

Microscopic Theory of Dielectric Screening and Lattice Dynamics in the Wannier Representation. I. Theory*

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The microscopic quantum-mechanical expressions for the dielectric screening matrix and for the electronic contribution to lattice dynamics are derived in terms of generalized Wannier functions. The Wannier representation makes practical an inversion of the dielectric matrix within the random-phase approximation and thus allows for an explicit calculation of local field effects in the dielectric response. This result leads, with the help of the dynamical matrix of an electron-nucleus system, to a multipole model of lattice dynamics. The formalism can be applied to conducting as well as to nonconducting crystals, and in this way provides a possibility to examine the relationship between the various methods and models used to describe lattice vibrations in all these solids.

I. INTRODUCTION

The general lattice-dynamical formulations focus on the problem of expressing the phonon self-energy in terms of the response of the crystal electrons to an external probe. This so-called microscopic dielectric approach permits the complete solution of the lattice vibrational problem within the harmonic approximation once the charge of the nuclei and a properly defined inverse dielectric matrix for the electrons, $\epsilon^{-1}(\vec{q} + \vec{G}, \vec{q} + \vec{G}')$, are known.¹⁻³ From this it follows that the dielectric matrix and its inverse reveal the basic difference between the commonly used theory of lattice dynamics of metals and the corresponding theory of nonmetallic crystals.

A particularly noteworthy feature of the dielectric matrix of an insulating crystal is that the $\vec{G} \neq \vec{G}'$ elements of ϵ^{-1} have to be nonzero—indeed, these elements are necessary for the satisfaction of an exact sum rule which ensures that all acoustic-mode frequencies approach zero as the wave vector of the mode goes to zero.^{1,3} Since the nondiagonal elements of ϵ^{-1} are, as well, a manifestation of local-field effects, one might also expect the importance of $\vec{G} \neq \vec{G}'$ elements in insulators from the local character of the wave functions.

In a simple metal, on the other hand, the common approach to the lattice dynamics is afforded by means of the free-electron approximation for the conduction electrons, which renders the Hartree dielectric-screening function diagonal ($\vec{G} = \vec{G}'$). But in transition metals the requirement of $\vec{G} \neq \vec{G}'$ elements follows again from band-structure arguments: in these metals the conduction-band states are of free-electron (*s* and *p*) character as well as of local (*d*) character. Moreover the *d*-like electrons of the filled bands are not rigidly bound to the core and therefore must be included in the dynamical screening process.

It follows that, with the exception of simple

metals, a practical microscopic treatment of the lattice dynamics of solids is not possible unless the problem of inversion of the nondiagonal dielectric matrix has been solved.

In two previous papers^{4,5} a formalism was developed whereby an exact inversion of the random-phase-approximation (RPA) dielectric matrix could be achieved in terms of localized or Wannier functions. As a result, the dynamical matrix leads to a generalized multipole model of lattice dynamics. The formalism can be applied to normal and transition metals as well as to covalent and ionic crystals and thus provides a unified treatment of the theory of lattice vibrations in all these solids. An application of this unified approach was performed on the transition metal Ni in the paramagnetic phase.⁵ This application represents the first microscopic phonon calculation in a metal which takes into account the nondiagonality of the dielectric matrix, i. e., local-field effects in the dielectric response.

In the first paper (I) in this series we consider explicitly the theory of dielectric screening for a crystal the relevant energy bands of which overlap with other bands [in Ref. 5 the case of isolated, simple (noncomposite) bands was treated in detail, but for composite (hybridizing, crossing, touching) bands only the final results were stated]. This analysis is of importance because in many, if not most, calculations of the dielectric matrix ϵ the condition of isolated bands entering does not hold. For example in all of the transition metals the *s* and *d* bands are split by the *s-d* hybridization and overlap with other bands. The Wannier functions corresponding to these single *s* and *d* bands do not converge exponentially but only with $1/r^4$ and this poor convergence renders the inversion procedure of ϵ in practice rather unmanageable. The $1/r^4$ convergence can be improved to an exponential decay if one takes into account the whole “complex”

of bands which are connected to each other but are disconnected from the rest of the energy spectrum. The set of Wannier functions corresponding to this "band complex" is related to the original set by a unitary transformation, and we shall see that this generalized Wannier representation allows in a completely analogue manner for an exact inversion of the dielectric matrix as was outlined in Refs. 4 and 5. For practical purposes one may introduce instead of these pseudo-Wannier functions, linear-combination-of-atomic-orbitals (LCAO) functions based on the Slater-Koster interpolation scheme.⁶ Because from the group-theoretical point of view the arrangement of the atomic orbitals in the LCAO treatment is equivalent to that of the pseudo-Wannier-functions in the exact treatment, the general results stated in this paper are

$$\epsilon(\vec{q} + \vec{G}, \vec{q} + \vec{G}') = \delta_{\vec{G}, \vec{G}'} - v(\vec{q} + \vec{G}) \sum_{n, n', \vec{k}} \langle n, \vec{k} | e^{-i(\vec{q} + \vec{G}) \cdot \vec{r}} | n', \vec{k} + \vec{q} \rangle \frac{f(n, \vec{k}) - f(n', \vec{k} + \vec{q})}{E(n, \vec{k}) - E(n', \vec{k} + \vec{q})} \langle n', \vec{k} + \vec{q} | e^{i(\vec{q} + \vec{G}') \cdot \vec{r}} | n, \vec{k} \rangle, \quad (1)$$

where \vec{G} and \vec{G}' are reciprocal-lattice vectors, $f(n, \vec{k})$ is the occupation number of a Bloch electron with wave vector \vec{k} , band index n , and energy $E(n, \vec{k})$. The electron-electron interaction $v(\vec{q} + \vec{G})$ may include corrections for exchange and correlation in the Hubbard approximation.⁹ Extension of our inversion result to apply to the frequency-dependent RPA dielectric matrix requires just the introduction of the frequency ω in the energy denominator of Eq. (1).

We now choose for the Bloch functions $\psi_{n, \vec{k}}(\vec{r})$ a description in terms of Wannier functions of the "band complex." For this we consider the set of eigenfunctions $\psi_{n, \vec{k}}(\vec{r})$, satisfying the crystal Schrödinger equation

$$H\psi_{n, \vec{k}}(\vec{r}) = E(n, \vec{k})\psi_{n, \vec{k}}(\vec{r}), \quad n = 1, \dots, \tilde{n} \quad (2)$$

whose eigenvalues $E(n, \vec{k})$ are disconnected from all other sets of bands (band complexes). The $E(n, \vec{k})$ may all be connected to each other, or they may be partly or wholly disconnected from each other. Then it is possible to construct a set of Wannier functions (In order to keep the notation from becoming too cumbersome we assume only one atom per unit cell. The generalization to crystals with more than one atom per unit cell is straightforward.)

$$\varphi_{\nu}(\vec{r} - \vec{R}^I), \quad \nu = 1, \dots, \tilde{n} \quad (3)$$

which have the following properties¹⁰⁻¹²: They decay exponentially at infinity, are orthonormal,

$$\langle \varphi_{\nu}(\vec{r} - \vec{R}^I) | \varphi_{\nu'}(\vec{r} - \vec{R}^{I'}) \rangle = \delta_{\nu\nu'} \delta_{II'}, \quad (4)$$

also applicable in the tight-binding limit. A similar lattice-dynamics approach in terms of localized Wannier functions was independently derived by Sham,⁷ while for ideal insulators Pick has given a corresponding treatment.⁸

In the second paper (II) in this series we examine the problems raised by the application of our formalism to the transition metals Pd and Ni. It is shown how local-field corrections due to the d electrons, i. e., off-diagonal elements of the dielectric matrix, enter into an explicit calculation; it is also shown that they are important for a satisfactory explanation of the experimental phonon results.

II. ELECTRONIC SCREENING

The dielectric matrix in the RPA is given by

and the Bloch waves can be obtained from them by a unitary transformation:

$$\psi_{n, \vec{k}}(\vec{r}) = N^{-1/2} \sum_{\nu} e_{\nu}(n, \vec{k}) \sum_I e^{i\vec{k} \cdot \vec{R}^I} \varphi_{\nu}(\vec{r} - \vec{R}^I). \quad (5)$$

Here N is the number of lattice vectors \vec{R}^I , and $\vec{e}(n, \vec{k})$ are the so-called vectors of the band complex satisfying the following relations:

$$\begin{aligned} \sum_{\nu} e_{\nu}^*(n, \vec{k}) e_{\nu}(n', \vec{k}) &= \delta_{n, n'}, \\ \sum_{n=1}^{\tilde{n}} e_{\nu}^*(n, \vec{k}) e_{\nu'}(n, \vec{k}) &= \delta_{\nu\nu'}, \end{aligned} \quad (6)$$

i. e., $e_{\nu}(n, \vec{k})$ is a unitary matrix in the indices n and ν . The general construction of the Wannier functions $\varphi_{\nu}(\vec{r} - \vec{R}^I)$ for composite bands in terms of the Bloch waves has been described in detail by Des Cloizeaux¹⁰ and requires elaborate group-theoretic considerations. Here we need only the result in that these functions have exponential decay at infinity, that their number per unit cell is equal to the number of connected bands and that they are related to the Bloch functions by the unitary transformation of Eq. (5). Using this relation, the dielectric matrix can be written in the form

$$\begin{aligned} \epsilon(\vec{q} + \vec{G}, \vec{q} + \vec{G}') &= \delta_{\vec{G}, \vec{G}'} - v(\vec{q} + \vec{G}) \sum_{s, s'} A_s(\vec{q} + \vec{G}) \\ &\quad \times N_{ss'}(\vec{q}) A_{s'}^*(\vec{q} + \vec{G}'), \end{aligned} \quad (7)$$

where

$$A_s(\vec{q} + \vec{G}) = \int \varphi_{\nu}^*(\vec{r}) e^{-i(\vec{q} + \vec{G}) \cdot \vec{r}} \varphi_{\mu}(\vec{r} + \vec{R}^I) d^3r$$

and

$$N_{ss'}(\vec{q}) = \sum_{n, n', \vec{k}} \frac{f(n, \vec{k}) - f(n', \vec{k} + \vec{q})}{E(n, \vec{k}) - E(n', \vec{k} + \vec{q})} e^{-i(\vec{k} + \vec{q}) \cdot (\vec{R}^I - \vec{R}^{I'})} e_{\nu}^*(n, \vec{k}) e_{\mu}^*(n', \vec{k} + \vec{q}) e_{\mu}(n', \vec{k} + \vec{q}) e_{\nu}(n, \vec{k}).$$

The index s stands for the lattice vector index l and for the indices ν and μ of the band complex. The separable form of the susceptibility matrix ANA^\dagger in Eq. (7) enables us to calculate the inverse dielectric matrix

$$\epsilon^{-1}(\vec{q} + \vec{G}, \vec{q} + \vec{G}') = \delta_{\vec{q}, \vec{q}'} + v(\vec{q} + \vec{G}) \sum_{s, s'} A_s(\vec{q} + \vec{G}) \times S_{ss'}^{-1}(\vec{q}) A_{s'}^*(\vec{q} + \vec{G}'), \quad (8)$$

where

$$S_{ss'}(\vec{q}) = N_{ss'}^{-1}(\vec{q}) - \sum_{\vec{q}''} A_s^*(\vec{q} + \vec{G}'') \times A_{s'}(\vec{q} + \vec{G}'') v(\vec{q} + \vec{G}'').$$

This result can be proved immediately with the help of the following matrix expansion for

$$\epsilon^{-1}(\vec{q} + \vec{G}, \vec{q} + \vec{G}')$$

$$\begin{aligned} \epsilon^{-1} &= (1 - vANA^\dagger)^{-1} = 1 + vANA^\dagger + (vANA^\dagger)^2 + \dots \\ &= 1 + vAN\{1 + A^\dagger vAN + (A^\dagger vAN)^2 + \dots\} A^\dagger \\ &= 1 + vA\{N^{-1} - A^\dagger vA\}^{-1} A^\dagger. \end{aligned} \quad (9)$$

It follows that by changing to a localized description, the inversion problem of the infinite-dimensional matrix ϵ has been reduced to that of an inversion of the matrix S defined above. As may be seen from Eq. (7), the dimension of this matrix is greatly reduced, provided the functions $\varphi_\nu(\vec{r} - \vec{R}^l)$ are well localized. Because we presumed to use the best localized set of Wannier functions $\{\varphi_\nu\}$ of the band complex, we now have the important result that even in crystals with hybridized and crossing energy bands as transition metals an explicit solution of the inversion problem becomes possible.

In practice, difficulties arise from the fact that, while the existence of well-localized and orthogonal Wannier functions has often been involved in theoretical discussions, only first steps in the direction of a quantitative construction of these functions have been taken.^{12,13} However, we can suggest a certain simplification which makes the approach just considered practical. This is the LCAO approximation.^{6,14} In this method we take a finite set of atomic orbitals $\{\varphi_{nlm}\}$ on each of the atoms of the unit cell, considering only the atomic orbitals whose energy is somewhere near that of the energy bands in which we are interested. From each of these atomic orbitals, we construct a Bloch sum

$$\sum_l e^{i\vec{k}\cdot\vec{R}^l} \varphi_{nlm}(\vec{r} - \vec{R}^l) \quad (10)$$

and set up a wave function consisting of a linear combination of all these Bloch sums of the form given in Eq. (5). There will be matrix components of energy between all these Bloch sums, so that we have a secular problem whose order equals that

of the total number of atomic orbitals concerned. In practice, one replaces the matrix components by disposable constants which are chosen to fit accurate determination of energies at highly symmetric \vec{k} values and reduces the number of these constants by symmetry arguments. The resulting eigenvectors (eigenvectors of the band complex) $\vec{e}(n, \vec{k})$ of the secular equation form the wave functions of Eq. (5), while the eigenvalues give a useful approximation of the energy spectrum. In this way we get an interpolation scheme for the energies and wave functions throughout the whole Brillouin zone,⁶ which allows for a practical solution of our inversion problem of Eq. (8). To determine the electron-phonon matrix element within this tight-binding treatment the electron-ion potential is replaced by the self-consistent potential of the actual band-structure calculation.

In the case of a metal, specifically in the case of a noble or transition metal, it is necessary for practical purposes to combine the LCAO description applied to the localized d electronic states with the nonlocalized orthogonal-plane-wave (OPW) scheme for the s - and p -conduction-band states. This combined band-structure scheme^{15,16} is based on the fact that except for hybridization effects, the d bands closely resemble those obtained in the tight-binding approximation, whereas the conduction bands are similar to those resulting from nearly-free-electron calculations. Accordingly, the unhybridized d bands are represented in terms of linear combinations of the atomic d orbitals, and the conduction bands in terms of linear combinations of OPW's.¹⁷ Effects from the hybridization of the conduction and d basis functions are included in this approach in the eigenvector coefficients $\vec{e}(n, \vec{k})$ of the corresponding energy eigenvalue problem.

Using this combined LCAO-OPW description, the dielectric matrix Eq. (1) can be split into two parts^{4,5}:

$$\epsilon(\vec{q} + \vec{G}, \vec{q} + \vec{G}') = \epsilon_0(\vec{q} + \vec{G}) \delta_{\vec{q}, \vec{q}'} - v(\vec{q} + \vec{G}) \chi'(\vec{q} + \vec{G}, \vec{q} + \vec{G}'), \quad (11)$$

where $\epsilon_0(\vec{q} + \vec{G})$ contains the free-electron part

$$-v(\vec{q} + \vec{G}) \sum_{\vec{k}} \frac{f(n_0, \vec{k}) - f(n_0, \vec{k} + \vec{q})}{E(n_0, \vec{k}) - E(n_0, \vec{k} + \vec{q})}$$

of the intraband transition in the s -type conduction band n_0 (corresponding to the plane wave $e^{i(\vec{k}\cdot\vec{r})}$ with reciprocal lattice vector $\vec{k} \equiv (0, 0, 0)$ of the OPW basis set). We now assume the combined band-structure scheme takes a simplified form which approximately corresponds to the band description of Hodges, Ehrenreich, and Lang and which consists of treating the OPW conduction-band states in a one-plane-wave approximation [with $\vec{k} \equiv (0, 0, 0)$] and the remaining d states in the

LCAO approximation with all hybridization neglected. In this band model (which will be discussed in detail in Paper II in the special case of the transition metal Pd) the susceptibility matrix χ' of Eq. (11) contains only the tight-binding d transitions which can again be written in the factorized form of Eq. (7). We finally obtain for the dielectric matrix

$$\epsilon(\vec{q} + \vec{G}, \vec{q} + \vec{G}') = \epsilon_0(\vec{q} + \vec{G}) \delta_{\vec{G}, \vec{G}'}, \\ - v(\vec{q} + \vec{G}) \sum_{s, s'} A_s(\vec{q} + \vec{G}) N_{ss'}(\vec{q}) A_{s'}^*(\vec{q} + \vec{G}'), \quad (12)$$

where in this case the index s denotes the lattice vector index l and the indices ν and μ of the tight-binding band complex. The idea behind this splitting (which is *not* necessary in the exact Wannier function treatment above) is to pick out the smooth part of the conduction-band wave function in a metal and treat it separately. If we would include this part in the factorization procedure, then the number of lattice vectors \vec{R}' and \vec{R}'' needed in the summation of Eq. (12) would be very large, and therefore a practical solution of the inversion problem within the framework of the tight-binding approximation would be difficult. Using a similar matrix expansion as in Eq. (9), we get for the inverse dielectric matrix

$$\epsilon^{-1}(\vec{q} + \vec{G}, \vec{q} + \vec{G}') = \frac{1}{\epsilon_0(\vec{q} + \vec{G}')} \\ \times \left(\delta_{\vec{G}, \vec{G}'} + \frac{v(\vec{q} + \vec{G})}{\epsilon_0(\vec{q} + \vec{G})} \sum_{s, s'} A_s(\vec{q} + \vec{G}) \right. \\ \left. \times S_{ss'}^{-1}(\vec{q}) A_{s'}^*(\vec{q} + \vec{G}') \right), \quad (13)$$

where the matrix S is now defined by

$$S_{ss'}(\vec{q}) = N_{ss'}^{-1}(\vec{q}) - \sum_{G''} A_s^*(\vec{q} + \vec{G}'') A_{s'}(\vec{q} + \vec{G}'') \frac{v(\vec{q} + \vec{G}'')}{\epsilon_0(\vec{q} + \vec{G}'')}. \quad (14)$$

There exist several possibilities of going beyond the simplified one-plane-wave LCAO band description and still retaining the separable form of the dielectric matrix in Eq. (12). It is not the purpose of this paper to discuss all these possibilities, because here we are mainly interested in the factorization and inversion of the dielectric matrix in a localized or Wannier representation. But we would like to point out that by employing the combined OPW-LCAO scheme in the form proposed by Mueller,¹⁶ a separable dielectric matrix can be established by assuming all OPW and OPW-tight-binding interactions to be given in the diagonal (in \vec{G} and \vec{G}') form and to be incorporated in the diagonal matrix $\epsilon_0(\vec{q} + \vec{G}) \delta_{\vec{G}, \vec{G}'}$. Then the main contributions to off-diagonal screening which are due to d - d tight-binding interactions can again be calculated in using Eq. (13). It should be mentioned that a similar inversion result for the di-

electric matrix has been given by Sinha, Gupta, and Price.¹⁸ But in their paper the factorization of the susceptibility matrix is an *ansatz* and therefore cannot be discussed in terms of Wannier or LCAO functions.

In conclusion we note that the above inversion formalism enables us to calculate explicitly local-field corrections in the dielectric response, i. e., the induced electric fields which fluctuate on the scale of the atoms involved rather than with the wavelength of the applied field. The applicability of this important result is not limited to a calculation of phonon spectra where we specifically consider the electronic response to the external field of the nuclei or ions.

III. DYNAMICAL MATRIX

We suppose the adiabatic and harmonic approximation to be valid for the crystal considered. Then, for a system of interacting electrons and nuclei, the dynamical matrix is determined completely by the inverse dielectric matrix $\epsilon^{-1}(\vec{q} + \vec{G}, \vec{q} + \vec{G}')$ of the electrons¹⁻³

$$D_{\alpha\beta}(\vec{q}) = \bar{D}_{\alpha\beta}(\vec{q}) - \bar{D}_{\alpha\beta}(0)$$

with

$$\bar{D}_{\alpha\beta}(\vec{q}) = \frac{4\pi Z^2 e^2}{\Omega_0} \sum_{\vec{G}, \vec{G}'} \frac{(\vec{q} + \vec{G})_\alpha (\vec{q} + \vec{G}')_\beta}{|\vec{q} + \vec{G}'|^2} \\ \times \epsilon^{-1}(\vec{q} + \vec{G}, \vec{q} + \vec{G}') \\ = \frac{4\pi Z^2 e^2}{\Omega_0} \sum_{\vec{G}} \frac{(\vec{q} + \vec{G})_\alpha (\vec{q} + \vec{G})_\beta}{|\vec{q} + \vec{G}|^2} \\ + N \sum_{\vec{G}, \vec{G}'} (\vec{q} + \vec{G})_\alpha V(\vec{q} + \vec{G}) \chi(\vec{q} + \vec{G}, \vec{q} + \vec{G}') \\ \times V(\vec{q} + \vec{G}') (\vec{q} + \vec{G}')_\beta, \quad (14)$$

where

$$\chi_{\vec{G}, \vec{G}'} = \frac{-\delta_{\vec{G}, \vec{G}'} + \epsilon_{\vec{G}, \vec{G}'}^{-1}}{v(\vec{q} + \vec{G})}$$

denotes the density response matrix. The structure of this formula is exactly preserved if we introduce, instead of the Coulomb potential $V(\vec{q} + \vec{G}) = -4\pi Z e^2 / N \Omega_0 |\vec{q} + \vec{G}|^2$ of the nucleus, a local pseudopotential $W(\vec{q} + \vec{G})$ of an ion.¹⁹ In this local description as well as in the general nonlocal pseudopotential formulation, the main problem in connection with phonon calculations is associated with finding the inverse of ϵ in order to correctly account for local-field effects.

Armed with the knowledge of the RPA inversion result, we are now in a position to construct an explicit expression for the dynamical matrix in terms of generalized Wannier functions. From Eqs. (8), (13), and (14) we have

$$\bar{D}_{\alpha\beta}(\vec{q}) = C_{\alpha\beta}(\vec{q}) + E_{\alpha\beta}(\vec{q})$$

$$+ N \sum_{s, s'} W_{\alpha, s}(\vec{q}) S_{ss'}^{-1}(\vec{q}) W_{\beta, s'}^{\dagger}(\vec{q}), \quad (15)$$

where

$$W_{\alpha, s}(\vec{q}) = \sum_{\vec{G}} (\vec{q} + \vec{G})_{\alpha} A_s(\vec{q} + \vec{G}) \frac{V(\vec{q} + \vec{G})}{\epsilon_0(\vec{q} + \vec{G})}.$$

This formula is the same as that given in Ref. 5 for the dynamical matrix, the only difference being that the localized functions involved in Eq. (15) are constructed from the band complexes rather than from single energy bands. In Eq. (15), $C_{\alpha\beta}(\vec{q})$ denotes the Coulomb matrix and corresponds to the electrostatic coupling between the nuclei, while $E_{\alpha\beta}(\vec{q})$ is the expression used for the dynamical matrix of simple metals with a scalar screening function $\epsilon_0(\vec{q} + \vec{G})$. In the case of such a simple metal ϵ_0 is usually approximated by the free-electron dielectric function and the terms $A_s(\vec{q} + \vec{G}) N_{ss'}(\vec{q}) A_s^*(\vec{q} + \vec{G}')$ of the dielectric matrix, Eq. (12), are neglected. This corresponds to the free-electron model of lattice dynamics. Putting $\epsilon_0(\vec{q} + \vec{G}) = 1$ for ideal insulators leads to vanishing $E_{\alpha\beta}(\vec{q})$ and to dipole models, where the dipole distributions are not necessarily centered on the lattice sites.^{5,8} For noble and transition metals both the $\epsilon_0(\vec{q} + \vec{G})$ and $A_s(\vec{q} + \vec{G}) N_{ss'}(\vec{q}) A_s^*(\vec{q} + \vec{G}')$ are important ingredients of $\epsilon(\vec{q} + \vec{G}, \vec{q} + \vec{G}')$. Here the electron-electron interactions $v(\vec{q} + \vec{G})$ are screened by the scalar function $\epsilon_0(\vec{q} + \vec{G})$ and we get a kind of "screened multipole model".⁵ In this way, Eq. (15) for the dynamical matrix enables us to examine the validity of the various methods and models used to describe lattice vibrations in all these solids and to study their explicit relationship to each other. In this context it should be mentioned that one can derive from this general model a "shell model" where the dipole distributions are centered at the atom sites.^{20,21} This microscopic shell model is not only valid in ionic crystals and here provides a quantum-mechanical justification for the commonly used macroscopic shell-model treatments of phonons in these crystals. It is also applicable as a kind of "screened shell model" to metals.^{20,21} It is interesting to note that corresponding macroscopic shell-model calculations

in the noble and transition metals Cu, Ag, Au, Ni, and Pd yielded excellent agreement with experiment.²² In the application of our lattice-dynamics formalism to Pd and paramagnetic Ni in the following paper II we introduce approximations which are equivalent to the assumptions necessary for a microscopic shell model to hold.

IV. SUMMARY

We have examined in this paper the harmonic and adiabatic theory of lattice dynamics in terms of generalized Wannier functions. The Wannier representation allows for an exact inversion of the infinite-dimensional dielectric matrix within the RPA and thus makes possible an explicit calculation of local-field effects in the dielectric response. This result leads, with the help of the dynamical matrix of an electron-nucleus system, to a screened dipole model of lattice dynamics. Because we have considered the general case of a crystal the relevant energy bands of which may overlap or touch each other, the formalism can be applied to simple and transition metals as well as to semiconducting and ionic crystals. Thus our approach provides a unified treatment of the theory of lattice dynamics in all these solids, showing for example, that the equations of motion of simple metals and ionic crystals are in a way two limiting cases of the same formalism. It is possible to analyze the results of this microscopic formalism so that the connection to the familiar phenomenological theories (shell model) can be made.

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where a is the lattice constant.

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