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The Hermiticity of the dynamical matrix. By C. SCHERINGER, *Fritz-Haber-Institut der Max-Planck-Gesellschaft*, 1 *Berlin* 33, *Faradayweg* 4–6, *Germany*

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In the usual calculation of the dynamical matrices of a crystal the self terms can give rise to non-Hermitian dynamical matrices, particularly when the crystal symmetry is low. It has been suggested that Hermiticity be imposed upon the interatomic force constants as an extra condition. It is shown that this is not necessary and that, with a correct treatment of the basic equations of lattice dynamics, the dynamical matrices will automatically be Hermitian.

Recently, Powell (1970) assumed that in the common formulation of the Born–von Karman theory, *cf.*, for example, Born & Huang (1954), the dynamical matrices of the crystal, for low-symmetry crystals, may not be Hermitian. The origin of this non-Hermiticity was found, by Powell, to be in the calculation of the so-called self terms of the dynamical matrices. In the common formulation of the lattice-dynamical theory the (non-mass-normalized) elements of the dynamical matrices of wave vector $\mathbf{q}$ have the form

$$L_{\alpha\beta}(\mathbf{q},kk')=\sum_{l'}\phi_{\alpha\beta}(lk,l'k')\exp\{i\mathbf{q}\,.\,[\mathbf{X}(l'k')-\mathbf{X}(lk)]\}\,, \quad (1)$$

cf., for example, Born & Huang [1954, equation (24·7)], where $\alpha,\beta=1,2,3$ denote the directions of space; l,l' the cells in the crystal; and k,k' the atoms in the cell. $\mathbf{X}(lk)$ is the position vector of the atom lk in the crystal, and $\phi_{\alpha\beta}(lk,l'k')$ are the interatomic force constants. For these constants the symmetry conditions

$$\phi_{\alpha\beta}(lk,l'k')=\phi_{\beta\alpha}(l'k',lk) \quad (2)$$

hold, by virtue of their definition as second derivatives of the potential energy of the crystal, *cf.* Born & Huang [1954, equations (23·3) and (24·11)]. Equation (2) ensures that the dynamical matrices, as calculated from (1), will be Hermitian. The self terms $l'k'=lk$ of the interatomic force constants, however, are not the second derivatives of the crystal potential but are calculated from the condition of translation invariance. Powell (1970) applied this generally accepted equation

$$\phi_{\alpha\beta}(lk,lk)=-\sum_{l'k'}{}'\phi_{\alpha\beta}(lk,l'k')\,, \quad (3)$$

cf. Born & Huang [1954, equation (23·16)]; Maradudin, Montroll & Weiss [1963, equation (2.1.12*b*)]. In $\sum'$ the term $l'k'=lk$ is excluded. With the example of the low-symmetry Te crystal, Powell (1970) found that the symmetry condition (2) was not fulfilled for the self terms $\phi_{\alpha\beta}(lk,lk)$. Hence, Powell concluded that, in general, the dynamical matrices may not be Hermitian.

Martin (1971) corrected Powell (1970) by showing that Powell had not used the correct formulation for the condition of translation invariance. According to Martin [1971, equation (8)] the condition of translation invariance properly reads

$$\tfrac{1}{2}[\phi_{\alpha\beta}(lk,lk)+\phi_{\beta\alpha}(lk,lk)]=-\sum_{l'k'}{}'\phi_{\alpha\beta}(lk,l'k')\,, \quad (4)$$

instead of (3). Since the left-hand side of (4) is certainly symmetric in α,β for any matrix $\phi_{\alpha\beta}(lk,lk)$, (4) imposes a symmetry condition on the interatomic force constants used on the right-hand side of (4) so that the total sum must be symmetric in α and β. Force constants which do not satisfy this condition are physically not acceptable. Since (4) does not imply that the self-term matrix $\phi_{\alpha\beta}(lk,lk)$, as it occurs in the dynamical matrix (1), must also be symmetric in α and β but may have any form, Martin (1971) concluded that in the usual lattice-dynamical theory '... the Hermitian character of the dynamical matrix can therefore be imposed as a condition upon any general force constant model'. Martin (1971) further emphasized that '... Hermiticity can be added to rotation and translation invariance as fundamental requirements for acceptability of any force constant model'.

It is the purpose of this note to show that there is no need to impose Hermiticity as a condition on the interatomic force constants, but rather that, in a correct treatment of the basic equations of lattice dynamics, Hermiticity is automatically obtained, provided the correct form of the condition (4) of translation invariance is used. If one uses the Hamiltonian of the crystal in order to set up the equations of motion and *does not assume* the symmetry condition (2), the result is

$$\frac{\partial\phi}{\partial u_\alpha(lk)}=\tfrac{1}{2}\sum_{l'k'\beta}[\phi_{\alpha\beta}(lk,l'k')+\phi_{\beta\alpha}(l'k',lk)]u_\beta(l'k')\,. \quad (5)$$

Thus, in the equations of motion we have half the sum of the two terms $\phi_{\alpha\beta}(lk,l'k')$ and $\phi_{\beta\alpha}(l'k',lk)$, and not the single term $\phi_{\alpha\beta}(lk,l'k')$, as given, for example, by Born & Huang [1954, equation (24·2)]. If one proceeds with the calculation in the normal manner, one obtains for the elements of the dynamical matrices

$$L_{\alpha\beta}(\mathbf{q},kk')=\tfrac{1}{2}\sum_{l'}[\phi_{\alpha\beta}(lk,l'k')+\phi_{\beta\alpha}(l'k',lk)]\times\exp\{i\mathbf{q}\,.\,[\mathbf{X}(l'k')-\mathbf{X}(lk)]\}\,, \quad (6)$$

instead of (1). For the cases $l'k'\neq lk$ the symmetry condition (2) holds, and (5) and (6) reduce to the expressions which have been used in the past. For the self terms $l'k'=lk$, however, the formulation with (5) and (6) is necessary, since the single terms $\phi_{\alpha\beta}(lk,lk)$ are neither defined in the Hamiltonian of the crystal, nor by the condition of translation invariance. Hence, any attempt to calculate the single terms $\phi_{\alpha\beta}(lk,lk)$, or to make statements about the symmetry of the single terms, is rendered meaningless. Thus, if one calculates the sum of the two terms from the condition (4) of translation invariance, observing that the sum on the right-hand side of (4) must be symmetric in α and β, then with the use of (6) one always obtains dynamical matrices which are Hermitian.

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