

The effects of nuclear reactions in solids on the phonon dispersion relation

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Abstract. In this study, theoretical predictions about the effects of nuclear reactions in solids on the phonon dispersion relations are shown. As an initial model, we have tried to treat one dimensional palladium deuteride and have obtained possible changes of phonon dispersion relations due to DD reactions. This method will be applied to the detection of nuclear reactions in solid by using nuclear scattering or Raman spectroscopy.

1. Introduction

In solid state physics, phonon dispersion relation is one of the most important information, because it includes not only the geometric and the electronic structures of the crystals but also the interaction between ions or electrons. For example, a lattice with only one atom in a primitive cell only has acoustic lattice vibration. However, a lattice with more than one atom in a primitive cell has acoustic and optical modes of vibration. Furthermore, interactions between impurity ions are also effective on the features of phonons. If nuclear reactions occur in solids, they will change the lattice and electronic structures and interactions between impurity ions and so on. Therefore, the changes of phonon dispersion relations will give us the evidences of nuclear reactions in solids. In this study, the possible changes of the phonon dispersion relation due to the nuclear reaction in solid were discussed. In this section, the theories of phonon dispersion relation of solids [1,2] are introduced briefly. Firstly, we define the equilibrium positions of ions in crystalline solids as $\mathbf{R}_{nm} = \mathbf{r}_n + \mathbf{k}_m$, where \mathbf{r}_n and \mathbf{k}_m mean position of the n-th primitive cell and position of the k-th ion in the cell, respectively. Then the total kinetic energy is defined using displacement vector \mathbf{u}_{nm} from \mathbf{R}_{nm} as

$$K = \frac{1}{2} \sum_{nmj} M_m (\dot{u}_{nm}^j)^2, \quad (1)$$

where M_m is the mass of the m-th ion in the primitive cell and the summation for j is done over Cartesian components. And the total potential energy is written in the 2nd order approximation as

$$V = V_0 + \frac{1}{2} \sum_{nmj} \sum_{n'm'j'} C_{nmn'm'}^{jj'} u_{nm}^j u_{n'm'}^{j'}, \quad (2)$$

where C is the second order partial derivative of V with respect to the displacement. They give the equation of motion for each ions which is written as

$$M_m \ddot{u}_{nm}^j = - \sum_{n'm'j'} C_{nmn'm'}^{jj'} u_{n'm'}^{j'}. \quad (3)$$

In this equation, every components of the displacement satisfy Bloch theorem

$$u_{nm}^j = e^{i\mathbf{q} \cdot \mathbf{r}_n} u_{0m}^j \quad (4)$$

and coefficient C has a symmetry

$$C_{nmn'm'}^{jj'} \rightarrow C_{m'm'n'n}^{j'j}(\mathbf{h}) \quad (\mathbf{h} = \mathbf{r}_{n'} - \mathbf{r}_n). \quad (5)$$

Defining the Fourier transformed formula as

$$\sum_{\mathbf{h}} C_{mm'}^{jj'}(\mathbf{h}) e^{i\mathbf{q}\cdot\mathbf{h}} \equiv G_{mm'}^{jj'}(\mathbf{q}) \quad (6)$$

and assuming the oscillatory solution as

$$\mathbf{u}_{nm}^j(t) = e^{-i\omega t} \mathbf{u}_{nm}^j(0), \quad (7)$$

we obtain phonon dispersion relation as

$$\sum_{m'=1}^M \sum_{j'=1}^3 G_{mm'}^{jj'}(\mathbf{q}) \mathbf{u}_{0m}^{j'}(0) = M_m \omega^2 \mathbf{u}_{0m}^j(0) \quad (8)$$

where M is the number of ions including in a primitive cell. For the case of a monatomic lattice which has only one ion in a primitive cell, eq.(8) is rewritten as

$$\begin{pmatrix} G^{11} & G^{12} & G^{13} \\ G^{21} & G^{22} & G^{23} \\ G^{31} & G^{32} & G^{33} \end{pmatrix} \begin{pmatrix} u^1 \\ u^2 \\ u^3 \end{pmatrix} = M\omega^2 \begin{pmatrix} u^1 \\ u^2 \\ u^3 \end{pmatrix}. \quad (9)$$

This equation has three solutions. One corresponds to the longitudinal mode, and the other two corresponds to the doubly-degenerated transverse mode. The typical solutions are sketched in Fig.1. For the case of a diatomic lattice with a heavy and a light ion in a primitive cell, eq.(8) is rewritten as

$$\begin{pmatrix} G_{11}^{11} & G_{11}^{12} & G_{11}^{13} & G_{12}^{11} & G_{12}^{12} & G_{12}^{13} \\ G_{11}^{21} & G_{11}^{22} & G_{11}^{23} & G_{12}^{21} & G_{12}^{22} & G_{12}^{23} \\ G_{11}^{31} & G_{11}^{32} & G_{11}^{33} & G_{12}^{31} & G_{12}^{32} & G_{12}^{33} \\ G_{21}^{11} & G_{21}^{12} & G_{21}^{13} & G_{22}^{11} & G_{22}^{12} & G_{22}^{13} \\ G_{21}^{21} & G_{21}^{22} & G_{21}^{23} & G_{22}^{21} & G_{22}^{22} & G_{22}^{23} \\ G_{21}^{31} & G_{21}^{32} & G_{21}^{33} & G_{22}^{31} & G_{22}^{32} & G_{22}^{33} \end{pmatrix} \begin{pmatrix} u_1^1 \\ u_1^2 \\ u_1^3 \\ u_2^1 \\ u_2^2 \\ u_2^3 \end{pmatrix} = \omega^2 \begin{pmatrix} M_1 u_1^1 \\ M_1 u_1^2 \\ M_1 u_1^3 \\ M_2 u_2^1 \\ M_2 u_2^2 \\ M_2 u_2^3 \end{pmatrix}. \quad (10)$$

The typical solutions are sketched in Fig.2. In real case, these relations should be calculated for each directions of wave propagation.

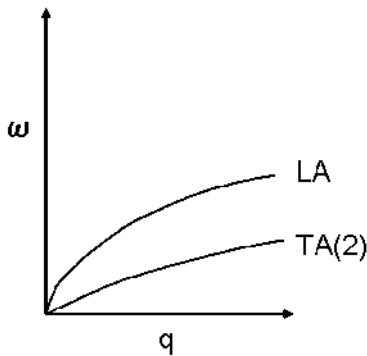


Fig. 1 - Phonon dispersion relation of monatomic lattice. Characters LA and TA(2) mean longitudinal acoustic and doubly-degenerate transverse acoustic modes, respectively.

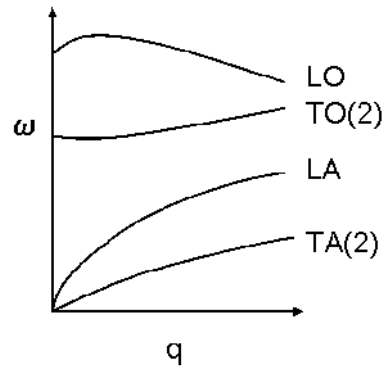


Fig. 2 - Phonon dispersion relation of diatomic lattice. Characters LA and TA(2) mean longitudinal acoustic and doubly-degenerate transverse acoustic modes, respectively. Characters LO and TO(2) mean longitudinal

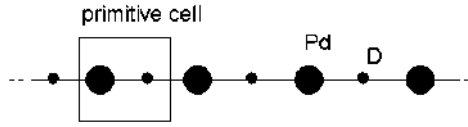


Fig. 3 - One-dimensional PdD lattice

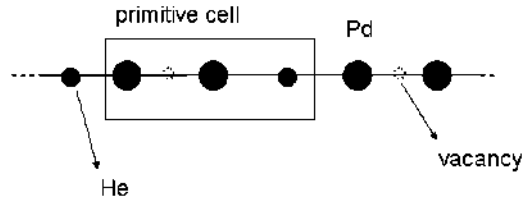


Fig. 4 - A model of structure changed one dimensional PdD lattice due to nuclear reaction

2. Change of the phonon dispersion relation due to the nuclear reaction

We discuss the phonon dispersion relation of palladium deuteride using the one dimensional PdD lattice. It is a very simple model which is illustrated in Fig.3. In this case, the dimension of the dynamical matrix is 2×2 with 2 solutions. The solutions are easily obtained by using the method described in section 1. And the typical dispersion curves are similar to Fig.2. If we consider some nuclear reactions and they change the structure of the lattice in Fig.3, they will also change phonon dispersion relation. For example as illustrated in Fig.4, if deuterons are disappeared and He's are produced, a primitive cell contains two palladiums, one He and one vacancy. This lattice roughly corresponds to the He concentration of 10^{22} atom/cm³ in the three-dimensional lattice. It is unrealistic situation. However, our model gives possible changes of phonon dispersion relation by assuming the lowest dimensional lattice and the highest deuteron concentration. For example, the occurrence of the nuclear reactions at a definite small domain with high deuteron concentration in the palladium deuteride may influence the phonon states of the whole lattice. At the moment, phonon dispersion relation will be changed under the influence from the domain. It will be detected by using neutron scattering or Raman spectroscopy. Of course it suggests the DD reaction. At this time, the dimension of the dynamical matrix is changed into 3×3 with 3 solutions. This means that some unknown phonon dispersion branches are created at the reaction. Therefore, if we measure the phonon dispersion relation of palladium deuteride and find unknown extra dispersion branches, they may be evidences of nuclear reactions. It is a model with oversimplification. In real case, more complicated and local reactions should be considered. For example, multi-body fusion or more complicated reaction [3] should be considered. However, the structure changes due to the nuclear reaction always tend to increase dispersion branches

Here, we would like to show the examples by doing one dimensional calculation of longitudinal mode for the lattices illustrated in Figs.3 and 4. Firstly, calculation for PdD lattice illustrated in Fig.3 has done. It is a simplified one dimensional lattice with alternate arrangement of Pd and D atoms. If we only consider the vibrations along the line, it means longitudinal mode. In this case, the total kinetic energy is written as

$$K = \frac{1}{2} M \sum_n \dot{u}_{2n}^2 + \frac{1}{2} m \sum_n \dot{u}_{2n+1}^2, \quad (11)$$

where M and m mean mass of Pd and mass of D, respectively. And the total potential energy in harmonic approximation with neighboring interaction is written as

$$V = \frac{1}{2} k \sum_n (u_n - u_{n+1})^2, \quad (12)$$

where k means elastic constant. Using them, the equations of motion are derived as

$$\begin{aligned} M \ddot{u}_{2n} &= -k(2u_{2n} - u_{2n+1} - u_{2n-1}) \\ m \ddot{u}_{2n+1} &= -k(2u_{2n+1} - u_{2n+2} - u_{2n}), \end{aligned} \quad (13)$$

where displacement u_n for the n -th atom is a function of time. Using a similar procedure with the general case in section 1, we obtain secular equation

$$\begin{vmatrix} 2k - M\omega^2 & -2k \cos(qa) \\ -2k \cos(qa) & 2k - m\omega^2 \end{vmatrix} = 0. \quad (14)$$

The well known solutions of eq.(14) are

$$\omega_{\pm}^2 = k \left(\frac{1}{m} + \frac{1}{M} \right) \pm k \sqrt{\left(\frac{1}{m} + \frac{1}{M} \right)^2 - \frac{4 \sin^2(qa)}{mM}}. \quad (15)$$

They give phonon dispersion relation which has one acoustic mode and one optical mode. Secondly, we consider one dimensional lattice illustrated in Fig.4. In this case the total kinetic energy is written as

$$K = \frac{1}{2} M \sum_n \dot{u}_{3n}^2 + \frac{1}{2} (2m) \sum_n \dot{u}_{3n+1}^2 + \frac{1}{2} M \sum_n \dot{u}_{3n+2}^2, \quad (16)$$

where the 1st, the 2nd and the 3rd terms mean total kinetic energy of Pd sitting on the left side of He, of He and of Pd sitting on the right side of He, respectively. The total potential energy is written as

$$V = \frac{1}{2} \alpha \sum_n (u_{3n+2} - u_{3n+1})^2 + \frac{1}{2} \alpha \sum_n (u_{3n+1} - u_{3n})^2 + \frac{1}{2} \beta \sum_n (u_{3n} - u_{3n-1})^2. \quad (17)$$

In this equation, the 1st and the 2nd terms mean interactions between neighboring Pd and He with elastic constant α . The 3rd term means interaction between neighboring Pd's with elastic constant β . Equations of motion for each atom are written as

$$\begin{aligned} M\ddot{u}_{3n} &= -(\alpha + \beta)u_{3n} + \alpha u_{3n+1} + \beta u_{3n-1} \\ 2m\ddot{u}_{3n+1} &= -\alpha(2u_{3n+1} - u_{3n+2} - u_{3n}) \\ M\ddot{u}_{3n+2} &= -(\alpha + \beta)u_{3n+2} + \beta u_{3n+3} + \alpha u_{3n+1}. \end{aligned} \quad (18)$$

And the secular equation is obtained as

$$\begin{vmatrix} \alpha + \beta - M\omega^2 & -\alpha e^{iq} & -\beta e^{-iq} \\ -\alpha e^{-iq} & 2\alpha - 2m\omega^2 & -\alpha e^{iq} \\ -\beta e^{iq} & -\alpha e^{-iq} & \alpha + \beta - M\omega^2 \end{vmatrix} = 0. \quad (19)$$

Expanding the secular equation, we obtain a cubic equation for ω^2 as

$$mM^2 \omega^6 - \{\alpha M + 2(\alpha + \beta)m\}M\omega^4 + \alpha(\alpha + 2\beta)(m + M)\omega^2 - \alpha^2 \beta \{1 - \cos(3ql)\} = 0, \quad (20)$$

which has three positive real solutions. They correspond to one acoustic mode and two optical modes. For the case of $q=0$, this equation reduces to

$$\omega_0^2 [mM^2 \omega_0^4 - \{\alpha M + 2(\alpha + \beta)m\}M\omega_0^2 + \alpha(\alpha + 2\beta)(m + M)] = 0. \quad (21)$$

The solution for acoustic mode is $\omega_0=0$ and the solutions for the optical mode are

$$\omega_{0\pm}^2 = \frac{\alpha M + 2(\alpha + \beta) \pm |\alpha M - 2\beta m|}{2mM}. \quad (22)$$

For the case of $\alpha M > 2\beta m$, we obtain

$$\omega_{0+}^2 = \alpha \left(\frac{1}{m} + \frac{1}{M} \right) \quad \text{and} \quad \omega_{0-}^2 = (\alpha + 2\beta) \frac{1}{M}. \quad (23)$$

For the case of $\alpha M < 2\beta$, we obtain

$$\omega_{0+}^2 = (\alpha + 2\beta) \frac{1}{M} \quad \text{and} \quad \omega_{0-}^2 = \alpha \left(\frac{1}{m} + \frac{1}{M} \right). \quad (24)$$

They give the difference of two optical mode at $q=0$. If we find these changes of dispersion relation, we can know the change of the structure from the lattice in Fig.3 to the lattice in Fig.4. This corresponds to the detection of some nuclear reactions in solids.

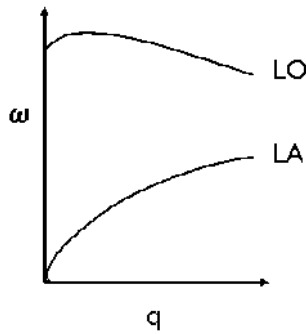


Fig. 5 - Phonon dispersion relation of the longitudinal vibration of the one dimensional lattice illustrated in Fig.3.

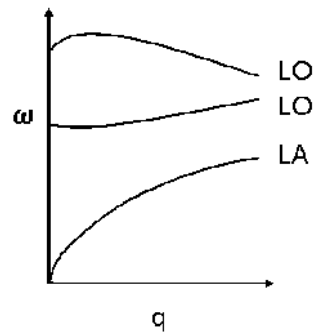


Fig. 6 - Phonon dispersion relation of the longitudinal vibration of the one dimensional lattice illustrated in Fig.4.

3. Conclusions

If some nuclear reactions have happened in solids, the lattice structure is changed and the number of the ions in a primitive cell is increased. Even if these changes are spatially inhomogeneous, this tendency is unaltered. In this case, the dimension of the dynamical matrix becomes larger and this causes an increase of dispersion branches. Therefore, if we measure the phonon dispersion relation of palladium deuteride and find unknown extra dispersion branches, they may be evidences of nuclear reactions. These discussions can be applied to the analysis of Raman observation of the palladium deuterides [4,5] .

Acknowledgments

The authors wish to thank Dr. A.Takahashi (Technova inc.) and Professor H.Yamada (Iwate Univ.) for helpful discussions and encouragements. We also wish to thank Professor N.Akuzawa (TNCT) for technical and financial supports to this study.

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