

## Generation of lattice Wannier functions via maximum localization

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A method is presented for generating approximate lattice Wannier functions (LWFs) for lattice dynamics problems, using the dynamical matrix for a supercell as input. The lattice Wannier functions fit selected phonon frequencies and eigenvectors exactly, are orthonormal, and are optimized to be maximally localized. The method easily generalizes to the case where LWF centered on more than one distinct chemical species are desired, as well as to the case of solid solutions. The method is successfully applied to a one-dimensional toy model.

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### I. INTRODUCTION

In many lattice dynamics problems, the temperature-dependent physical properties are dominated by the low-frequency phonons. For example, the low-temperature  $c_v \propto T^3$  heat capacity relationship in crystals arises from the linear dispersion of acoustic phonons near zero wave vector. A second example is the dielectric constant of dielectrics. The phonon contribution to the dielectric tensor for a crystal can be written in the form<sup>1,2</sup>

$$\kappa_{\alpha\beta} = \frac{1}{V\epsilon_0 m_0} \sum_{\omega_\mu^2 \neq 0} \frac{\overline{Z}_{\mu\alpha}^* \overline{Z}_{\mu\beta}^*}{\omega_\mu^2}, \quad (1.1)$$

where  $V$  is the unit cell volume,  $\epsilon_0$  the permittivity of free space,  $\omega_\mu$  the (angular) frequency of phonon  $\mu$ ,  $\overline{Z}_\mu^*$  the electric polarization induced by phonon mode  $\mu$  divided by the phonon amplitude, and  $m_0$  an arbitrary mass that appears in the definition of  $\overline{Z}_\mu^*$ . In high- $\kappa$  materials, the dielectric constant is typically dominated by the contribution of low-frequency zone-center optical phonons. A third example is ferroelectric phase transitions. Typically in ferroelectrics, the ground state is largely determined by the freezing in of a particular mode which is *unstable* in the paraelectric phase.<sup>3</sup> To the extent that the anharmonic coupling of the instabilities of the paraelectric phase to other modes is small, the thermodynamics of the ferroelectric phase transition is determined by the properties of the unstable modes.<sup>4</sup> In all of the above cases, the number of degrees of freedom of the lattice dynamics problem, as it affects the temperature-dependent physical properties, can be greatly reduced by including only those degrees of freedom corresponding to the relevant phonons.<sup>5</sup> Furthermore, as shown by Rabe and Waghmare,<sup>4</sup> these degrees of freedom can be spanned by a localized “lattice Wannier function” (LWF) basis set. The projection of the original Hamiltonian onto the LWF yields the harmonic lattice dynamical part of an “effective Hamiltonian” in a form amenable to Monte Carlo and molecular dynamics simulations.

Despite their usefulness, the generation of LWF to date has been largely done on a case-by-case basis. Difficulties in making the generation of LWFs more automatic include

symmetrization, localization,<sup>6</sup> basis set completeness, and, most importantly, the band-mixing or “entangled-band” problem. Souza, Mazari, and Vanderbilt<sup>7</sup> discussed the entangled-band problem for the case of electronic Wannier functions. Rabe and Caracas<sup>8</sup> discussed the entangled-band problem for lattice Wannier functions and concluded that a practical solution is to impose LWF locality and to fit only the relevant (i.e., low frequency) parts of the phonon band(s) exactly.

In many cases, materials with the most useful properties are solid solutions. For example, temperature stability in dielectrics for microwave resonators typically requires solid solutions. Ultrahigh piezoelectric constants are found in solid solutions of  $\text{Pb}[\text{Mg}, \text{Zn}]_{1/3}\text{Nb}_{2/3}\text{O}_3$  with  $\text{PbTiO}_3$ . For such systems, LWFs designed for solid solutions would be useful in elucidating the physics responsible for their properties.

In this paper, an automatic procedure is given for generating LWFs. As in the previous work by Rabe and Caracas,<sup>8</sup> the key idea is to impose LWF locality, while fitting only the relevant part of the phonon bands exactly (although it may be desirable to include additional phonons). Furthermore, by eliminating symmetry as a consideration in generating the LWF, and using the principle of maximum localization only, the method easily generalizes to solid solutions.

### II. METHOD

To generate the LWF of a solid, one begins with phonon information. Although it is desirable in principle to know the full phonon dispersion (including eigenvectors), this is not usually possible in practice. Instead, the method presented here gives an approximate LWF based on phonon results for finite supercells. The required input is the eigenvalues and eigenmodes of a supercell dynamical matrix, as might be determined from first-principles (FP) calculations on the supercell, FP linear response calculations on a primitive cell, or “interpolated” results for a larger supercell using FP interatomic forces obtained for smaller (super)cells.<sup>9</sup>

For a supercell with  $N$  atoms, let  $i$  label the  $DN$  atomic coordinates (where  $D$  is the dimensionality of the system, and, for simplicity, the position  $\mathbf{r}_i$  and Cartesian direction coordinate  $\hat{a}_i$  are folded into a single label) and  $j$  the  $DN$

normal modes. Let  $v_j$  be the  $j$ 'th dynamical matrix eigenvalue (proportional to the squared frequency) and  $e_{ji}$  be the  $i$ 'th component of the (normalized) dynamical matrix eigenvector for mode  $j$ .

The LWF determination problem is to fit the  $v_j$  and  $e_{ji}$  for a chosen subset of modes  $\{u_l\} \subset \{v_j\}$ , using a set of supercell-periodic functions  $w_{ki}$ , where  $w_{ki}$  is the displacement of coordinate  $i$  in basis function  $w_k$ . The  $w_k$  become the exact lattice Wannier functions in the limit of an infinite supercell.

There is no absolute criteria for which modes  $u_l$  should be included, but, for problems where the physics is dominated by the low-frequency modes, it is necessary to include those modes for which  $v_j$  is less than some cutoff value. (As discussed in Sec. IV, it may be desirable to include some additional modes.) Note that the supercell approach, in effect, replaces continuous phonon dispersion branches with information on a discrete grid in  $q$ -space. In fact, it is this focus on individual phonons rather than phonon branches that makes it so easy to generalize to solid solutions, etc.

There are also no absolute criteria for the set of positions  $\{\mathbf{r}_k\}$  on which to center the  $\{w_{ki}\}$ . In many cases, certain atomic species displaced in certain Cartesian directions dominate the eigenvectors of the included modes. In such cases, it is natural to have the LWF set comprise each site  $\{\mathbf{r}_k\}$  on which an atom of the given species sits and each important direction of displacement  $\hat{\alpha}_k$ .

To fit the selected eigenfunctions of the original matrix exactly, one seeks (unknown) coordinates  $a_{lk}$  and  $w_{ki}$  such that  $\sum_k a_{lk} w_{ki} = e_{li}$ . There is no unique solution for  $w_{ki}$  because different choices of  $\{a_{lk}\}$  lead to different  $w_{ki}$ . To find the "optimal" set of  $w_{ki}$ , a "localization criterion" is applied, namely, to minimize

$$\sum_{ki} w_{ki}^2 d_{ki}^2. \quad (2.1)$$

The distance metric  $d_{ki}^2$  here is chosen to be  $d_{ki}^2 = \min[(r_k - r_i + \mathbf{R}_{\text{supercell}})^2]$ . Other metrics may be considered, for example, "anisotropic" metrics where  $d_{ki}^2$  is a function of  $\hat{\alpha}_k$  and  $\hat{\alpha}_i$  as well as the distance between the LWF centers.

Additional constraints are imposed to keep the functions  $w_k$  orthonormal and the coordinate sets  $a_{lk}$  orthonormal:  $\sum_i w_{ki} w_{mi} = 1$ ,  $k=m$ ;  $\sum_i w_{ki} w_{mi} = 0$ ,  $k \neq m$ ;  $\sum_k a_{lk} a_{nk} = 1$ ,  $l=n$ ;  $\sum_k a_{lk} a_{nk} = 0$ ,  $l \neq n$ . The problem is then set up as a constrained minimization problem. Using Lagrange multipliers, we write

$$\begin{aligned} f = & \sum_{ki} w_{ki}^2 d_{ki}^2 + \sum_{li} \lambda_{1li} \left( \sum_k a_{lk} w_{ki} - e_{li} \right) \\ & + \sum_k \lambda_{2kk} \left( \sum_i w_{ki} w_{ki} - 1 \right) + \sum_{k < m} \lambda_{2km} \left( \sum_i w_{ki} w_{mi} \right) \\ & + \sum_l \lambda_{3ll} \left( \sum_k a_{lk} a_{lk} - 1 \right) + \sum_{l < n} \lambda_{3ln} \left( \sum_k a_{lk} a_{nk} \right). \end{aligned}$$

One then needs to solve  $\partial f / \partial w_{ki} = 0$ ;  $\partial f / \partial a_{lk} = 0$ ;  $\partial f / \partial \lambda_{1li} = 0$ ;  $\partial f / \partial \lambda_{2k \leq m} = 0$ ;  $\partial f / \partial \lambda_{3l \leq n} = 0$ .

Since the above partial derivatives are nonlinear, an analytic solution does not exist in general. To extract a solution numerically, it is simpler instead to minimize the function,



FIG. 1. Primitive cell of a one-dimensional toy model for lattice dynamics.

$$\begin{aligned} F = & 1 + \sum_{ki} (\partial f / \partial w_{ki})^2 + \sum_{lk} (\partial f / \partial a_{lk})^2 + \sum_{li} (\partial f / \partial \lambda_{li})^2 \\ & + \sum_{k \leq m} (\partial f / \partial \lambda_{2km})^2 + \sum_{l \leq n} (\partial f / \partial \lambda_{3ln})^2. \end{aligned}$$

By construction,  $F$  is minimized to 1 if and only if all partial derivatives of  $f$  are zero. There may, however, be more than one solution. A hypothesis for a reasonable starting guess for the variables is  $w_{ki} = 1$  if  $\mathbf{r}_k = \mathbf{r}_i$  and  $\hat{\alpha}_k = \hat{\alpha}_i$ , else  $w_{ki} = 0$ ;  $a_{lk} = c_l e_{li}$  for  $i$  such that  $\mathbf{r}_k = \mathbf{r}_i$  and  $\hat{\alpha}_k = \hat{\alpha}_i$ , with normalization constants  $c_l$  such that  $\sum_k a_{lk}^2 = 1$ , and all Lagrange multipliers set to zero.

After the above results are obtained, the dynamical matrix  $D$  can be replaced by  $D'$ , a reduced dynamical matrix over LWF variables. The components of  $D'$  are, in bra-ket notation,

$$D'_{km} = \langle w_k | D | w_m \rangle. \quad (2.2)$$

It is straightforward to show that

$$\langle a_l | D' | a_n \rangle = \langle e_l | D | e_n \rangle = \delta_{ln} v_l; \quad (2.3)$$

that is, the selected eigenvalues of the original dynamical matrices are also eigenvalues of  $D'$ , and the corresponding eigenvectors are related to the origin dynamical matrix eigenvectors through the LWF.  $D'$  also gives the harmonic lattice terms of an effective Hamiltonian for Monte Carlo or molecular dynamics simulations.

### III. MODEL

The methods of this paper will be applied to a toy model for the lattice dynamics of a one-dimensional chain ( $D=1$ ). Figure 1 shows the toy linear chain model, created so that the phonon dispersion would be qualitatively similar to that often observed in ferroelectric perovskites such as  $\text{BaTiO}_3$ .<sup>10</sup> The chain has four atoms per primitive cell: an "A" or "A'" cation at each  $x = \text{integer } n$ , a "B" or "B'" cation at  $x = n + 0.5$ , and "C" anions at  $x = n + 0.25$  and  $x = n + 0.75$ . For simplicity, all ions have mass  $m=1$ , and the length of the unit cell is set to  $a=1$ . Defect and solid solution phenomena are incorporated into the model through the distribution of  $[A, A']$  and  $[B, B']$  ions on the corresponding sublattices.

The intersite force constants are given in Table I. The force constants involving A' and B' are set to mimic certain common characteristics of perovskite solid solutions. B' is designed to be chemically very similar to B and to create relatively minor perturbations of the phonon dispersion, as might occur for the substitution of one similarly sized isovalent ion for another. A' is designed so that instabilities involving A' offcentering (but not A offcentering) will occur, as happens when Li substitutes for K on the perovskite A site of  $[\text{K, Li}]\text{TaO}_3$ .

TABLE I. Interatomic force constants in model. For C—C interactions at distance 0.50, (A) and (B) indicate which ion is between the two “C” ions.

1	2	$d_{12}$	FC	1	2	$d_{12}$	FC
A, A'	A, A'	1.00	-37.5	B, B'	C	0.25	25.0
A, A'	B, B'	0.50	-15.0	B, B'	C	0.75	25.0
A	C	0.25	-15.0	C	C	0.50(A)	-140.0
A'	C	0.25	70.0	C	C	0.50(B)	40.0
A, A'	C	0.75	-15.0	C	C	1.00	-55.0
B, B'	B, B'	1.00	-107.5				
B	B, B'	2.00	28.75				
B'	B'	2.00	37.5				

Figure 2 shows the dynamical matrix eigenvalue dispersion for the “ideal”  $ABC_2$  chain. There is a single unstable mode at  $q=0$ , dominated by B participation. The instability only extends through part of the Brillouin zone (BZ). The mode with largest B participation at  $q=\pi/a$  is not the lowest one, but rather the second-highest one (eigenvalue 360). Note the following analogies with the case of  $BaTiO_3$ .<sup>5,10</sup> In  $BaTiO_3$ , there are instability branches dominated by Ti motion that do not extend throughout the entire BZ. Ti-dominated zone-boundary modes, where they occur, are not always the lowest-frequency modes at these points.

#### IV. RESULTS

The procedure for generating LWFs was applied to four structures within the toy model: (1) the ideal  $ABC_2$  cell, (2) a doubled cell with composition  $AB_{1/2}B'_{1/2}C_2$  (“ $AB_{1/2}B'_{1/2}C_2$ -ordered”), (3) an octupled cell with composition  $AB_{1/2}B'_{1/2}C_2$ , and the B and B' cations arranged in the quasirandom<sup>11</sup> arrangement  $BBBB'B'B'BB'$  (“ $AB_{1/2}B'_{1/2}C_2$ -quasirandom”), and (4) an octupled cell with the composition  $A_{7/8}A'_{1/8}BC_2$ .

For a one-dimensional supercell of period  $Na$ , the BZ goes from  $-\pi/(Na)$  to  $\pi/(Na)$ . It is possible to plot the dispersions of all the examples studied in this work in a common zone from  $-\pi/(8a)$  to  $\pi/(8a)$ , but that yields complicated diagrams with 32 bands in each case. For simplicity,

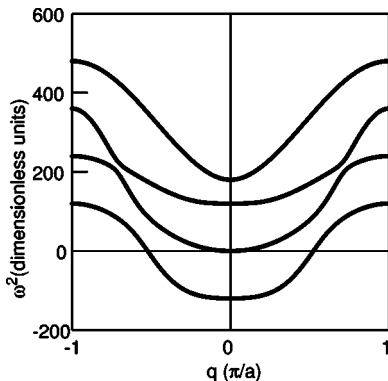


FIG. 2. Phonon dispersion for the ideal  $ABC_2$  structure.

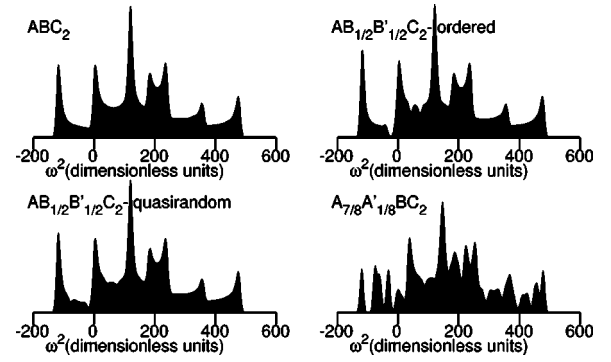


FIG. 3. Density of states of  $\omega^2$  for the configurations studied.

we compare instead the density of states (DOS) in each case, as shown in Fig. 3.

Relative to the DOS in the  $ABC_2$  structure, the replacement of B with B' in  $AB_{1/2}B'_{1/2}C_2$ -ordered leads to a gap in the DOS just below  $\omega^2=0$ , a typical phenomenon in period-doubling perturbations. While the DOS in the quasirandom  $AB_{1/2}B'_{1/2}C_2$  structure is similar, there is only a pseudogap, and the structure of the DOS is more complicated. The replacement of A with A' in the  $A_{7/8}A'_{1/8}BC_2$  structure gives an additional instability branch which is nearly dispersionless. Because of coupling of A' to the other ions, the DOS is strongly perturbed over the whole frequency range, with many more singularities.

LWFs were then generated for each structure. In each case, a (super)cell of length  $8a$  containing 32 ions was sufficient to generate LWFs that reproduced very well the phonon DOS in the low-frequency bands. Each structure had a similar set of optical low-frequency modes dominated by B, B' and with  $\omega^2 < 10$ . Based on the analogy of the toy model to lattice dynamics for ferroelectric transitions, where the modes of interest are the low frequency optical modes (in particular, the instabilities), all supercell optical modes with  $\omega^2 < 10$  were included in the LWF fits. This frequency range encompasses the additional A'-dominated instability branch in the  $A_{7/8}A'_{1/8}BC_2$  structure. Based on the ions whose motion dominates the low-frequency modes, LWFs centered on B and B' sites were included in all cases. One additional LWF, centered on the A' site, was included for the  $A_{7/8}A'_{1/8}BC_2$  structure. In each case, there is a van Hove singularity in the phonon DOS at  $\omega^2 \approx 360$  arising from  $q=\pi/a$ -type B-dominated modes. Based on the principle that the singularities in the phonon DOS of the LWF model should match the singularities in the original DOS as much as possible, the corresponding mode was included in each fit. Note that there is no absolute criterion requiring that these modes should be included. As long as the number of modes fit per supercell is less than or equal to the number of LWF centers, the procedure will generate LWFs that reproduce these modes, and as long as the phonon DOS in the region that affects the physical properties is correctly reproduced, it does not matter which higher-frequency modes are included.

In each case, the function  $F$  converges to 1. Using  $D'$  and setting the interactions to zero for distances larger than the maximum distance in  $D'$  allows phonon DOS and dispersions to be calculated from the effective Hamiltonian gener-

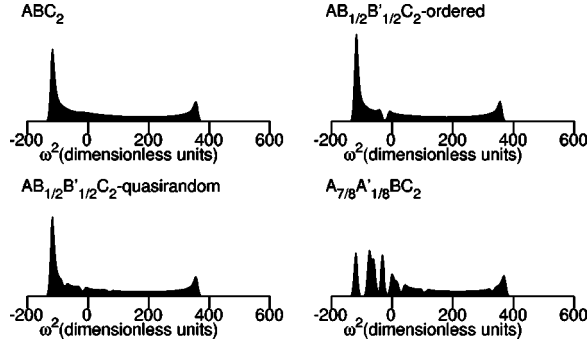


FIG. 4. Density of states of  $\omega^2$  for the effective Hamiltonians generated from the LWF for each configuration studied.

ated for each structure. For each case, the calculated DOS are shown in Fig. 4. The dispersion generated from the LWFs for  $ABC_2$  is shown in Fig. 5 and compared with the original full phonon dispersion. It reproduces the unstable part of the original model extremely well and rises smoothly to match the A-dominated mode at  $q = \pi/a$ . The values for the LWF displacement of the central cations and their nearest-neighbor C anions are given in Table II.

## V. DISCUSSION

The procedure for generating LWFs succeeds in reproducing the low-frequency phonon DOS for all of the one-dimensional model test cases. The phonon dispersion in the Brillouin zone or reduced Brillouin zone is also reproduced in each case. The results provide strong evidence that LWFs can be generated for arbitrary cation orderings in solid solutions.

In principle, the method should work in three dimensions. Polarization of phonon eigenvectors in dimensions  $D > 1$ , and the corresponding possibility for multiple LWF components centered on the same ion, are technical issues to be resolved. Also, the method requires minimization of a function of  $(2DNn_\mu + n_\mu n_w + n_w^2 + n_w/2 + n_\mu^2 + n_\mu/2)$  dimensions, where  $n_\mu$  is the number of modes to be fit and  $n_w$  is the

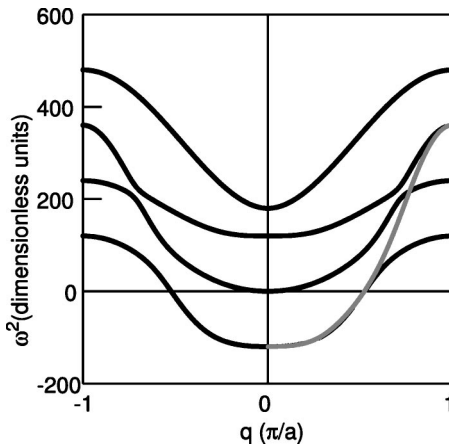


FIG. 5. (Left side) Dispersion in  $\omega^2$  of  $ABC_2$ . (Right side) Same as left side, with the effective Hamiltonian dispersion added (in gray).

TABLE II. Lattice Wannier functions generated from each structure. Although the LWFs extend over the unit cells only the components for ions within distance 0.25 from the central ion are shown. Structures: (1)  $ABC_2$ ; (2)  $AB_{1/2}B'_{1/2}C_2$ -ordered; (3)  $AB_{1/2}B'_{1/2}C_2$ -quasirandom; (4)  $A_{7/8}A'_{1/8}BC_2$ .

structure	ion	position	$w_{x-0.25}$	$w_x$	$w_{x+0.25}$
1	B	(all)	-0.233	0.860	-0.233
2	B	(all)	-0.232	0.860	-0.232
2	B'	(all)	-0.206	0.898	-0.206
3	B	0.5	-0.231	0.859	-0.234
3	B	1.5	-0.237	0.860	-0.238
3	B	2.5	-0.234	0.859	-0.231
3	B'	3.5	-0.211	0.880	-0.226
3	B'	4.5	-0.235	0.860	-0.232
3	B'	5.5	-0.201	0.899	-0.201
3	B	6.5	-0.232	0.860	-0.235
3	B'	7.5	-0.226	0.880	-0.211
4	A'	0.0	-0.491	0.706	-0.491
4	B	0.5	-0.068	0.916	-0.234
4	B	1.5	-0.249	0.854	-0.231
4	B	2.5	-0.236	0.861	-0.211
4	B	3.5	-0.246	0.832	-0.263
4	B	4.5	-0.263	0.832	-0.246
4	B	5.5	-0.211	0.861	-0.236
4	B	6.5	-0.231	0.854	-0.249
4	B	7.5	-0.234	0.916	-0.068

number of LWF centers chosen. This gives a practical limit to how large a supercell can be chosen for the fit.

In the case of the  $ABC_2$  and  $AB_{1/2}B'_{1/2}C_2$ -ordered structures, the LWFs have “ $u$ ” symmetry under reflection, and the LWFs for the same species with different centers are related by translational symmetry. While these reflect the symmetry of the lattice, symmetry was not imposed by design. Rather, symmetry resulted from maximum localization. In fact, in the  $AB_{1/2}B'_{1/2}C_2$ -quasirandom and  $A_{7/8}A'_{1/8}BC_2$  structures, LWFs centered on ions that are not on sites with reflection symmetry do not have reflection symmetry, and LWFs on sites that are not symmetry-related are not translationally identical (see Table II). Remarkably, even though all the phonons selected for the fit preserve the center of mass of the crystal, the individual LWFs do not, in general, conserve the center of mass. The fact that LWFs centered on the same species are not in general related by symmetry means that, in the effective Hamiltonian for a solid solution, the interactions involving these LWF centers will be environment-dependent.

Tests where the centers of the LWFs were inappropriately chosen gave interesting results. For the  $A_{7/8}A'_{1/8}BC_2$  structure, if  $A'$  centers were not included in the LWF fits, neither the density of the low-frequency states nor the phonon dispersion were correctly reproduced. For the  $ABC_2$  structure, if the LWF centers were initially put on the A sites, the final solution had LWFs centered on the B sites. In all the structures investigated here, various different combinations of



species on which to center the LWFs were tested. In each case, the choice described in Sec. IV had a combination of small  $n_w$  and small localization function (2.1) that was clearly superior to the other choices. Complete automation of the LWF procedure will require algorithms to decide which low-frequency modes are essential to the fit, where to center the LWFs, and, perhaps, which higher-frequency modes to include in order to fit singularities in the phonon DOS as much as possible.

## VI. CONCLUSIONS

A simple, flexible, method has been presented for generating lattice Wannier functions and the harmonic lattice dy-

namical part of the corresponding effective Hamiltonians, based on constrained LWF localization. The method requires only that the user choose the sites on which to localize the LWFs and which eigenmodes of a supercell to exactly fit. When applied to various configurations of a toy one-dimensional model, the method reproduces the desired features of the full lattice dynamics problem in each case. The procedure works equally well for simple compounds, ordered solid solutions, and disordered solid solutions.

## ACKNOWLEDGMENT

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- <sup>1</sup>M. Born and K. Huang, *Dynamical Theory of Crystal Lattices* (Oxford University Press, Oxford, 1954).  
<sup>2</sup>E. Cockayne and B. P. Burton, Phys. Rev. B **62**, 3735 (2000).  
<sup>3</sup>M. E. Lines and A. M. Glass, *Principles and Applications of Ferroelectrics and Related Materials* (Oxford University Press, Oxford, 1977), Chap. 8.  
<sup>4</sup>K. M. Rabe and U. V. Waghmare, Phys. Rev. B **52**, 13 236 (1995).  
<sup>5</sup>W. Zhong, D. Vanderbilt, and K. M. Rabe, Phys. Rev. Lett. **73**, 1861 (1994).  
<sup>6</sup>J. Íñiguez, A. García and J. M. Pérez-Mato, Phys. Rev. B **61**, 3127 (2000).  
<sup>7</sup>I. Souza, N. Marzari, and D. Vanderbilt, Phys. Rev. B **65**, 035109 (2002).  
<sup>8</sup>K. Rabe and R. Caracas, “Maximally-localized lattice Wannier functions,” [http://www.tcm.phy.cam.ac.uk/~mdt26/talks/karin\\_rabe/karinrabe\\_wannier\\_functions.pdf](http://www.tcm.phy.cam.ac.uk/~mdt26/talks/karin_rabe/karinrabe_wannier_functions.pdf) (2002).  
<sup>9</sup>X. Gonze, J.-C. Charlier, D. C. Allan, and M. P. Teter, Phys. Rev. B **50**, R13 035 (1994).  
<sup>10</sup>Ph. Ghosez, E. Cockayne, U. V. Waghmare, and K. M. Rabe, Phys. Rev. B **60**, 836 (1999).  
<sup>11</sup>A. Zunger, S.-H. Wei, L. G. Ferreira, and J. E. Bernard, Phys. Rev. Lett. **65**, 353 (1990).