Anharmonicity and superconductivity in metal hydrides. I. Formulation

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The role of anharmonicity of the phonon spectrum in superconductivity is discussed. The anharmonic phonon-electron coupling is calculated under quasiharmonic approximation. A procedure for calculating this coupling from the neutron data is outlined. The superconductivity in metal-hydride (deuteride) systems is discussed in the light of aforementioned anharmonic phonon-electron coupling.

I. INTRODUCTION

In the recent past the author has formulated a phenomenological theory of superconductivity and opposite isotope effect in PdH(D) systems based upon a simple three-square-well-potential model.[1] In contradistinction to many other theories[2-4] the three-square-well-potential model takes into account the participation of relatively soft optic-phonon modes in BCS-type[5] coupling in palladium hydride (deuteride) systems. It was also pointed out that the anharmonicity emanating from the large zero-point amplitude of the hydrogen motion, relative to that of deuterium, is the main cause of the observed opposite isotope effect. I would like to point out, in passing, that the aforementioned explanation of the opposite isotope effect, which is based on tha anharmonic phonon spectrum, is basically distinct from the explanation of Miller and Satterthwaite,[6] which takes into account only the *differences in the electronic densities of states* in PdH compared to PdD and thus has nothing to do with the role of *anharmonic phonons* in the superconductivity of these systems. Recent tunneling experiments[7-9] prove decisively that the optic phonons are participating in the electron-phonon coupling in these systems. Moreover, the tunneling experiments of Dynes and Garno[8] found the ratio of hydrogen to deuterium optic-mode frequencies to be 1.5 instead of $\sqrt{2}$; the latter value is expected from a harmonic theory. The most recent neutron scattering experiment[10] shows the shift of the optic mode in $PdH_{0.63}$ well beyond a factor of $\sqrt{2}$ of $PdD_{0.63}$, indicating a strong anharmonic optic phonon in PdH. These experimental results prompt us to reiterate the statement that the major contribution to the opposite isotope effect is the change in the *phonon spectrum* due to anharmonicity leading to a different electron-phonon coupling and *not* primarily due to the purely *electronic density-of-states* change as proposed by Miller and Satterthwaite.[6]

Superconductivity in palladium–noble-metal–hydrogen (deuterium) systems has been reported[11] and the highest T_c attained is about 17°K. The mechanism of enhanced superconductivity and other related observations has been discussed qualitatively by the author in a recent publication.[12] It was pointed out in that paper that since the Grüneisen constant of the optic mode, as calculated[13] from the pressure dependence of T_c in PdH(D) systems, is quite small the effect of lattice expansion by noble-metal addition may have little effect on the position of the optic mode. However, heat-of-absorption experiments[14] suggest different energies for hydrogen bonding to the lattices of these alloy systems compared to pure palladium and this may result in the changes in the position as well as the widths in the optic-mode spectra in noble-metal alloys. The neutron scattering experiment on the Pd-Ag-H system[15] shows a broad bandwidth of the optic mode.

The role of *anharmonicity* in the theory of superconductivity was neglected in the past because the phonons in most of the superconductors can essentially be described by the harmonic approximation. Moreover, the isotope mass, which resulted in a change of 1 in hundreds of mass units, affected the phonon frequencies but did not violate the harmonic approximation. The situation is drastically different in the metal-hydride (deuteride) systems. Because of the large zero-point amplitude of hydrogen compared to that of deuterium and the large relative changes in the isotope mass, one must take into account appreciable anharmonic effects in the theory of superconductivity. The purpose of this paper is just that, that is, to incorporate the anharmonic phonon-electron interaction in the Eliashberg formalism of the theory of superconductivity. It will also be shown that the effects of anharmonicity on the electron-phonon coupling λ can be calculated from the available neutron scattering information. A quantitative calculation of λ for the PdH(D) systems is postponed until a future publication while the formulations and the procedures of such a calculation

will be discussed in Sec. II. The paper is organized in the following way: In Sec. II we describe briefly the relevant modifications of the Eliashberg[16]-McMillan[17] theory of superconductivity due to anharmonicity in the phonon spectrum. The anharmonic phonon Green's function and the corresponding spectral weight function are described in Sec. III. In Sec. IV the superconductivity in the metal-hydride (deuteride) systems is disucssed in light of the theory and the procedure for calculating the anharmonic phonon-electron coupling constant using neutron data is described.

II. ANHARMONICITY AND ELIASHBERG-MC MILLAN THEORY

In the Eliashberg's formalism of the theory of superconductivity the normal and the superconducting electronic self-energies involve the electron-phonon interaction function $\alpha^2(\omega_{\bar{q}})F(\bar{q}, \omega_{\bar{q}})$. This function can be expressed in terms of the phonon spectral function[18,19] and the electron-phonon matrix element as

$$\alpha^2(\omega_{\bar{q}})F(\bar{q}, \omega_{\bar{q}}) = \sum_{\nu} g^2_{\bar{k}\bar{k}'\nu} A(\bar{q}, \omega_{\bar{q}})\delta(\epsilon_{\bar{k}'}), \qquad (2.1)$$

where $g_{\bar{k}\bar{k}'\nu}$ is the electron-phonon matrix element, $A(\bar{q}, \omega_{\bar{q}})$ is the spectral weight function of the phonon spectrum, and $\bar{k}' = \bar{k} + \bar{q}$. In the deformation-potential approximation the electron-phonon matrix element is given by

$$g_{\bar{k}\bar{k}'\nu} = \left(\frac{\hbar}{2MVN\omega_{\bar{q}}}\right)^{1/2} g_{\nu}(\bar{k}, \bar{k}'), \qquad (2.2)$$

where g_{ν} is the well-known electronic overlap integral for phonon polarization vector ν between electron momentum states $\bar{k}$ and $\bar{k}'$. The δ function on the final state ensures that the final state is on the Fermi surface. The averaging of $\alpha^2(\omega_{\bar{q}})F(\omega_{\bar{q}})$ over the Fermi surface is given by

$$\alpha^2(\omega_{\bar{q}})F(\omega_{\bar{q}}) = \frac{\sum_{\bar{k}''}\alpha^2(\omega_{\bar{q}})F(\omega_{\bar{q}})\delta(\epsilon_{\bar{k}''})}{\sum_{\bar{k}''}\delta(\epsilon_{\bar{k}''})}. \qquad (2.3)$$

Use of the identities

$$\sum_{\bar{k}} \equiv \int d\epsilon_{\bar{k}} \int d\theta_{\bar{k}} (2\pi)^3 m\bar{k}$$

and

$$\int d\theta_{\bar{k}} m\bar{k} \equiv \int_S \frac{d^2\bar{k}}{v_F}$$

yields

$$\alpha^2(\omega_{\bar{q}})F(\omega_{\bar{q}}) = \int_S \frac{d^2\bar{k}}{v_F} \int \frac{d^2\bar{k}'}{v_F'(2\pi\hbar)^3}$$

$$\times \sum_{\nu} g^2_{\bar{k}\bar{k}'\nu} A(\bar{q}, \omega_{\bar{q}}) \bigg/ \int_S \frac{d^2\bar{k}}{v_F}. \qquad (2.4)$$

The electron-phonon coupling constant λ is given by the integral

$$\lambda = 2\int d\omega_{\bar{q}} \alpha^2(\omega_{\bar{q}})F(\omega_{\bar{q}})/\omega_{\bar{q}}. \qquad (2.5)$$

For harmonic phonons the spectral function $A(\bar{q}, \omega_{\bar{q}})$ is merely a δ function, given by

$$A(\bar{q}, \omega_{\bar{q}}) = \delta(\omega - \omega_{\bar{q}}). \qquad (2.6)$$

Substitution of (2.6) into (2.4) readily gives

$$\int_0^{\infty} d\omega_{\bar{q}}\, \omega_{\bar{q}}\alpha^2(\omega_{\bar{q}})F(\omega_{\bar{q}}) = N(0)\hbar\langle g^2_{\bar{k}\bar{k}'}\rangle/2M, \qquad (2.7)$$

where $\langle g^2 \rangle$ is the Fermi-surface average of the electronic overlap integral. One can use (2.7) in order to integrate (2.5), leading to the McMillan expression[17] for λ:

$$\lambda = N(0)\langle g^2\rangle/M\langle\omega_{\bar{q}}^2\rangle, \qquad (2.8)$$

where

$$\langle\omega_{\bar{q}}^2\rangle = \frac{\int d\omega_{\bar{q}}\, \omega_{\bar{q}}\alpha^2(\omega_{\bar{q}})F(\omega_{\bar{q}})}{\int d\omega_{\bar{q}}\alpha^2(\omega_{\bar{q}})F(\omega_{\bar{q}})/\omega_{\bar{q}}}. \qquad (2.9)$$

It is customary to assume $\alpha^2(\omega_{\bar{q}})$ to be independent of the isotopic mass. However, in the PdH(D) systems, because the amplitudes and the frequencies of H and D vibrations are quite different, the values of $\alpha^2(\omega_{\bar{q}})$ should differ.

For anharmonic phonons one has to calculate the spectral function $A(\bar{q}, \omega_{\bar{q}})$ in order to calculate $\alpha^2F(\omega)$ from Eq. (2.4). For anharmonic crystals, since the phonons are no longer independent normal modes of the system, the electron-phonon vertex is very complicated. Physically, the virtual phonon emitted by an electron at the Fermi surface couples with many phonons in the phonon bath through the anharmonic coupling. In this situation, to the author's knowledge, the modification of Eliashberg's formalism is not known. In what follows, we will restrict ourselves to the quasiharmonic approximation in which the third-order and the fourth-order anharmonicity will lead to the shift and broadening of the phonon modes keeping them independent normal modes of the crystal. The calculation of the phonon self-energy giving shift and broadening parameters is the subject of Sec. III.

III. ANHARMONICITY AND PHONON SPECTRAL FUNCTION

The spectral function $A(\bar{q}, \omega_{\bar{q}})$ is given in terms of the phonon Green's function $D(\bar{q}, \omega_{\bar{q}})$ by the relation

$$A(\bar{q}, \omega_{\bar{q}}) = -(1/\pi)\mathrm{Im}D(\bar{q}, \omega_{\bar{q}}). \qquad (3.1)$$

The calculation of the phonon Green's function for

the anharmonic crystal is summarized as follows: The Hamiltonian of the anharmonic crystal inclusive of third- and fourth-order anharmonicity is given by[20]

$$H_0 = \sum_{\vec{q}} \tfrac{1}{2}\hbar\Omega_{\vec{q}}(a_{\vec{q}}a_{\vec{q}}^* + a_{\vec{q}}^* a_{\vec{q}}),$$ (3.2)

$$H_A^{(3)} = \sum_{\vec{q}_1,\vec{q}_2,\vec{q}_3} V(\vec{q}_1\vec{q}_2\vec{q}_3)A_{\vec{q}_1} A_{\vec{q}_2} A_{\vec{q}_3},$$ (3.3)

$$H_A^{(4)} = \sum_{\vec{q}_1,\vec{q}_2,\vec{q}_3,\vec{q}_4} V(\vec{q}_1\vec{q}_2\vec{q}_3\vec{q}_4)A_{\vec{q}_1} A_{\vec{q}_2} A_{\vec{q}_3} A_{\vec{q}_4},$$ (3.4)

where $A_{\vec{q}} = a_{\vec{q}} + a_{\vec{q}}^*$, $a_{\vec{q}}^*$ and $a_{\vec{q}}$ being phonon creation and annihilation operators and

$$V(\vec{q}_1\vec{q}_2\vec{q}_3) = \frac{1}{6N^{1/2}} \Delta(\vec{q}_1 + \vec{q}_2 + \vec{q}_3)$$
$$\times \left(\frac{\hbar^3}{8\Omega_{\vec{q}_1}\Omega_{\vec{q}_2}\Omega_{\vec{q}_3}}\right)^{1/2} \Phi(\vec{q}_1\vec{q}_2\vec{q}_3),$$ (3.5)

$$V(\vec{q}_1\vec{q}_2\vec{q}_3\vec{q}_4) = \frac{1}{24N} \Delta(\vec{q}_1 + \vec{q}_2 + \vec{q}_3, \vec{q}_4)$$
$$\times \left(\frac{\hbar^4}{8\Omega_{\vec{q}_1}\Omega_{\vec{q}_2}\Omega_{\vec{q}_3}\Omega_{\vec{q}_4}}\right) \Phi(\vec{q}_1\vec{q}_2\vec{q}_3\vec{q}_4).$$ (3.6)

Here H_0 is the harmonic Hamiltonian, and $H_A^{(3)}$ and $H_A^{(4)}$ are the third- and the fourth-order an-

harmonic Hamiltonians, respectively. $\Omega_{\vec{q}}$ is the harmonic frequency of the crystal and Φ's are Coulomb coefficients defined by Born and Huang. Δ functions conserve the total crystal momentum. If the harmonic phonon frequencies of the system are known one can calculate the anharmonic effects by switching on the anharmonic interaction and calculating the third- and fourth-order potentials from (3.5) and (3.6).

The self-energy contributions from the third- and fourth-order diagrams are given by[20]

$$\Sigma^{(3)} = 18\beta^2 \sum_{\vec{q}_1\vec{q}_2} \sum_{n_1} V(-\vec{q}\vec{q}_1\vec{q}_2)V(\vec{q}' - \vec{q}_1 - \vec{q}_2)G_T(\vec{q}_1, i\omega_{n_1})$$
$$\times G_T(\vec{q}_2, i(\omega_n - \omega_{n_1})),$$ (3.7)

$$\Sigma^{(4)} = -12\beta \sum_{\vec{q}_1} \sum_n V(-\vec{q}\vec{q}'\vec{q}_1 - \vec{q}_1)G_T(\vec{q}_1, i\omega_n),$$ (3.8)

where G_T's are matrix elements of the thermodynamic Green's functions. The total self-energy can be expressed in terms of Hermitian and anti-Hermitian parts as

$$\Sigma(\vec{q}\vec{q}'\omega) = \beta\hbar\Delta(\vec{q}\vec{q}'\omega) + i\beta\hbar\Gamma(\vec{q}\vec{q}'\omega),$$ (3.9)

where

$$\Delta(\vec{q}\vec{q}'\omega) = \frac{12}{\hbar} \sum_{\vec{q}_1} V(-\vec{q}\vec{q}'\vec{q}_1 - \vec{q}_2)(2n_{\vec{q}_1} + 1)$$
$$- \frac{18}{\hbar^2} \sum_{\vec{q}_1,\vec{q}_2} V(-\vec{q}\vec{q}_1\vec{q}_2)V(\vec{q}' - \vec{q}_1\vec{q}_2)\left(\frac{n_{\vec{q}_1} + n_{\vec{q}_2} + 1}{(\omega + \Omega_{\vec{q}_1} + \Omega_{\vec{q}_2})_P} + \frac{n_{\vec{q}_1} + n_{\vec{q}_2} + 1}{(\Omega_{\vec{q}_1} + \Omega_{\vec{q}_2} - \omega)_P} + \frac{n_{\vec{q}_2} - n_{\vec{q}_1}}{(\Omega_{\vec{q}_1} - \Omega_{\vec{q}_2} - \omega)_P}\right.$$
$$\left. + \frac{n_{\vec{q}_1} - n_{\vec{q}_2}}{(\Omega_{\vec{q}_1} - \Omega_{\vec{q}_2} - \omega)_P}\right),$$ (3.10)

and

$$\Gamma(\vec{q}\vec{q}'\omega) = \frac{18\pi}{\hbar} \sum_{\vec{q}_1\vec{q}_2} V(-\vec{q}\vec{q}_1\vec{q}_2)V(\vec{q}' - \vec{q}_1 - \vec{q}_2)\{-(n_{\vec{q}_1} + n_{\vec{q}_2} + 1)[\delta(\omega + \Omega_{\vec{q}_1} + \Omega_{\vec{q}_2}) - \delta(\omega - \Omega_{\vec{q}_1} - \Omega_{\vec{q}_2})]$$
$$- (n_{\vec{q}_1} - n_{\vec{q}_2})[\delta(\omega - \Omega_{\vec{q}_1} - \Omega_{\vec{q}_2}) - \delta(\omega + \Omega_{\vec{q}_1} - \Omega_{\vec{q}_2})]\}.$$ (3.11)

$n_{\vec{q}}$'s are Bose-Einstein distribution functions and P refers to principal-value summation. For an external probe of frequency Ω, the Δ and Γ can be calculated if the harmonic analogs of the system are known.[20] Once the self-energy of the anharmonic phonon is known the phonon Green's function corresponding to anharmonic crystal can be written as

$$D(\vec{q}, \omega) = \frac{2\Omega_{\vec{q}}}{\Omega_{\vec{q}}^2 - \omega^2 + 2\Omega_{\vec{q}}(\Delta - i\Gamma)},$$ (3.12)

and thus the spectral function is

$$A(\vec{q}, \omega) = \frac{1}{\pi} \frac{2\Omega_{\vec{q}}\Gamma}{(\Omega_{\vec{q}}^2 - \omega^2 + 2\Omega_{\vec{q}}\Delta)^2 + 4\Omega_{\vec{q}}^2\Gamma^2}.$$ (3.13)

The anharmonic phonon spectral function can now be used in Eqs. (2.4) and (2.5) in order to calculate the anharmonic phonon-electron coupling constant λ. Note that for small ω the $\Delta(q, \omega)$ is independent of ω whereas $\Gamma(q, \omega)$ is linear in ω.

IV. ANHARMONIC PHONON-ELECTRON COUPLING AND SUPERCONDUCTIVITY IN METAL HYDRIDE (DEUTERIDE) SYSTEMS

Since the spectral function for the anharmonic phonon spectrum is no longer a δ function the integrals involved in calculating the $\alpha^2 F(\omega)$ and the coupling parameter λ are nontrivial. However, one can make use of the following identity to carry out the integral:

$$\int_0^\infty \frac{d\omega}{\omega} A(q, \omega) = - \frac{D(q, \omega = 0)}{2} . \qquad (4.1)$$

Identity (4.1) is valid for any spectral function A. The static phonon Green's function for the anharmonic phonon is given by [cf. Eq. (3.12)]

$$\frac{D(q, \omega = 0)}{2} = \frac{\Omega_{\vec{q}}}{\Omega_{\vec{q}}^2 + 2\Omega_{\vec{q}} \Delta(\omega = 0)} \qquad (4.2)$$

The static anharmonic frequency Ω_A is defined by

$$\Omega_A^2 = \Omega_{\vec{q}}^2 + 2\Omega_{\vec{q}} \Delta(\omega = 0). \qquad (4.3)$$

If we assume that the electron-phonon coupling parameter g is inversely proportional to the static anharmonic phonon frequency and use the well-known sum rule

$$\int_0^\infty d\omega \, \omega A(\vec{q}, \omega) = \Omega_{\vec{q}}, \qquad (4.4)$$

then the first moment of $\alpha^2 F(\omega)$ is independent of the phonon parameter and only dependent on the electronic parameter P_e of the system:

$$\int_0^\infty d\omega \, \omega \alpha^2 F(\omega) = \frac{P_e}{2M} . \qquad (4.5)$$

This leads to the well-known[17] expression for the electron-phonon coupling parameter with the only difference that the harmonic phonon frequencies have to be replaced by *static* anharmonic frequencies Ω_A, i.e.,

$$\lambda = P_e / M \langle \Omega_A^2 \rangle . \qquad (4.6)$$

It is interesting to note here that, under the quasi-harmonic approximation, neither phonon lifetime effects nor the dynamical frequency shifts enter into the anharmonic phonon-electron coupling. *The effect of anharmonicity on λ is totally governed by the static shift $\Delta(\Omega = 0)$ of the phonon mode from its harmonic position.*

Let us now discuss superconductivity in the metal-hydride (deuteride) systems in light of the anharmonic phonon-electron coupling. It was pointed out early in this paper that the hydrogen vibrations in the palladium cage are much more anharmonic than that of deuterium. Let us assume also that the phonons in PdD are the harmonic analog of phonons in PdH except for the

mass factor of 2. Moreover, we will assume that the electronic parameters of PdH and PdD are not drastically different. The latter assumption is supported by the fact that the lattice constants in PdH and PdD are almost identical and the densities of states at the Fermi surface are not much different. Under these realistic assumptions one can write

$$\frac{\lambda_{PdD}}{\lambda_{PdH}} = \frac{M_H \langle \Omega_A^2 (PdH) \rangle}{M_D \langle \Omega_{\vec{q}}^2 (PdD) \rangle} ; \qquad (4.7)$$

where Ω_A(PdH) is the static anharmonic phonon frequencies in PdH, and Ω_q(PdD) the harmonic phonon frequencies in PdD. Now Ω_A^2(PdH) can be written in terms of harmonic frequencies of PdD and the static frequency shift $\Delta(\Omega = 0)$ as

$$\Omega_A^2 (PdH) = 2\Omega_{\vec{q}}^2 (PdD) + 2\sqrt{2} \, \Omega_{\vec{q}}(PdD) \Delta(\omega = 0). \qquad (4.8)$$

Substituting (4.8) in (4.7) and decoupling the averaging scheme for two terms in Eq. (4.8) one gets

$$\lambda_{PdD} \simeq [1 + \sqrt{2} \, \langle \Delta(\omega = 0) / \Omega_{\vec{q}}(PdD) \rangle] \lambda_{PdH}. \qquad (4.9)$$

Equation (4.9) indicates a reduction in the electron-phonon coupling due to anharmonicity if the static anharmonic phonon frequency shift is positive, i.e., $\Delta(\Omega = 0) > 0$. This will imply a stiffening of the force constant by anharmonicity. As mentioned earlier the tunneling data and the neutron data do indicate a stiffening of the force constant between Pd-H compared to that of Pd-D. Equation (4.9) in conjunction with available neutron data and Born–von Kármán analysis provides us with a prescription for calculating the change in λ from the PdD to the PdH system. A numerical calculation of that sort will be attempted in the future.

In conclusion, we have attempted to demonstrate the effect of anharmonicity in the theory of superconductivity in metal-hydride (deuteride) systems using existing ideas on anharmonic phonons[20] and a general spectral-function representation.[18] We have indicated a simple procedure for calculating the change in the electron-phonon coupling parameter λ due to anharmonicity using available neutron data and their analysis. The theory outlined above clearly shows that for a positive static shift of the harmonic phonon modes, the anharmonicity will lower the magnitude of the coupling parameter. This result is in qualitative agreement with the opposite isotope effect in PdH(D) systems, and the tunneling and the neutron scattering data indicating a stiffening of the force constant in PdH compared to that of PdD. However, a quantitative calculation of the change in λ and thus the superconducting T_c for PdH(D) systems will be the real test of this theory. Finally, there are a few assumptions in this theory, viz., the assumptions that the

electronic parameters are equivalent in PdH(D) systems, that the PdD phonons are almost harmonic, that the anharmonicity is not so important for Pd vibrations, and that there is no optic-phonon-induced anharmonicity in the acoustical phonons. In view of experimental information on lattice constants, densities of states at the Fermi surface, changes in the acoustical-phonon spectra from neutron data on PdH(D) systems

we feel that the assumptions are realistic and will not change the physics of the situation in a qualitative way.

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