parts: the set $-\infty \leqslant n < 0$ and the set $0 < n \leqslant \infty$. The problem of setting up the solution for localised vibrations can be viewed as a question of matching the allowed solution on the two sides of the local imperfection. In each part, the general solution associated with the set of localised modes of the couple (1) and (2) is the equation (4*a*). Clearly, the boundary conditions, i.e. the localisation condition of the solution around the defect, can be used in order to determine two of the coefficients. In the region $-\infty \leqslant n < 0$, the only sequence that is permitted if $y_n^{(2)}$ and $x_n^{(2)}$ are not increased without limit as $n \to -\infty$ is the sequence that converges. This determines one of the constants to be zero and in this region

$$\begin{aligned} y_n^{(2)} &= C_2 E_{1,2}^{-n} && n < 0 \\ x_n^{(2)} &= C_2[(E_{1,2}+1)/(2-x)]E_{1,2}^{-(n+1)} && n \leqslant -1. \end{aligned} \tag{7}$$

In (7) $E_1 E_2 = 1$ was used. Similarly, in the region to the right, $0 < n \leqslant \infty$, $y_n^{(1)}$ and $x_n^{(1)}$ satisfy equations (1) and (2) so the solution has the form

$$\begin{aligned} y_n^{(1)} &= C_1 E_{1,2}^{n} && n > 0 \\ x_n^{(1)} &= C_1[(E_{1,2}+1)/(2-x)]E_{1,2}^{n} && n \geqslant 0 \end{aligned} \tag{8}$$

where only the sequence that decreases appears. Of course the other constant is zero, otherwise $y_n^{(1)}$ and $x_n^{(1)}$ would increase without limit as $n \to \infty$.

Seeing that the impurity atom forms a symmetry centre, all solutions of this problem can be divided into symmetric and antisymmetric. It is easy to show that the localised antisymmetric modes have only trivial solution. In this case the impurity mass is at rest in the y_0 site. The previous result was pointed out by Mazur, Montroll and Potts

[17]. The constants C_1 and C_2 are connected by the continuity of the solution. This blends the two parts of the solution, equations (7) and (8), suitably in the imperfection region. Since the proposed solution must be continuous at site y_0 then $C_1 = C_2$. As C_1 is arbitrary, any constant multiple of y_n and x_n is also a solution of the boundary value problem. This arbitrary constant can be used for normalisation.

Finally, to ensure that the solution on the two sides of the impurity atom, equations (7) and (8), join properly across the impurity, they must satisfy a suitable matching condition, which is provided by the equation of motion of the maximum displacement for the impurity atom m' at site y_0. Only symmetric modes exist and non-trivial solutions are sought for this problem. So, if $m' = \varepsilon_m m$ and equations (7) and (8) are substituted in the equation of motion of the maximum displacement of the impurity atom, it is easily seen that

$$E_{1,2} = (\varepsilon_m/2)x^2 - (\varepsilon_m + 1)x + 1. \tag{9}$$

Writing the roots of the auxiliary equation of equation (3) in terms of x and ε and substituting into (9) gives the eigenvalue equation for the frequencies of the localised vibrations. Solving for these frequencies one obtains

$$\varepsilon_m(\varepsilon_m - 2\varepsilon)x^2 + 2[\varepsilon_m(2\varepsilon - \varepsilon_m) + 2\varepsilon]x - 4(\varepsilon + 1) = 0. \tag{10}$$

The reader is to be warned that in (10) appear roots that do not satisfy the localisation condition. A root must always be checked for appropriateness. Negative roots have no physical meaning and must be discarded. Once the frequency of the localised mode is known, the value of $E_{1,2}$ in equation (9) can be obtained and the eigenvector (maximum atomic displacements) can be calculated through equations (7) and (8).

Now a brief analysis of equation (10) is presented. For the special case $m = M$ ($\varepsilon = 1$), the monatomic lattice, the results obtained from (10) are in accordance with previous calculations. These are well known and a full analysis of this case has been made by many authors using several different mathematical techniques. The reader is referred to the original papers for details in the monatomic case [39]; see also [2, 4, 6, 8, 9, 13, 22]. When the difference equations method explained previously is applied to this monatomic model the same results are obtained [31].

For the case $m' = M$, $\varepsilon = \varepsilon_m$, the perfect lattice, the eigenvalue equation (10) gives the frequencies of the edges of the branches. Equation (10) shows that as $m' \to M$ the frequencies of the localised modes return to the edges of the branches. Observe that the solution for the edges of the branches (equation (4*b*)) with $C_2 = 0$ is included in the solution (equation (4*a*)). This special solution does not satisfy the localisation condition, but it is mentioned in order to complete the picture. Using (9) and the values of the roots from (10) the corresponding eigenvectors of each mode are obtained from (7) and (8). The eigenvectors have a simple harmonic spatial dependence. At the top of the acoustic branch only the heavy atoms move, whereas at the bottom of the optical branch only the light atoms move. Finally, in the highest-frequency mode of the perfect lattice, the particle pairs move against each other. These results are in accordance with those obtained analytically by Brillouin [33] and numerically by Barker and Sievers [22] for the edge modes of the perfect lattice.

An illustration of the dependence of the localised mode frequencies on the mass m' and the relative magnitudes of the masses M and m for the perfect lattice obtained

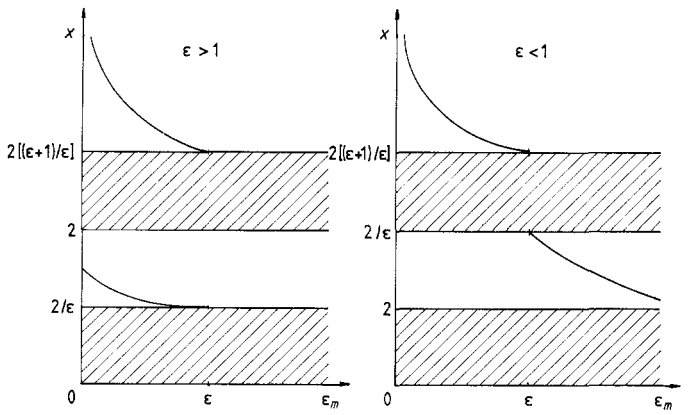

Figure 2. The dependence on the impurity mass of the frequencies of the localised modes when a heavy mass and a light mass are replaced by an isotopic impurity, respectively.

from equation (10) is shown schematically in figure 2. This dependence is also well known and may be summarised with the following statements: When substitution is made on the heavy mass ($\varepsilon > 1$), a lighter impurity mass ($\varepsilon_m < \varepsilon$) produces two localised modes, a local mode with frequency situated above the optical branch and a gap mode with frequency situated above the acoustic branch but below the optical branch of the host lattice. When substituting with a heavier impurity for the heavier atom ($\varepsilon_m > \varepsilon$), no new localised modes, local or gap, are generated. On the other hand, when substituting for the lighter mass ($\varepsilon < 1$) by a lighter impurity mass ($\varepsilon_m < \varepsilon$), one gets a localised mode, which is a local mode, with a frequency that lies above the top of the optical branch. And a heavier mass ($\varepsilon_m > \varepsilon$) causes a localised gap mode with frequency between optical and acoustic branches of the host lattice. In the case of replacing the heavy atom by a lighter mass, when two new localised modes appear simultaneously, for $m' \to 0$, the local mode raises its frequency to a very high value. However, the frequency of gap mode shows a different behaviour. It approaches the centre of the forbidden gap. For both local and gap localised modes a reduction of the mass impurity m' increases their frequencies, and vice versa if the mass impurity m' is larger, the two values of the frequencies of these local and gap modes diminish and their frequencies return to the edges of the branches for $m' = M$. The behaviour of the local mode frequency in the case of replacing the light host atom by a lighter one is like that described above for replacing the heavy atom by a lighter isotope. For $m' \to 0$ the frequency increases to a very high value and it the mass m' is larger this frequency decreases and returns to the top of the optical branch for $m' = M$. The other gap mode has its frequency in the gap between the optical and acoustic branches of the perfect lattice. This gap mode frequency diminishes its value from the top of the forbidden gap when the mass of the atomic impurity m' is equal to M, to the bottom of the gap as the substituted mass becomes very large $m' \to \infty$. These are exactly the results which have been obtained analytically by Mazur and co-workers [17], Maradudin and co-workers [2], Sen and co-workers [35] and Andrade and co-workers [36–38], and numerically by Barker and Sievers [22], but all these calculations contain unnecessary complications in their mathematical techniques.

The detailed nature of the localised mode will be examined by means of the investigation of the spatial form of this vibration. Figure 3 shows the eigenvector for the local mode in the case of replacing the heavy atom ($\varepsilon > 1$). All atoms move against each other in the local mode. The form of the vibrational mode does not change

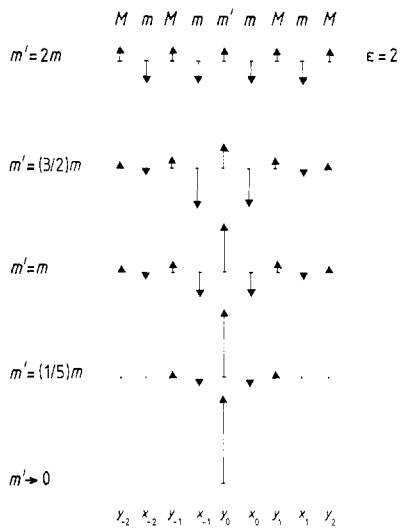

Figure 3. Local mode displacement pattern as a function of impurity mass when a heavy atom is replaced.

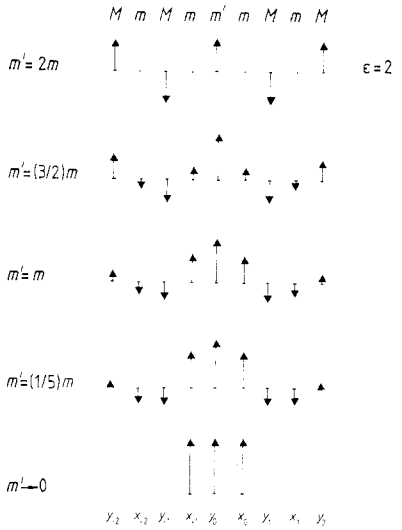

Figure 4. Gap mode displacement pattern as a function of impurity mass when a heavy atom is replaced.

abruptly with the variation of the mass m'. Rather, it evolves from the case when the particle pairs move in opposite directions to each other, the mode of maximum frequency of the perfect lattice and the mass impurity $m' = M$, to the case where there is practically no amplitude of displacement of any atom and the mass impurity tends to zero. In this case the atomic impurity is moving with large amplitude, whereas the impurity neighbours are moving with considerably less amplitude. For the gap mode, when substitution is made on the heavy mass ($\varepsilon > 1$), the eigenvector also has sequence spatial dependence. However, the displacement pattern is quite different in detail. All atoms move in pairs against each other and always maintain this displacement pattern. From $m' = M$, to $m' = 0$, this gap mode changes its form from the shape of the mode of maximum frequency of the perfect lattice where only heavy atoms are in motion to the so-called 'surface mode' coined by Mazur, Montroll and Potts [17]. In this gap mode the atom on each side of the atomic impurity only follows the mass impurity and the maximum atomic displacement of this mass impurity has a large amplitude. The impurity neighbours have considerably less amplitude. Note from figure 4 that for the gap mode the mass impurity plays almost no role in this type of motion. The maximum amplitude of displacement of the atoms in this mode falls to zero far from the impurity site. Mazur *et al* [17] interpreted the case $m' = 0$ in this mode in the following way: 'the lattice is reduced to two chains each with one end fixed and the other free'. Observe from figure 4 that when $m' = 0$ there is no relative displacement between the atomic impurity and its nearest neighbours, so that there is no force between them. The behaviour of the masses in this case confirms the interpretation of Mazur *et al*, for this like 'surface mode'. But, this procedure of interpretation is, however, physically unjustifiable because in this case, though the mass impurity is zero, there remains a force constant between the two chains. Indeed from a physical point of view the lattice cannot be broken into two independent portions and for this reason there is no surface. An analysis of the true surface mode has been given in detail by many workers [40].

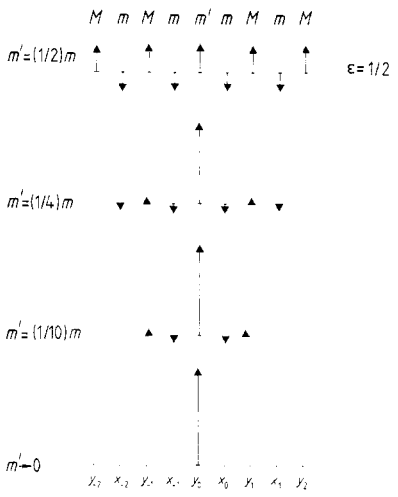

Figure 5. Local mode displacement pattern as a function of impurity mass when a light atom is replaced.

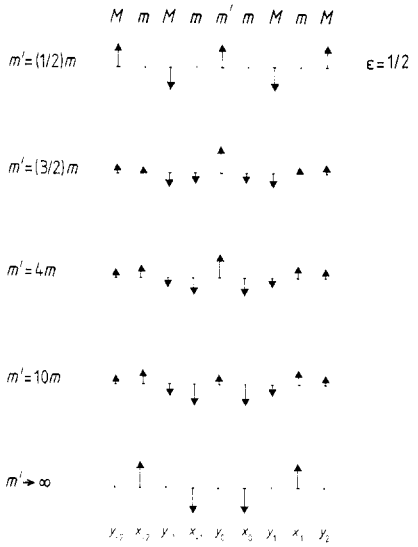

Figure 6. Gap mode displacement pattern as a function of impurity mass when a light atom is replaced.

It was mentioned previously that if the light host atom is replaced by a lighter one, then a local mode forms with a frequency above the maximum frequency of the perfect lattice. This mode is also localised spatially. Its eigenvector exhibits the impurity atom moving with large amplitude as compared with the amplitude of its neighbours. All atoms move against each other. It has a sequence eigenvector, with maximum amplitude which falls towards zero far from m'. One of the mode's properties is that it becomes more localised as $m' \to 0$. Figure 5 presents the local mode displacement pattern as a function of m'. It also develops its form from the mode of maximum frequency of the perfect lattice when $m' = M$, to the very localised mode where only the atomic impurity m' has displacement different from zero, when $m' \to 0$. Lastly, for the gap mode eigenvector, figure 6 shows the development of it from the minimum frequency mode of the optical branch where only the light atoms are moving when $m' = M$, to the case of the maximum frequency mode of the acoustic branch of the perfect lattice where only the heavy atoms move when $m' \to \infty$. In this limit the mass impurity does not have motion. This gap mode has a displacement pattern which is different from the displacement pattern of the gap mode discussed above. The results for eigenvectors complement those obtained numerically by Barker and Sievers [22] in a linear chain of 48 atoms using cyclic boundary conditions.

3.2. Force-constant defect

The next model of local imperfection consists of a single anomalous force constant. As before, it will also be proposed to obtain the solution of this problem as a characteristic value problem. There are many calculations [6, 16, 20, 22, 25] where force-constant changes were made in a linear diatomic lattice. Apparently, no work has been done on the calculation of frequencies and forms of the localised modes in a diatomic linear chain with a single anomalous force constant as a local imperfection.

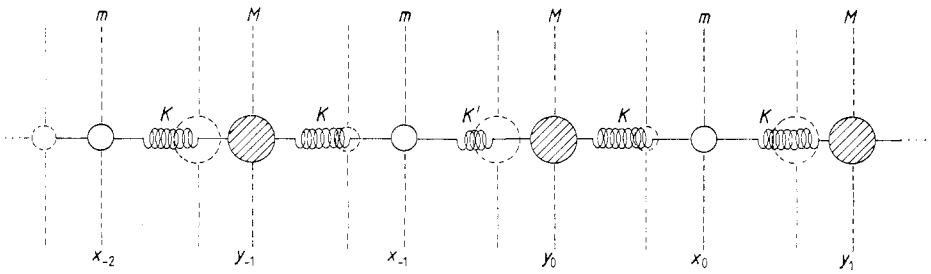

Figure 7. Diatomic linear lattice with a single force-constant defect.

The solution of this problem is not only of interest in itself but also serves as an additional model to the range of specific imperfection models in the systems cited above [1–25].

Now the local imperfection will be considered by replacing one constant K of the perfect lattice by K' between the atoms of mass m at site x_{-1} and mass M at site y_0. The imperfection model is shown in figure 7. The interval in which the pair of equations (1) and (2) are valid will be split into two parts: $-\infty \leqslant n \leqslant -1$ and $0 \leqslant n \leqslant \infty$. In order to solve the problem, use again one of the general solutions of the diatomic lattice (4*a*). Clearly, then, equation (7) is the solution for the interval $-\infty \leqslant n \leqslant -1$ and equation (8) is the solution for the interval $0 \leqslant n \leqslant \infty$.

The two constants C_1 and C_2 are determined by the two matching conditions, which blend the two parts of the solution adequately in the imperfection region. The matching conditions are provided by the two equations of motion of the maximum displacement of the atoms m at site x_{-1} and M at site y_0. If $K = K'\rho$ and by substituting (7) and (8) in the two equations of motion yields a set of two simultaneous linear equations in the two unknowns C_1 and C_2. A non-trivial solution is required, so that it is necessary and sufficient for the determinant formed by the two unknowns to be zero. Expanding this gives for $E_{1,2}$

$$\rho(\rho-1)E_{1,2}^2+[\rho b+a(\rho-1)+(x-2)]E_{1,2}+ab+x-2=0 \tag{11}$$

where $a = (\varepsilon\rho x - \rho - 1)(2 - x) + \rho$ and $b = \rho x - \rho - 1$.

Inserting the roots of the auxiliary algebraic equation of equation (3) in terms of x and ε and by substitution of it in (11), after some algebraic manipulation the equation for the frequencies of the localised modes is

$$\varepsilon^2\rho(\rho-2)x^3+2\varepsilon(\varepsilon+1)(-\rho^2+2\rho+1)x^2 + [(\varepsilon^2+1)(\rho^2-2\rho-3)+2\varepsilon(\rho^2-2\rho-5]x+8(\varepsilon+1)=0. \tag{12}$$

In (12) roots can appear that do not satisfy the localisation condition, so that the roots must be checked for appropriateness. The negative root when its exists has no physical meaning. The value of $E_{1,2}$ (equation (11)) can be calculated. First notice that the equation of motion of the atoms m at x_{-1} and M at y_0 determines one of the constants in terms of the other. Since one of the constants is arbitrary, any constant multiple of y_n and x_n is also a solution of this boundary value problem. The arbitrary constant can be used for normalisation.

For the special case $m = M(\varepsilon = 1)$ of the monatomic lattice, equation (12) supplies results that are in accordance with a previous calculation given by Montroll and Potts

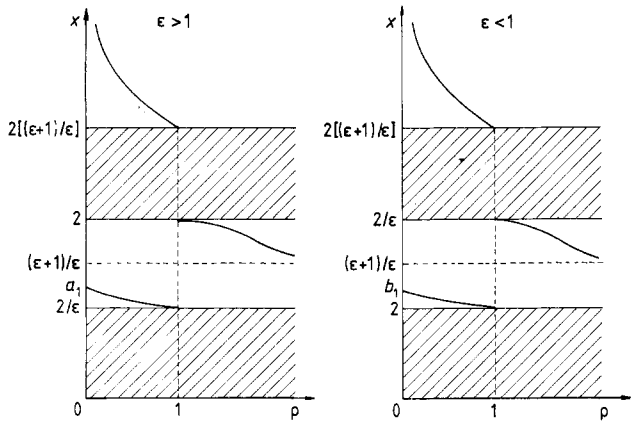

Figure 8. The dependence of the frequencies of the localised modes on the impurity force constant.

[2], using the Green function method, and by Andrade and Borau [41], using a heuristic solution. If the difference equations method is applied to this monatomic model the same results are obtained [32].

It is interesting to note that the frequencies of the localised modes given by (12) return to the edges of the branches as $\rho \to 1$. In the case $\rho = 1$ the eigenfrequency equation has three roots which correspond with the frequencies of the edges of the branches of the perfect lattice.

The results of solving the eigenfrequency equation of the localised modes (equation (12)) are shown in figure 8. The localised mode frequency depends on the force-constant impurity K' and the relative magnitudes of the masses M and m of the perfect lattice. This dependence is summarised for both cases when the mass M is larger than the mass m ($\varepsilon > 1$) or vice versa ($\varepsilon < 1$): If the force-constant impurity K' is larger than the force constant K of the perfect lattice ($\rho < 1$), two modes, a local mode with frequency situated above the optical branch and a gap mode with frequency situated in the forbidden gap of the modes of the perfect lattice, are generated. When the force-constant impurity K' is less than the force constant K of the perfect lattice ($\rho > 1$), there is a gap mode. The frequency of the local mode is larger if the impurity constant K' increases, and for $K' \to \infty$ it rises to a very high value; for $K' \to K$, this frequency tends to the maximum frequency of the optical branch of the normal modes in the perfect lattice. The gap mode frequency ranges from the value

$$a_1 = (3\varepsilon^2 + 10\varepsilon + 3) + [(3\varepsilon^2 + 10\varepsilon + 3)^2 - 64\varepsilon(\varepsilon+1)^2]^{1/2}/4\varepsilon(\varepsilon+1)$$

when $\varepsilon > 1$ or the value

$$b_1 = (3\varepsilon^2 + 10\varepsilon + 3) - [(3\varepsilon^2 + 10\varepsilon + 3)^2 - 64\varepsilon(\varepsilon+1)^2]^{1/2}/4\varepsilon(\varepsilon+1)$$

when $\varepsilon < 1$, for $K' \to \infty$, to the maximum frequency of the acoustic mode of the perfect lattice for $K' = K$; and from the minimum frequency of the optical branch for the normal modes of the perfect lattice when $K' = K$ to the frequency of the surface mode when $K' = 0$.

In figures 9, 10 and 11 the maximum atomic displacements of the localised modes are plotted as a function of the positions of the atoms. The eigenvectors show simple spatial dependence. It is clear from figure 9 that in the case of the local mode all atoms move against each other. For $K' \to K$ the form of the mode is exactly like the mode

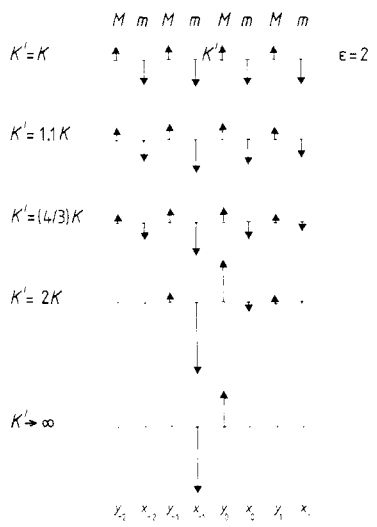

Figure 9. Eigenvectors for local modes when the impurity force constant is larger than the force constant of the perfect lattice.

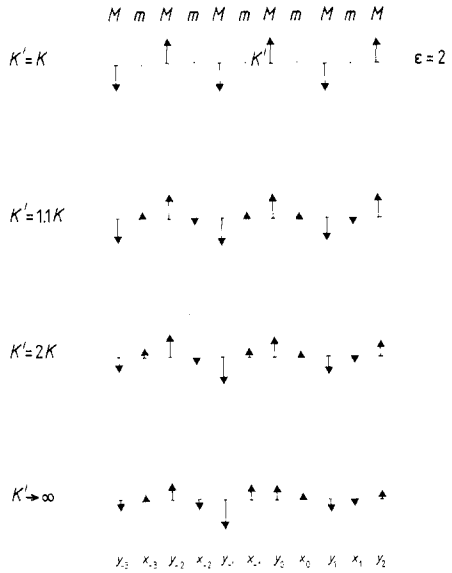

Figure 10. Eigenvectors for gap modes when the impurity force constant is larger than the force constant of the perfect lattice.

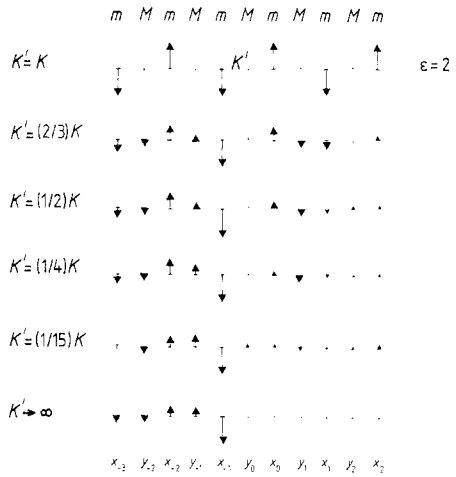

Figure 11. Eigenvectors for gap modes when the impurity force constant is reduced.

of maximum frequency of the optical branch in a perfect lattice, and for $K' \to \infty$, this vibration type shows two atoms with amplitude different from zero. They are the atoms linked by the force-constant impurity. In this case the atom with larger mass has an amplitude of displacement shorter than the atom of lighter mass. For the gap mode in the case $\rho < 1$ notice from figure 10 that this mode evolves in shape in a simple form, from the form of the mode of maximum frequency in the acoustic branch of the perfect lattice for $\rho = 1$, when only the atoms with larger mass have an amplitude different from zero, to the mode that is similar in shape when $K' \to \infty$ to the Wallis type 'surface mode' [40]. Though the maximum atomic displacements have the same direction as in the 'surface mode' only the two atoms linked by the constant impurity

K' have the same amplitude. The nearest neighbour to the impurity K' with the larger mass has the maximum atomic displacement in the lattice. Thus, in this mode the maximum atomic displacements of the two atoms of masses m and M of the lattice are different. Finally, the shape of the gap mode when $\rho > 1$, figure 11, evolves from the form of the mode of minimum frequency in the optical branch of the perfect lattice when $\rho = 1$ to the surface mode when $\rho \to \infty$. In the latter case when $K' = 0$ there are two semi-infinite lattices with surfaces so that in the semi-infinite lattice with lighter mass at the surface there appears a surface mode, and in another semi-infinite lattice with heavier mass at the surface there are no localised modes. For this limit all the atoms in the latter semi-infinite lattice have zero amplitude.

The behaviour of the present model of imperfection in the diatomic lattice is not markedly different from the behaviour of other models previously studied. One local mode was found when the value of force-constant defect is larger than the value of the force constant of the perfect lattice. One gap mode was obtained for any value of the force-constant defect. The shape of the eigenvector for this gap mode shows that indeed there are two different gap modes, one when $\rho > 1$ and another when $\rho < 1$. It should be pointed out that a single force-constant defect in a diatomic linear chain with nearest-neighbour interactions can never produce an in-branch localised mode.

4. Conclusions

The lattice dynamics for a diatomic lattice with nearest-neighbour interactions was examined by using the difference equations method. Specific results were obtained in the case of one dimension. Therefore it was demonstrated that a closed analysis for the lattice structure is possible through finite difference techniques. The application of the approach is quite systematic and the procedure is essentially the same as for ordinary differential equations.

As a first result, the general solution of the diatomic lattice was obtained analytically via the operator method. This contains three different sets of solution types, i.e. sequence (4*a*), edge branch (4*b*) and periodic (4*c*). An analysis of the range of frequencies in which the different sets of the general solution appear has been given. Although the distinct sets of the general solution had already been employed before by a large number of workers [1–25], apparently it is the first time that this general solution has been obtained without heuristic propositions and in a systematic way. It is of great importance, because in the eigenfunction method, this general solution supplies the mathematical basis which must be used in characteristic value problems. Also, the density of modes or frequency distribution function for the perfect lattice was calculated.

Furthermore, a theory to study localised vibrational modes in a lattice with local imperfection was presented.

In order to apply the technique, the frequencies and the forms of the localised vibrational modes when the local imperfection model is a single isotopic mass or an anomalous force constant were calculated. All results in these cases were obtained firstly in a simpler and more straightforward manner than those calculated with all other mathematical methods mentioned above [2, 6, 17, 22, 35–38] and secondly for the first time. Notice that in this work the author tried mostly to underline the methodological side of the problem, and naturally could not dwell upon the major

part of the cases with other different imperfection models. These are necessary, since they can be applied to understand optical effects in real crystals [27, 28].

Finally, in spite of the fact that only one class of diatomic linear lattice was examined here, it seems that this method could have advantages and profound significance in the analysis of the lattice dynamics for other more complex lattices.

References

[1] Lifshitz I M 1943 *J. Phys. USSR* **7** 211, 249; 1944 *J. Phys. USSR* **8** 89; 1956 *Nuovo. Cimento Suppl.* **3** 716

[2] Montroll E W and Potts R B 1955 *Phys. Rev.* **100** 525
Maradudin A A, Mazur P, Montroll E W and Weiss G H 1958 *Rev. Mod. Phys.* **30** 175
Maradudin A A, Montroll E W and Weiss G H 1963 *Solid State Phys. Suppl.* (New York: Academic) ch 5

[3] Lax M 1954 *Phys. Rev.* **94** 1391

[4] Dean P 1967 *J. Inst. Math. Applic.* **3** 98

[5] Litzman O 1957 *Czech. J. Phys.* **7** 410, 690; 1959 *Czech. J. Phys.* **9** 692

[6] Hori J and Asahi T 1957 *Prog. Theor. Phys.* **17** 523

[7] Hori J 1961 *J. Phys. Soc. Japan* **16** 23

[8] Fukuda Y 1962 *J. Phys. Soc. Japan* **17** 766

[9] Andrade L and Saavedra H C 1977 *Bull. Phys. Mex. Soc.* **4** 182

[10] Teramoto E 1960 *Prog. Theor. Phys.* **24** 1296

[11] Toda M, Kotera T and Kogure K 1962 *J. Phys. Soc. Japan* **17** 426

[12] Teramoto E and Takeno S 1960 *Prog. Theor. Phys.* **24** 1349

[13] Krumhansl J A 1962 *J. Appl. Phys. Suppl.* **33** 307

[14] Dean P 1968 *J. Phys. C: Solid State Phys.* **1** 22

[15] De Loach B C Jr and Shaffer W H 1961 *J. Mol. Spectrosc.* **6** 229
Lee S M and Shaffer W H 1966 *J. Mol. Spectrosc.* **21** 183

[16] Bala S, Malik D P S and Ghatak A K 1972 *J. Phys. Chem. Solids* **33** 1885
Bala S, Ghatak A K and Malik D P S 1972 *Phys. Lett.* **39A** 179

[17] Mazur P, Montroll E W and Potts R B 1956 *J. Washington Acad. Sci.* **46** 2

[18] Genzel L, Renk K F and Weber R 1965 *Phys. Status Solidi* **12** 639

[19] Andrade L 1977 *Proc. 5th Latinamerican Symp. Solid State Physics* (*Lima: UNI*) p 7

[20] Bjork R L 1957 *Phys. Rev.* **105** 456

[21] Bacon M D, Dean P and Martin J L 1962 *Proc. Phys. Soc.* **80** 174

[22] Barker A S Jr and Sievers A J 1975 *Rev. Mod. Phys. Suppl.* **47** 56

[23] Barker A S Jr, Berman R and Verleur H W 1973 *J. Phys. Chem. Solids* **34** 123

[24] Wagner M 1963 *Phys. Rev.* **131** 2520
Munn R W 1970 *J. Chem. Phys.* **52** 64

[25] Behera S N and Patnaik K 1975 *Phys. Rev.* B **12** 4547; 1976 *Phys. Rev.* B **13** 2705; 1976 *Pramāna* **7** 102

[26] Andrade L 1981 *Recent Progress in Many-Body Theories* vol 142 (Berlin: Springer) p 309

[27] Lucovsky G, Brodsky M H and Burstein E 1970 *Phys. Rev.* B **2** 3295
Hattori T, Ehara K, Mitsuishi A, Sakuragi S and Kanzaki H 1973 *Solid State Commun.* **12** 545

[28] Rosenstock H B and Klick C C 1960 *Phys. Rev.* **119** 1198
Mitsuishi A and Yoshinaga H 1962 *Prog. Theor. Phys.* (*Kyoto*) *Suppl.* **23** 241
Angress J F, Goodwin A R and Smith S D 1965 *Proc. R. Soc.* A **287** 64

[29] Vandevyver M, Talwar D N, Plumelle P, Kunc K and Zigone M 1980 *Phys. Status Solidi* b **99** 727
Vandevyver M and Plumelle P 1977 *J. Phys. Chem. Solids* **38** 765; 1978 *Phys. Rev.* B **17** 675
Talwar D N and Agrawal B K 1974 *Phys. Rev.* B **9** 2539; 1975 *Phys. Rev.* B **12** 1432
Grimm A, Maradudin A A, Ipatova I P and Subashiev A V 1972 *J. Phys. Chem. Solids* **33** 775
Gaur S P, Vetelino J F and Mitra S S 1971 *J. Phys. Chem. Solids* **32** 2737
Govindarajan J and Haridasan T M 1969 *Phys. Lett.* **29A** 387
Sievers A J, Maradudin A A and Jaswal S S 1965 *Phys. Rev.* **138** A272
Nolt I G, Westwig R A, Alexander R W Jr and Sievers A J 1967 *Phys. Rev.* **137** 730
Benedek G and Maradudin A A 1968 *J. Phys. Chem. Solids* **29** 423

Bäuerle D and Hübner R 1970 *Phys. Rev.* B **2** 4252
Benedek G 1970 *Phys. Status Solidi* **42** 389
de Jong C 1971 *Solid State Commun.* **9** 527
Zavt G S 1963 *Fiz. Tverd. Tela* **5** 1086 (Engl. Transl. 1963 *Sov. Phys.–Solid State* **5** 792)
Takeno S, Kashiwamura S and Teramoto E 1962 *Prog. Theor. Phys. Suppl.* **22** 124
Jaswal S S and Montgomery D D 1964 *Phys. Rev.* **135** A1257
Jaswal S S 1965 *Phys. Rev.* **140** A687
Sebastian R, Savari Raj G A and Haridasan T M 1979 *Phys. Status Solidi* b **91** 371
Page J B and Strauch D 1967 *Phys. Status Solidi* **24** 469
Benedek G and Nardelli G F 1967 *Phys. Rev.* **155** 1004
Fieschi R, Nardelli G F and Terzi N 1965 *Phys. Rev.* **138** A203
[30] Hildebrand F B 1968 *Finite-Difference Equations and Simulations* (Englewood Cliffs, NJ: Prentice-Hall)
Boole G 1970 *Calculus of Finite Differences* (New York: Chelsea)
[31] Andrade L 1983 *Internal Report* Facultad de Ciencias, UNAM, Winter, Dec. 1982–Mar. 1983
[32] Andrade L 1980 *Internal Report* Facultad de Ciencias, UNAM, Fall
[33] Brillouin L 1953 *Wave Propagation in Periodic Structures* (New York: Dover)
[34] Blackman M 1935 *Proc. R. Soc.* A **148** 365
[35] Sen P N and Hartmann W M 1974 *Phys. Rev.* B **9** 367
[36] Andrade L 1988 *Phys. Mex. Rev.* submitted
[37] Andrade L, Miranda A and Saavedra C H 1977 *Bull. Phys. Mex. Soc.* **4** 183
[38] Miranda G A 1981 *Thesis* Facultad de Ciencias, UNAM
[39] Andrade L and Saavedra H C 1976 *Bull. Phys. Mex. Soc.* **4** 192
[40] Wallis R F 1957 *Phys. Rev.* **105** 540; 1964 *Surf. Sci.* **2** 146
Asahi T and Hori J 1963 *Proc. Int. Conf. Lattice Dynamics* (*Copenhagen*) (Oxford: Pergamon) p 571
Asahi T 1963 *J. Res. Inst. Catal. Hokkaido Univ.* **11** 133
Hori J and Asahi T 1964 *Prog. Theor. Phys.* **31** 49
Cheng Y 1969 *Physica* **42** 124
Collett T J, Bishop F M and Trullinger E S 1978 *Phys. Rev.* B **18** 2464
Andrade L and Alvarez M 1976 *CAP–APS–SMF Joint Congr.* (*Quebec*); 1976 *Bull. Am. Phys. Soc.* **21** 794
[41] Andrade L and Borau J 1979 *Bull. Mex. Phys. Soc.* **4** 190