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A Nonlinear Theory of Electron-Phonon Interaction in Semiconductors

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Starting from a usual hamiltonian in the electron-phonon system, a canonical transformation is carried out up to the third order of the coupling constant. The three-phonon process is the lowest order one which remains after taking the expectation value over electron distribution. The effective third-order elastic constant for long wavelength phonons at high temperatures is found to be identical with that suggested by Hutson and derived from a semiclassical treatment. The other three-phonon processes arising from the strain dependence of the coupling constant are of less importance than the above-mentioned process.

§1. Introduction

A few years ago, Hutson¹⁾ suggested that a three-phonon process via a nonlinear interaction of acoustic waves with conduction electrons might be responsible for the establishment of steady states of amplified waves in piezoelectric semiconductors in a strong electric field. Later experiments of ultrasonic harmonic generation $^{2\sim 5}$ in CdS have shown that the new process at ultrasonic frequencies predominates over the usual process due to the anharmonic interaction The phenomenological in lattice vibrations. treatments^{2~5)} are limited to the case of ql < 1(q: wave number of acoustic waves, l: mean free path of electrons), so that one could not know all about the new process. We have therefore developed in a previous paper⁶) a formalism based on the semiclassical Boltzmann equation treatment which can be applied to all values of ql. It has been shown that the new process, irrespective of the applied electric field when ql>1, predominates over the usual process in a restricted range of wave number in both piezoelectric and covalent semiconductors.

In this paper we shall develop a quantum theory of electron-phonon interaction by using the method of canonical transformation in order to lay the foundation for the semiclassical treatment which is still phenomenological. We shall also discuss the other three-phonon processes arising from the strain dependence of the coupling constant of electron-phonon interaction.

§2. Canonical Transformation

We start from a usual hamiltonian,

$$H' = H_e' + H_p' + H_i'$$
, (2.1 a)

$$H_{e}' = \sum_{k,s} E_{k} A_{k,s}^{+} A_{k,s}$$
, (2.1 b)

$$H_p' = \sum_q \hbar \Omega_q B_q^+ B_q , \qquad (2.1 \text{ c})$$

$$H_i' = \sum_{k,s,q} U(q) B_q A_{k+q,s}^+ A_{k,s} + c.c.$$
, (2.1 d)

where E_k is the energy of an electron with the wave vector k, $\hbar\Omega_q$ is the energy of a phonon with q, A^+ , A and B^+ , B are the creation and annihiration operators for electrons and phonons, respectively and s is a spin index. For simplicity, the polarization index of phonons has been omitted in (2.1 c). The coupling constant U(q)is given by

$$U(q) = i(C/\varepsilon(q))(\hbar q/2 \rho v_s)^{1/2}, \qquad (2.2 a)$$

$$= (4 \pi \beta e/\varepsilon \varepsilon(q))(\hbar/2 \rho v_s q)^{1/2} , \qquad (2.2 b)$$

for the deformation-potential and piezoelectric coupling, respectively. Here C is the deformation-potential constant, β the piezoelectric constant, ρ the density, e the electronic charge, v_s the velocity of sound, and ε is the static dielectric constant. The Coulomb interaction between electrons has been taken into account by introducing the wave-number dependent dielectric constant, $\varepsilon(q)$. In the Debye approximation we have

$$\varepsilon(q) = 1 + q_0^2/q^2$$
, $q_0^2 = 3 \omega_p^2/v_0^2$, (2.3)

where ω_p is the plasma frequency and v_0 is the root-mean-square velocity of electrons.

We shall now eliminate H_i' by a canonical transformation from operators (A, B) to a new set of operators (a, \tilde{b}) and calculate a new hamiltonian up to the third order of the coupling constant. The new hamiltonian is expressed by

$$\mathscr{H} = e^{iS} H e^{-iS} , \qquad (2.4)$$

where H represents the hamiltonian which is the same function of the new operators as H' is of the old. Besides the transformation (2.4), the Bogoliubov transformation on phonon operators,

$$\tilde{b}_q = u_q b_q + v_q^* b_{-q}^+$$
, $|u_q|^2 - |v_q|^2 = 1$, (2.5)

is carried out in order to eliminate terms of pair creation and annihiration of phonons with opposite momenta. We then rewrite H in the form,

$$H = H_0 + H_1 + H_2$$
, (2.6 a)

$$H_0 = \sum_{k,s} \varepsilon_k a^+_{k,s} a_{k,s} + \sum_q \hbar \omega_q b_q^+ b_q , \quad (2.6 \text{ b})$$
$$H_1 = \sum V(q) b_q a^+_{k+q,s} a_{k,s} + c.c. ,$$

$$V(q) = U(q)u_q + U^*(q)v_q$$
, (2.6 c)

$$H_{2} = \sum_{k,s} (E_{k} - \varepsilon_{k}) a_{k,s}^{+} a_{k,s} - \sum_{q} \hbar \{ \omega_{q} - \Omega_{q} (|u_{q}|^{2} + |v_{q}|^{2}) \} b_{q}^{+} b_{q} + \sum_{q} \hbar \Omega_{q} (u_{q} v_{q} b_{-q} b_{q} + c.c.) ,$$
(2.6 d)

where ε_k , $\hbar \omega_q$ and V(q) are the renormalized c electron energy, phonon energy and coupling constant, respectively.

If we write the generating function S in (2.4) as

$$S = S_1 + S_2$$
, (2.7)

then we have the new hamiltonian,

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_1 + \mathcal{H}_2 + \mathcal{H}_3, \quad (2.8 a)$$

$$\mathscr{H}_0 = H_0$$
, (2.8 b)

$$\mathscr{H}_1 = H_1 + i[S_1, H_0],$$
 (2.8 c)

$$\mathscr{H}_{2} = H_{2} + i[S_{1}, H_{1}] + i[S_{2}, H_{0}] - \frac{1}{2}[S_{1}, [S_{1}, H_{0}]],$$
(2.8 d)

$$\mathcal{H}_{3} = i[S_{2}, H_{1}] + i[S_{1}, H_{2}] - \frac{1}{2}[S_{1}, [S_{2}, H_{0}]]$$

$$- \frac{1}{2}[S_{2}, [S_{1}, H_{0}]] - \frac{1}{2}[S_{1}, [S_{1}, H_{1}]]$$

$$- (i/6)[S_{1}, [S_{1}, [S_{1}, H_{0}]]] . \qquad (2.8 \text{ e})$$

Let us determine unknown quantities S_1 , S_2 , ε_k , $\hbar \omega_q$, u_q and v_q by using the standard procedure employed in the theory of electron-phonon interaction in normal metals⁷ and superconductors.⁸ First, S_1 is determined by $\mathscr{H}_1=0$. Then, ε_k and $\hbar \omega_q$ are chosen so as to cancel diagonal terms of $a_{k,s}^+ a_{k,s}$ and $b_q^+ b_q$ in \mathscr{H}_2 in the Hartree approximation. Similarly, u_q and v_q can be determined so as to eliminate terms of $b_{-q}b_q$ and $b_q^+ b_{-q}^+$ in \mathscr{H}_2 . Assuming that V(q) is real or pure imaginary and u_q and v_q are real, we obtain $\varepsilon_k = E_k + \sum_q |V(q)|^2 \{(n_q+1-f_{k-q})(\varepsilon_k - \varepsilon_{k-q} - \hbar \omega_q)^{-1}\}$

+
$$(n_q+f_{k+q})(\varepsilon_k-\varepsilon_{k+q}+\hbar\omega_q)^{-1}\}$$
, (2.9 a)

$$\omega_q^2 = \Omega_q^2 + (2 \omega_q/\hbar) |V(q)|^2 \times \sum_{k,s} (f_k - f_{k+q}) (\varepsilon_k - \varepsilon_{k+q} + \hbar \omega_q)^{-1} , \qquad (2.9 \text{ b})$$

$$|V(q)|^2 = |U(q)|^2 \Omega_q / \omega_q$$
, (2.9 c)

where $f_k = \langle a_{k,s}^* a_{k,s} \rangle$ and $n_q = \langle b_q^+ b_q \rangle$ are the distribution functions of electron and phonon, respectively. Finally, S_2 can be determined by $\mathcal{H}_2 = 0$. It should be noted here that the parts of \mathcal{H}_1 and \mathcal{H}_2 which are diagonal in energy and hence responsible for the real processes cannot be eliminated by the canonical transformation. Thus, the explicit form of the new hamiltonian is given by

$$\mathscr{H} = \mathscr{H}_0 + \mathscr{H}_1^r + \mathscr{H}_2^r + \mathscr{H}_3$$
, (2.10 a)

$$\mathscr{H}_{0} = \sum_{k,s} \varepsilon_{k} a^{+}_{k,s} a_{k,s} + \sum_{q} \hbar \omega_{q} b_{q}^{+} b_{q} , \quad (2.10 \text{ b})$$

$$\mathscr{H}_{1}^{r} = \sum_{k,s,q} V(q) b_{q} a_{k+q,s}^{+} a_{k,s} + c.c.$$
, (2.10 c)

$$\begin{aligned} \mathscr{H}_{2}^{r} &= \frac{1}{2} \sum_{k,s,q,p} \left\{ (\varepsilon_{k} - \varepsilon_{k+q} + \hbar \omega_{q})^{-1} \\ &- (\varepsilon_{k+p} - \varepsilon_{k+p+q} + \hbar \omega_{q})^{-1} \right\} \\ &\times \left[\left\{ V(q)V(p)b_{p}b_{q}a_{k+p+q,s}^{+}a_{k,s} + c.c \right\} \\ &+ \left\{ V(q)V^{*}(p)b_{-p}^{+}b_{q}a_{k+p+q,s}^{+}a_{k,s} + c.c. \right\} \right] \\ &+ \frac{1}{2} \sum_{k,s,\lambda,\sigma,q} |V(q)|^{2} (\varepsilon_{k+q} - \varepsilon_{k} - \hbar \omega_{q})^{-1} \\ &\times (a_{k+q,s}^{+}a_{k,s}a_{\lambda-q,\sigma}^{+}a_{\lambda,\sigma} + c.c.) , \end{aligned}$$
(2.10 d)

$$\begin{aligned} \mathscr{H}_{3} &= \frac{1}{4} \sum_{k,s,p,q,r} \\ &\times \left[P\{V(p)V(q)V(r)b_{r}b_{p}b_{q}a_{k+p+q+r,s}^{+}a_{k,s} + c.c. \} \\ &+ Q\{V(p)V(q)V^{*}(r)b_{-r}^{+}b_{p}b_{q}a_{k+p+q+r,s}^{+}a_{k,s} + c.c. \} \\ &+ R\{V^{*}(p)V(q)V(r)b_{r}b_{-p}^{+}b_{q}a_{k+p+q+r,s}^{+}a_{k,s} + c.c. \} \\ &+ J\{V^{*}(p)V(q)V^{*}(r) \\ &\times b_{-r}^{+}b_{-p}^{+}b_{q}a_{k+p+q+r,s}^{+}a_{k,s} + c.c. \} \right], \quad (2.10 e) \end{aligned}$$

$$P \equiv B \{ (\varepsilon_{k+p+q} - \varepsilon_k - \hbar \omega_p - \hbar \omega_q)^{-1} \\ + (1/3) (\varepsilon_{k+p+q} - \varepsilon_{k+p+q+r} + \hbar \omega_r)^{-1} \} \\ + D \{ (\varepsilon_{k+p+q+r} - \varepsilon_{k+r} - \hbar \omega_p - \hbar \omega_q)^{-1} \\ + (1/3) (\varepsilon_k - \varepsilon_{k+r} + \hbar \omega_r)^{-1} \} ,$$

$$Q \equiv B\{(\varepsilon_{k+p+q} - \varepsilon_k - \hbar\omega_p - \hbar\omega_q)^{-1} + (1/3)(\varepsilon_{k+p+q} - \varepsilon_{k+p+q+r} - \hbar\omega_r)^{-1}\} + D\{(\varepsilon_{k+p+q+r} - \varepsilon_{k+r} - \hbar\omega_p - \hbar\omega_q)^{-1} + (1/3)(\varepsilon_k - \varepsilon_{k+r} - \hbar\omega_r)^{-1}\},$$

$$\begin{split} R &\equiv B \{ (\varepsilon_{k+p+q} - \varepsilon_k + \hbar \omega_p - \hbar \omega_q)^{-1} \\ &+ (1/3) (\varepsilon_{k+p+q} - \varepsilon_{k+p+q+r} + \hbar \omega_r)^{-1} \} \\ &+ D \{ (\varepsilon_{k+p+q+r} - \varepsilon_{k+r} + \hbar \omega_p - \hbar \omega_q)^{-1} \\ &+ (1/3) (\varepsilon_k - \varepsilon_{k+r} + \hbar \omega_r)^{-1} \} \end{split}$$

$$\begin{split} J &= B\{(\varepsilon_{k+p+q} - \varepsilon_k + \hbar \omega_p - \hbar \omega_q)^{-1} \\ &+ (1/3)(\varepsilon_{k+p+q} - \varepsilon_{k+p+q+r} - \hbar \omega_r)^{-1}\} \\ &+ D\{(\varepsilon_{k+p+q+r} - \varepsilon_{k+r} + \hbar \omega_p - \hbar \omega_q)^{-1} \\ &+ (1/3)(\varepsilon_k - \varepsilon_{k+r} + \hbar \omega_r)^{-1}\} , \end{split}$$

$$B &\equiv (\varepsilon_{k+p} - \varepsilon_{k+p+q} + \hbar \omega_q)^{-1} - (\varepsilon_k - \varepsilon_{k+q} + \hbar \omega_q)^{-1} ,$$

$$D &\equiv (\varepsilon_{k+r} - \varepsilon_{k+q+r} + \hbar \omega_q)^{-1} \\ &- (\varepsilon_{k+p+r} - \varepsilon_{k+p+q+r} + \hbar \omega_q)^{-1} , \end{split}$$

where we have neglected terms of the type of $b_q a_{k+p+q,s}^+ a_{k,s} a_{\lambda-q,\sigma}^+ a_{\lambda,\sigma}$ in (2.10 e).

§ 3. Three-Phonon Process

We shall only deal with \mathcal{H}_3 . As the diagonal terms of $a_{k,s}^+a_{k,s}$ in \mathcal{H}_2^r have been eliminated by the Bogoliubov transformation, the threephonon process is the lowest order one which remains after taking the expectation value over electron distribution. In order to compare the present theory with the semiclassical treatment, we choose the simplest terms proportional to $b_{2q}^+b_qb_{q}a_{k,s}^+a_{k,s}$ and replace $a_{k,s}^+a_{k,s}$ by its expectation value f_k :

$$f_{k} = (N/N_{c}) \exp(-\varepsilon_{k}/k_{B}T) ,$$

$$N_{c} = 2(2 \pi m k_{B}T/h^{2})^{3/2} , \quad \varepsilon_{k} = \hbar^{2}k^{2}/2 m , \quad (3.1)$$

where m is the effective mass of electrons and N is the electron concentration. If we make the approximations,

$$k \gg q$$
, $k \gg 2 m v_s / \hbar$, (3.2)

for long wavelength phonons at high temperatures, we obtain

$$\mathscr{H}_{3}(q+q\to 2q) = (N/2k_{B}^{2}T^{2})V^{*}(2q)V^{2}(q)b_{2q}^{+}b_{q}b_{q} .$$
(3.3)

On the other hand, the hamiltonian for the same process as (3.3) due to the anharmonic interaction in lattice vibrations⁹⁾ is given by

$$\mathscr{H}(q+q\to 2\,q) = \frac{1}{2}(\hbar/2\,\rho)^{3/2}(2\,q^3A/2^{1/2}\omega q^{3/2})b_{2q}^+b_qb_q$$
(3.4)

in terms of the third-order elastic constant A. By comparing (3.3) with (3.4) we get an effective third-order elastic constant,

$$A_{\rm eff} = (N/2^{1/2} k_B^2 T^2) (2 \rho v_s / \hbar q)^{3/2} V^* (2 q) V^2(q) .$$
(3.5)

This is identical with that derived from the semiclassical theory in the case of ql > 1 and has been discussed in detail in the previous paper.⁶ So far we have not taken care of the selection rule due to the dispersion of phonons. However,

it is expected that eq. (3.3) can be applied to the case where all three phonons belong to the same polarization branch as long as the lifetime of phonons is sufficiently short at high temperatures.

§4. Other Three-Phonon Processes

We have neglected so far the strain dependence of the coupling constant which may lead to the two-phonon and three-phonon processes. We shall consider for simplicity the case of deformation-potential coupling for longitudinal phonons, neglecting the screening effect. If we write

$$C = C_1 + C_2 \varDelta + C_3 \varDelta^2 , \qquad (4.1)$$

 \varDelta being a dilation, then we have extra hamiltonians,

$$\begin{split} H_{i2} &= -\sum_{k,s,p,q} V_2(p,q)(b_q - b_{-q}^+) \\ &\times (b_p - b_{-p}^+) a_{k+p+q,s}^+ a_{k,s} . \\ V_2(p,q) &= C_2(\hbar/2 \ \rho)(\omega_q \omega_p)^{-1/2} p q \ , \qquad (4.2) \\ H_{i3} &= -i \sum_{k,s,p,q,r} V_3(p,q,r)(b_q - b_{-q}^+)(b_p - b_{-p}^+) \\ &\times (b_r - b_{-r}^+) a_{k+p+q+r,s}^+ a_{k,s} \ , \\ V_3(p,q,r) &= C_3(\hbar/2 \ \rho)^{3/2} (\omega_q \omega_p \omega_r)^{-1/2} p q r \ . \qquad (4.3) \end{split}$$

Thus, we have the other two three-phonon processes arising from the second order perturbation of H_1 in (2.6 c) and H_{i_2} , and from H_{i_3} . The hamiltonian for the former process is given by

$$\mathscr{H}_{3}(\boldsymbol{q}+\boldsymbol{q}\rightarrow 2\boldsymbol{q})=b_{2q}^{+}b_{q}b_{q}\sum_{\boldsymbol{k},\boldsymbol{s}}F(\boldsymbol{k},\boldsymbol{q})$$

$$\begin{split} F(k, q) \\ = V_1^*(2q) V_2(q, q) \{ f_k(1 - f_{k-2q}) / (\varepsilon_k - \varepsilon_{k-2q} - 2\hbar\omega_q) \\ + f_k(1 - f_{k+2q}) / (\varepsilon_k - \varepsilon_{k+2q} + 2\hbar\omega_q) \} \\ - 2 V_1(q) V_2(q, 2q) \{ f_k(1 - f_{k-q}) / (\varepsilon_k - \varepsilon_{k-q} - \hbar\omega_q) \\ + f_k(1 - f_{k+q}) / (\varepsilon_k - \varepsilon_{k+q} + \hbar\omega_q) \} , \end{split}$$

$$(4.4)$$

where we have used notations $V_1(q)$ and C_1 in place of V(q) and C in H_1 . Using the approximations, (3.2) and $f_k \ll 1$, we obtain

$$A'_{\rm eff} = -6 \, i C_1 C_2 N / k_B T$$
 (4.5)

for this process. On the other hand, the effective third-order elastic constant due to H_{i3} is simply given by

$$A_{\rm eff}^{\prime\prime} = 6 \, i C_3 N$$
 (4.6)

These constants, A'_{eff} and A''_{eff} , should be compared with

$$A_{\rm eff} = i C_1^{3} N / (k_B T)^2 \tag{4.7}$$

derived from (3.5). Therefore, if we assume $C_2 \sim C_3 \sim C_1$, then we have an inequality,

$$A_{\rm eff} \gg A_{\rm eff}^{\prime} \gg A_{\rm eff}^{\prime\prime} , \qquad (4.8)$$

because C_1/k_BT is much larger than 1 under ordinary conditions. It is expected that the strain dependence of the coupling constant does not contribute seriously to the three-phonon process even in piezoelectric semiconductors.

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References

- 1) A. R. Hutson: Phys. Rev. Letters 9 (1962) 296.
- 2) T. Ishiguro *et al.*: 1964 IEEE International Convention.
- 3) H. Kroger: Appl. Phys. Letters 4 (1964) 190.
- 4) C. Elbaum and R. Truell: Appl. Phys. Letters 4 (1964) 212.
- 5) B. Tell: Phys. Rev. 136 (1964) A772.
- N. Mikoshiba: J. Phys. Soc. Japan 20 (1965) 2160.
- J. Bardeen and D. Pines: Phys. Rev. 99 (1955) 1140.
- N. N. Bogoliubov et al.: Fortschr. Phys. 6 (1958) 605.
- 9) J. M. Ziman: *Electrons and Phonons* (Clarendon Press, Oxford, 1960).