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Electron–Phonon Interaction in Dielectric Bilayer Systems

Effect of the Electronic Polarizability

By

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A quantum-mechanical theory of the electron–(long-wave optical) phonon interaction in dielectric bilayer systems is developed. The operator describing the interaction between an electron and the phonon modes of the bilayer system is calculated including the electronic polarizability. All polarization eigenmodes together with their dispersion relation are derived and discussed. The dispersion curves of the surface phonons are calculated and presented for various cases in graphical form. The interaction of electrons outside and inside the bilayer system is studied and the coupling functions are calculated and discussed and they are shown in graphical form.

Es wird eine quantenmechanische Theorie der Elektron–(langwellig-optischen) Phonon-Wechselwirkung in dielektrischen Doppelschichtsystemen entwickelt. Dabei wird der Einfluß der elektronischen Polarisierbarkeit auf die Struktur des Hamilton-Operators der Wechselwirkung der Elektronen mit den Phononen berücksichtigt. Es werden alle Polarisationseigenmoden und die dazu gehörenden Dispersionsrelationen abgeleitet und diskutiert. Die Dispersionskurven der Oberflächen-Phononen werden für verschiedene Fälle berechnet und graphisch dargestellt. Die Wechselwirkung von Elektronen, welche sich innerhalb und außerhalb des Doppelschichtsystems befinden, wird untersucht und die Kopplungsfunktionen werden für verschiedene Fälle berechnet und graphisch dargestellt.

1. Introduction

In the last few years the interest in the investigation of surface properties of condensed media and of thin layer properties has been highly increased. There are several reasons for it: (i) surfaces are always present in real samples and (ii) due to the very large scale integration in microelectronics surface properties are of great importance. Therefore, experimental and theoretical investigations in the field of surface physics are in direct relation to microelectronics, optoelectronics, and other modern applications.

Solid state surfaces as well as interfaces between two media are connected with a variety of interesting phenomena. On the one hand their existence results in completely novel effects and, on the other hand, the properties of the bulk are changed. In finite crystals the electrons can couple with *longitudinal phonons* and with *surface phonons*. The investigation of the latter interaction is easier when the effect of retardation connected with surface modes is neglected. The effect of electron–phonon interaction results from the sum over all wave vectors, while the effect of retardation on surface modes is limited to small frequencies (wave vector of the light in the FIR $|\boldsymbol{q}| \approx 10^5\ \mathrm{m}^{-1}$, dimension of the first Brillouin zone $|\boldsymbol{q}| \approx 10^{10}\ \mathrm{m}^{-1}$). Therefore, the approximation neglecting retardation effects yields good results. Is the electron (or an ion) out of the medium an interaction takes place only with the surface modes. This effect is important for various *surface spectroscopic experiments* as for instance

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EELS and XPS. In layered *semiconductor devices* the understanding of the interaction of conduction band electrons with long-wave optical phonons is of great importance. The new developments concerning *heterostructures* and *superlattices* on the basis of the polar $A^{III}B^{V}$ semiconductors increase the interest in the understanding of the electron–phonon interaction. The electron–phonon interaction plays an important role to understand the *quasi-two-dimensional electron system* which forms the space charge layer in such novel devices.

Firstly electron–phonon interaction in a dielectric layer was considered by Lucas et al. [1] followed by works of Wang and Mahan [2], Evans and Mills [3], and others. The electronic polarizability was taken into account firstly by Licari and Evrard [4], the electron–phonon interaction in a bilayer system without it was studied by Lenac and Šunjić [5]. In fact, this electronic polarizability plays an important role in the interaction operators and can give a large contribution to their magnitudes. For instance in an infinite extended dielectric medium the strength of electron–phonon interaction is proportional to $(1/\varepsilon_\infty - 1/\varepsilon_s)^{1/2}$. Neglecting the electronic polarizability ($\varepsilon_\infty = 1$) in a $A^{III}B^{V}$ semiconductor the strength of interaction is overestimated by a factor of eight.

In our work a bilayer system composed of two dielectric media (ionic crystals or polar semiconductors) is studied. The importance of bilayer systems is given by the fact that they are often practically realized. Moreover the bilayer system serves as a model for multilayer systems. For such a bilayer system we have developed a quantum-mechanical theory of electron–phonon interaction (in the long-wave approximation) taking into account the effect of electronic polarizability. The latter is nontrivial for a bilayer system because the electronic polarizability enters the interaction operators in a complicate manner. Moreover the orthonormalization relation of the surface polarization eigenmodes is changed.

2. Polarization Eigenmodes of the Bilayer System: Long-Wave Optical Phonons

2.1 Equation of motion and polarization eigenmodes

We consider bilayer systems (Fig. 1) composed of diatomic ionic crystals or polar semiconductors consisting of one pair of positive and negative ions per Wigner-Seitz cell.

Because our interest is directed to long-wave optical modes we use the continuum model. In this case we can write the displacement of the positive and negative ions as $\boldsymbol{u}^+(\boldsymbol{x}, t)$ and $\boldsymbol{u}^-(\boldsymbol{x}, t)$, respectively. The equation of motion for the displacement of the ions in the n-th layer ($n = 1, 2$) is given by

$$m_n^+\ddot{\boldsymbol{u}}^+(\boldsymbol{x}, t) = -f_n\big(\boldsymbol{u}^+(\boldsymbol{x}, t) - \boldsymbol{u}^-(\boldsymbol{x}, t)\big) + e_n^*\boldsymbol{E}^{+\mathrm{loc}}(\boldsymbol{x}, t)\,, \tag{1}$$

$$m_n^-\ddot{\boldsymbol{u}}^-(\boldsymbol{x}, t) = f_n\big(\boldsymbol{u}^+(\boldsymbol{x}, t) - \boldsymbol{u}^-(\boldsymbol{x}, t)\big) - e_n^*\boldsymbol{E}^{-\mathrm{loc}}(\boldsymbol{x}, t)\,, \tag{2}$$

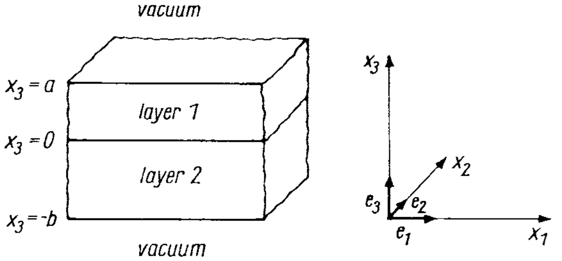


Fig. 1. Bilayer system geometry

where $\boldsymbol{u}^+$ and $\boldsymbol{u}^-$ are the displacement of the positive and negative ions, respectively, m_n^+ and m_n^- are the mass of the ions in the Wigner-Seitz cell of the n-th layer, e_n^* is the effective charge of the ions, f_n the short-range force constant (excluding the long-range Coulomb fields), and $\boldsymbol{E}^{+\text{loc}}$ and $\boldsymbol{E}^{-\text{loc}}$ are the local fields at the positions of the ions. Equation (2) yields two parts of interaction between the ions: a short-range part $(\sim f_n(\boldsymbol{u}^+ - \boldsymbol{u}^-))$ and a long-range part $(\sim e_n^*\boldsymbol{E}^{\text{loc}})$. For the short-range part only the harmonic interaction is taken into account. In each layer the short-range force constant is considered to be equal inside the layer and at the surfaces of the layer. Then the geometry of the problem is taken into account only by the long-range part of the interaction. This simplification leads to good results in the case of long-wave optical modes. With the reduced mass of the ion pair $\mu_n = m_n^+ m_n^-/(m_n^+ + m_n^-)$ and the relative diplacement $\boldsymbol{u}(\boldsymbol{x}, t) = \boldsymbol{u}^+(\boldsymbol{x}, t) - \boldsymbol{u}^-(\boldsymbol{x}, t)$ we can write (1), (2) in the form

$$\mu_n \ddot{\boldsymbol{u}}(\boldsymbol{x}, t) = -\mu_n \omega_{0n}^2 \boldsymbol{u}(\boldsymbol{x}, t) + e_n^* \boldsymbol{E}^{\text{loc}}(\boldsymbol{x}, t) , \tag{3}$$

where $\omega_{0n}^2 = f_n/\mu_n$ is the frequency associated with the short-range part of the interaction. The oscillating ions produce a polarization field $\boldsymbol{P}(\boldsymbol{x}, t)$ consisting of two parts: (i) the ionic polarization caused by the relative separation of the positive and negative ions when the crystal vibrates in an optical mode $(\sim \boldsymbol{u}(\boldsymbol{x}, t))$, and (ii) the electronic polarization caused by the local electric field associated with the optical modes acting on the electron shell of the ions $(\sim \boldsymbol{E}^{\text{loc}}(\boldsymbol{x}, t))$. $\boldsymbol{P}(\boldsymbol{x}, t)$ is given as the sum of both parts,

$$\boldsymbol{P}(\boldsymbol{x}, t) = n_n e_n^* \boldsymbol{u}(\boldsymbol{x}, t) + n_n \alpha_n \boldsymbol{E}^{\text{loc}}(\boldsymbol{x}, t) , \tag{4}$$

where n_n is the number of Wigner-Seitz cells per unit volume, and α_n is the electronic polarizability per Wigner-Seitz cell in the layer n. From the point of view of classical electrostatics (equivalent to the unretarded limit considered here) the Maxwell equations results in the Poisson equation for the scalar potential $\Phi(\boldsymbol{x}, t)$,

$$\Delta \Phi(\boldsymbol{x}, t) = -\frac{1}{\varepsilon_0} \varrho^{\text{total}}(\boldsymbol{x}, t) , \tag{5}$$

where ε_0 is the absolute dielectric constant and ϱ^{total} the total charge density. Since we assume the media to be nonconducting, nonmagnetic, and uncharged, ϱ^{total} is the sum of both bulk and surface polarization charge density.

Including both contributions (5) yields

$$\Phi(\boldsymbol{x}, t) = -\frac{1}{4\pi\varepsilon_0} \sum_\beta \int \mathrm{d}^3x' \frac{\partial}{\partial x_\beta} \frac{1}{|\boldsymbol{x} - \boldsymbol{x}'|} P_\beta(\boldsymbol{x}', t) . \tag{6}$$

Using

$$E_\alpha(\boldsymbol{x}, t) = -\frac{\partial}{\partial x_\alpha} \Phi(\boldsymbol{x}, t) \tag{7}$$

and the well-known Lorentz relation between the local field and the macroscopic electric field $\boldsymbol{E}$,

$$\boldsymbol{E}^{+\text{loc}} = \boldsymbol{E}^{-\text{loc}} = \boldsymbol{E} + \frac{1}{3\varepsilon_0} \boldsymbol{P} , \tag{8}$$

one obtains

$$E_\alpha^{\text{loc}}(\boldsymbol{x}, t) = \frac{1}{3\varepsilon_0} P_\alpha(\boldsymbol{x}, t) + \frac{1}{\varepsilon_0} \sum_\beta \int \mathrm{d}^3x' \, \Gamma_{\alpha\beta}(\boldsymbol{x} - \boldsymbol{x}') \, P_\beta(\boldsymbol{x}', t) . \tag{9}$$

Herein

$$\Gamma_{\alpha\beta}(\boldsymbol{x}-\boldsymbol{x}') = \frac{1}{4\pi}\frac{\partial^2}{\partial x_\alpha\,\partial x_\beta}\frac{1}{|\boldsymbol{x}-\boldsymbol{x}'|} \tag{10}$$

is the Green tensor. Then the equation of motion for the polarization follows from (3) and (4) with (9)

$$\begin{aligned}\ddot{P}_\alpha(\boldsymbol{x},t) &= \alpha_n n_n\left[\frac{1}{3\varepsilon_0}\ddot{P}_\alpha(\boldsymbol{x},t) + \frac{1}{\varepsilon_0}\sum_\beta\int \mathrm{d}^3x'\,\Gamma_{\alpha\beta}(\boldsymbol{x}-\boldsymbol{x}')\,\ddot{P}_\beta(\boldsymbol{x}',t)\right] - \\ &\quad - n_n e_n^* \omega_{0n}^2\left\{\frac{1}{n_n e_n^*}P_\alpha(\boldsymbol{x},t) - \frac{\alpha_n}{e_n^*}\left[\frac{1}{3\varepsilon_0}P_\alpha(\boldsymbol{x},t) + \right.\right. \\ &\quad \left.\left. + \frac{1}{\varepsilon_0}\sum_\beta\int \mathrm{d}^3x'\,\Gamma_{\alpha\beta}(\boldsymbol{x}-\boldsymbol{x}')\,P_\beta(\boldsymbol{x}',t)\right]\right\} + \\ &\quad + \frac{n_n e_n^{*2}}{\mu_n}\left[\frac{1}{3\varepsilon_0}P_\alpha(\boldsymbol{x},t) + \frac{1}{\varepsilon_0}\sum_\beta\int \mathrm{d}^3x'\,\Gamma_{\alpha\beta}(\boldsymbol{x}-\boldsymbol{x}')\,P_\beta(\boldsymbol{x}',t)\right].\end{aligned} \tag{11}$$

We assume a harmonic time dependence of the polarization

$$\boldsymbol{P}(\boldsymbol{x},t) = \boldsymbol{P}(\boldsymbol{x})\,\mathrm{e}^{-i\omega t}\,. \tag{12}$$

Equation (12) becomes then

$$\begin{aligned}&\frac{(\omega^2-\omega_{0n}^2)\left(1-\dfrac{1}{3\varepsilon_0}n_n\alpha_n\right) + \dfrac{1}{3\varepsilon_0}\dfrac{n_n e_n^*}{\mu_n}}{\dfrac{n_n e_n^*}{\mu_n} - n_n\alpha_n(\omega^2-\omega_{0n}^2)}\,P_\alpha(\boldsymbol{x}) = \\ &= -\frac{1}{\varepsilon_0}\sum_\beta\int \mathrm{d}^3x'\,\Gamma_{\alpha\beta}(\boldsymbol{x}-\boldsymbol{x}')\,P_\beta(\boldsymbol{x}')\,.\end{aligned} \tag{13}$$

With the abbreviations

$$\lambda_{0n} = \frac{\omega_{0n}^2}{\varepsilon_0\omega_{\mathrm{p}n}^2}, \qquad \lambda_n = \frac{\omega^2}{\varepsilon_0\omega_{\mathrm{p}n}^2}, \tag{14}$$

where

$$\omega_{\mathrm{p}n}^2 = \frac{n_n e_n^{*2}}{\varepsilon_0\mu_n} \tag{15}$$

is the ion plasma frequency, we get from (13)

$$\begin{aligned}&\frac{(\lambda_n-\lambda_{0n})\left(1-\dfrac{1}{3\varepsilon_0}n_n\alpha_n\right) + \dfrac{1}{3\varepsilon_0}}{1-n_n\alpha_n(\lambda_n-\lambda_{0n})}\,P_\alpha(\boldsymbol{x}) = \\ &= -\frac{1}{\varepsilon_0}\sum_\beta\int \mathrm{d}^3x'\,\Gamma_{\alpha\beta}(\boldsymbol{x}-\boldsymbol{x}')\,P_\beta(\boldsymbol{x}')\,.\end{aligned} \tag{16}$$

We are interested in wave propagation in the x_1–x_2 plane where $\boldsymbol{q}_{||}$ is a two-dimensional wave vector parallel to the surfaces of the bilayer system. The x_3-axis is chosen perpendicular to the interface at $x_3 = 0$ (see Fig. 1). This symmetry of the bilayer system will be used now. The translational invariance for continuous displacements in

$\boldsymbol{q}_{||}$-direction is exploited by introducing two-dimensional Fourier transforms,

$$\boldsymbol{P}(\boldsymbol{x}) = \left(\frac{1}{2\pi}\right)^2 \int_{-\infty}^{\infty} \mathrm{d}^2 q_{||}\, \mathrm{e}^{i\boldsymbol{q}_{||}\boldsymbol{x}_{||}} \boldsymbol{P}(\boldsymbol{q}_{||}, x_3)\,, \tag{17}$$

where $\boldsymbol{x}_{||}$ is the two-dimensional position vector in the x_1–x_2 plane. The two-dimensional Fourier transform

$$\frac{1}{4\pi\,|\boldsymbol{x} - \boldsymbol{x}'|} = \frac{1}{8\pi^2} \int_{-\infty}^{\infty} \mathrm{d}^2 q_{||}\, \mathrm{e}^{i\boldsymbol{q}_{||}(\boldsymbol{x}_{||} - \boldsymbol{x}'_{||})}\, \frac{\mathrm{e}^{-q_{||}|x_3 - x_3'|}}{q_{||}} \tag{18}$$

is derived from the three-dimensional one by a complex integration and $q_{||} = |\boldsymbol{q}_{||}|$ The two-dimensional wave vector $\boldsymbol{q}_{||}$ and the two-dimensional position vector $\boldsymbol{x}$ are defined as

$$\boldsymbol{x}_{||} = x_1 \boldsymbol{e}_1 + x_2 \boldsymbol{e}_2\,, \tag{19}$$

$$\boldsymbol{q}_{||} = q_1 \boldsymbol{e}_1 + q_2 \boldsymbol{e}_2\,, \tag{20}$$

where $\boldsymbol{e}_1$, $\boldsymbol{e}_2$, $\boldsymbol{e}_3$ are Cartesian unit vectors directed along the x_1, x_2, x_3 axis, respectively. Since $\boldsymbol{P}(\boldsymbol{x})$ is real its Fourier transform must satify the following condition:

$$\boldsymbol{P}(\boldsymbol{q}_{||}, x_3) = \boldsymbol{P}^*(-\boldsymbol{q}_{||}, x_3)\,. \tag{21}$$

To write the integral equation (16) in a more transparent form we use (17) and (18). Then the integral equation changes to

$$\sum_\beta \gamma_{\alpha\beta} P_\beta(\boldsymbol{q}_{||}, x_3) = \sum_\beta \int \mathrm{d}x_3'\, M_{\alpha\beta}(x_3 - x_3')\, P_\beta(\boldsymbol{q}_{||}, x_3') \tag{22}$$

with

$$\gamma_{\alpha\beta} = -\frac{(\lambda_n - \lambda_{0n})\,(\delta_{\alpha\beta} - \Lambda_{\alpha\beta} n_n \alpha_n) + \Lambda_{\alpha\beta}}{1 - n_n \alpha_n (\lambda_n - \lambda_{0n})}\,, \tag{23}$$

$$\Lambda_{\alpha\beta} = \begin{pmatrix} \frac{1}{3\varepsilon_0} & 0 & 0 \\ 0 & \frac{1}{3\varepsilon_0} & 0 \\ 0 & 0 & -\frac{2}{3\varepsilon_0} \end{pmatrix}, \tag{24}$$

$$M_{\alpha\beta}(x_3 - x_3') =$$

$$= \frac{1}{2\varepsilon_0} \begin{pmatrix} -q_1^2 & -q_1 q_2 & -i q_1 q_{||} \operatorname{sgn}(x_3 - x_3') \\ -q_1 q_2 & -q_2^2 & -i q_2 q_{||} \operatorname{sgn}(x_3 - x_3') \\ -i q_1 q_{||} \operatorname{sgn}(x_3 - x_3') & -i q_2 q_{||} \operatorname{sgn}(x_3 - x_3') & q_{||}^2 \end{pmatrix} \frac{\mathrm{e}^{-q_{||}|x_3 - x_3'|}}{q_{||}}, \tag{25}$$

and

$$\operatorname{sgn}(x_3 - x_3') = \begin{cases} 1; & x_3 > x_3', \\ -1; & x_3 < x_3'. \end{cases}$$

Equation (22) with $\gamma_{\alpha\beta}$ given by (23) is the *microscopic form* of the integral equation for the polarization. The integral equation (22) can be also obtained in a *macroscopic form*. We start with (6) and replace the electric field strength by the polarization according to the linear macroscopic equation

$$\boldsymbol{P}(\boldsymbol{q}_{||}, x_3) = \varepsilon_0 \chi_n(\omega) \, \boldsymbol{E}(\boldsymbol{q}_{||}, x_3), \tag{26}$$

where $\chi_n(\omega) = \varepsilon_n(\omega) - 1$ is the isotropic dielectric susceptibility. Using the same procedure as described above we find (22) with

$$\gamma_{\alpha\beta} = \frac{1}{\varepsilon_0} \begin{pmatrix} \chi_n^{-1}(\omega) & 0 & 0 \\ 0 & \chi_n^{-1}(\omega) & 0 \\ 0 & 0 & \chi_n^{-1}(\omega) \, \varepsilon_n(\omega) \end{pmatrix}. \tag{27}$$

The integral equation (22) defines the eigenvalues $\omega_i(\boldsymbol{q}_{||})$ corresponding to the polarization eigenmodes $\boldsymbol{P}^i(\boldsymbol{q}_{||}, x_3)$. Both forms of the integral equation (22), the microscopic (23), (24) and the macroscopic one (27), will be used in the following. The orthogonality relation

$$\sum_\alpha \int_{-b}^{a} \mathrm{d}x_3 \, [\chi_n^{-1}(\omega_i) - \chi_n^{-1}(\omega_j)] \, P_\alpha^{j*}(\boldsymbol{q}_{||}, x_3) \, P_\alpha^i(\boldsymbol{q}_{||}, x_3) = 0 \tag{28}$$

can be derived from (22) taking into account that $\boldsymbol{M}(x_3 - x_3') = \boldsymbol{M}^+(x_3' - x_3)$. The eigenvectors of (22) will then be orthonormalized according to

$$\sum_\alpha \int_{-b}^{a} \mathrm{d}x_3 \, \frac{\theta_n^{1/2}(\omega_i) \, \theta_n^{1/2}(\omega_j)}{\omega_{\mathrm{p}n}^2} \, P_\alpha^{j*}(\boldsymbol{q}_{||}, x_3) \, P_\alpha^i(\boldsymbol{q}_{||}, x_3) = \delta_{ij}, \tag{29}$$

with

$$\theta_n^{1/2} = \frac{1}{1 + n_n \alpha_n (\lambda_{0n} - \lambda_{in})}. \tag{30}$$

The orthonormalization relation given in (29) differs from that occurring in the case of a single free-standing layer by a weighting factor [4]. We define two new orthogonal unit vectors in the x_1–x_2 plane,

$$\boldsymbol{e}_{q_{||}} = \frac{\boldsymbol{q}_{||}}{q_{||}} = \frac{1}{q_{||}} [q_1 \boldsymbol{e}_1 + q_2 \boldsymbol{e}_2] \tag{31}$$

and

$$\boldsymbol{e}_\mathrm{s} = \frac{1}{q_{||}} [-q_2 \boldsymbol{e}_1 + q_1 \boldsymbol{e}_2]. \tag{32}$$

Using these unit vectors we can transform the polarization into the new system $\{\boldsymbol{e}_{q_{||}}, \boldsymbol{e}_\mathrm{s}, \boldsymbol{e}_3\}$,

$$\boldsymbol{P}(\boldsymbol{q}_{||}, x_3) = P_{q_{||}}(\boldsymbol{q}_{||}, x_3) \, \boldsymbol{e}_{q_{||}} + P_\mathrm{s}(\boldsymbol{q}_{||}, x_3) \, \boldsymbol{e}_\mathrm{s} + P_3(\boldsymbol{q}_{||}, x_3) \, \boldsymbol{e}_3 \tag{33}$$

with

$$P_{q_{||}}(\boldsymbol{q}_{||}, x_3) = \frac{1}{q_{||}} \big(q_1 P_1(\boldsymbol{q}_{||}, x_3) + q_2 P_2(\boldsymbol{q}_{||}, x_3)\big) \tag{34}$$

and

$$P_s(\boldsymbol{q}_{||}, x_3) = \frac{1}{q_{||}} \left(q_1 P_2(\boldsymbol{q}_{||}, x_3) - q_2 P_1(\boldsymbol{q}_{||}, x_3)\right) . \tag{35}$$

Realizing this transformation of coordinates in the integral equation (22) it is easy to separate the polarization field into an s-polarized part $(\boldsymbol{P} = (0, P_s, 0))$ and a p-polarized one $(\boldsymbol{P} = (P_{q_{||}}, 0, P_3))$. Then the analytical relations for both parts of polarization are completely decoupled. Because we are interested in the polarization eigenmodes, the macroscopic form of the integral equation is used. One obtains for s-polarization

$$\chi_n^{-1}(\omega)\, P_s(\boldsymbol{q}_{||}, x_3) = 0 . \tag{36}$$

The other two components (p-polarization) form a coupled system of Fredholm integral equations,

$$\chi_n^{-1}(\omega)\, P_{q_{||}}(\boldsymbol{q}_{||}, x_3) = \tfrac{1}{2} \int \mathrm{d}x_3'\, \mathrm{e}^{-q_{||}|x_3 - x_3'|} \left(-q_{||} P_{q_{||}}(\boldsymbol{q}_{||}, x_3') - \right.$$
$$\left. - iq_{||} \operatorname{sgn}(x_3 - x_3')\, P_3(\boldsymbol{q}_{||}, x_3')\right) , \tag{37}$$

$$\chi_n^{-1}(\omega)\, \varepsilon_n(\omega)\, P_3(\boldsymbol{q}_{||}, x_3) = \tfrac{1}{2} \int \mathrm{d}x_3'\, \mathrm{e}^{-q_{||}|x_3 - x_3'|} \times$$
$$\times \left(-iq_{||} \operatorname{sgn}(x_3 - x_3')\, P_{q_{||}}(\boldsymbol{q}_{||}, x_3') + q_{||} P_3(\boldsymbol{q}_{||}, x_3')\right) . \tag{38}$$

The usual way to solve this system of equations is to transform it into differential equations [6]. These are

$$\chi_n^{-1}(\omega) \frac{\mathrm{d}}{\mathrm{d}x_3} P_{q_{||}}(\boldsymbol{q}_{||}, x_3) = iq_{||} \chi_n^{-1}(\omega)\, P_3(\boldsymbol{q}_{||}, x_3) , \tag{39}$$

$$\chi_n^{-1}(\omega)\, \varepsilon_n(\omega) \frac{\mathrm{d}}{\mathrm{d}x_3} P_3(\boldsymbol{q}_{||}, x_3) = -iq_{||} \chi_n^{-1}(\omega)\, \varepsilon_n(\omega)\, P_{q_{||}}(\boldsymbol{q}_{||}, x_3) . \tag{40}$$

The solutions (39) and (40) form a complete set of p-polarized polarization eigenmodes: (i) surface phonons, (ii) LO phonons, and (iii) TO phonons.

2.1.1 *p-polarization: surface phonons*

For surface phonons $\varepsilon_n(\omega) \neq 0$ and $\chi_n^{-1}(\omega)\, \varepsilon_n(\omega) \neq 0$ is valid. From (39) and (40) we find

$$\frac{\mathrm{d}^2}{\mathrm{d}x_3^2} P^s_{q_{||}\mathrm{S}}(\boldsymbol{q}_{||}, x_3) = q_{||}^2 P^s_{q_{||}\mathrm{S}}(\boldsymbol{q}_{||}, x_3) \tag{41}$$

with the solutions

$$P^s_{q_{||}\mathrm{S}}(\boldsymbol{q}_{||}, x_3) = iC^s_{\mathrm{S}}[F^s_n\, \mathrm{e}^{q_{||}x_3} - G^s_n\, \mathrm{e}^{-q_{||}x_3}] \tag{42}$$

and

$$P^s_{3\mathrm{S}}(\boldsymbol{q}_{||}, x_3) = C^s_{\mathrm{S}}[F^s_n\, \mathrm{e}^{q_{||}x_3} + G^s_n\, \mathrm{e}^{-q_{||}x_3}] . \tag{43}$$

C^s_{S} is a normalization constant, S denotes surface phonons and $s = 1, 2, 3, \ldots$ Using (42) and (43) for the polarization modes in the integral equations (37) and (38) we obtain a set of homogeneous linear equations in F_n^s and G_n^s. For these field amplitudes we find

$$F_1^s = G_1^s \frac{1-\varepsilon_1}{1+\varepsilon_1} e^{-2q_{\|}a}, \qquad G_1^s = \varepsilon_0 \chi_1 \frac{1+\varepsilon_1}{2\varepsilon_1} e^{q_{\|}a},$$
$$F_2^s = \frac{\chi_2}{\chi_1} \frac{[(\varepsilon_1+\varepsilon_2) F_1^s + (\varepsilon_1-\varepsilon_2) G_1^s]}{2\varepsilon_2}, \tag{44}$$
$$G_2^s = \frac{\chi_2}{\chi_1} \frac{[(\varepsilon_1-\varepsilon_2) F_1^s + (\varepsilon_1+\varepsilon_2) G_1^s]}{2\varepsilon_2}.$$

The condition for the existence of nontrivial p-polarized solutions leads to the implicit dispersion relation given by

$$\frac{(\varepsilon_1+\varepsilon_2)(1-\varepsilon_1) + (\varepsilon_1-\varepsilon_2)(1+\varepsilon_1) e^{2q_{\|}a}}{(\varepsilon_1-\varepsilon_2)(1-\varepsilon_1) + (\varepsilon_1+\varepsilon_2)(1+\varepsilon_1) e^{2q_{\|}a}} - \frac{1+\varepsilon_2}{1-\varepsilon_2} e^{2q_{\|}b} = 0\,. \tag{45}$$

This result for a bilayer system can be easily applied to some special cases such as a layer on a semi-infinite substrate ($b \to \infty$), a free-standing layer of thickness $a (b = 0, \varepsilon_2 = 1)$, and for a half-space geometry ($a \to \infty$, $b \to \infty$).

In the limiting case $q_{\|} \to \infty$ (45) gives

$$(\varepsilon_1 + 1)(\varepsilon_2 + 1)(\varepsilon_1 + \varepsilon_2) = 0$$

and

$$\varepsilon_1 = -1, \qquad \varepsilon_2 = -1, \qquad \varepsilon_1 = -\varepsilon_2\,. \tag{46}$$

From the description above (see also (23) and (27)) the dielectric function of both media is

$$\varepsilon_n(\omega) = \varepsilon_{\infty n} \frac{\omega_{Ln}^2 - \omega^2}{\omega_{Tn}^2 - \omega^2}, \tag{47}$$

with

$$\varepsilon_{\infty n} = 1 + \frac{1}{\varepsilon_0} \frac{n_n \alpha_n}{1 - \frac{1}{3\varepsilon_0} n_n \alpha_n}, \tag{48}$$

$$\omega_{Ln}^2 = \omega_{0n}^2 + \frac{\frac{2}{3}\omega_{pn}^2}{1 + \frac{2}{3\varepsilon_0} n_n \alpha_n}, \tag{49}$$

$$\omega_{Tn}^2 = \omega_{0n}^2 - \frac{\frac{1}{3}\omega_{pn}^2}{1 - \frac{1}{3\varepsilon_0} n_n \alpha_n}, \tag{50}$$

where $\varepsilon_{\infty n}$ is the optical dielectric constant, ω_{Ln} and ω_{Tn} is the longitudinal and the transverse optical (LO and TO) phonon frequency, respectively.

The various physical possibilities of combining two dielectric media arise out of the relative positions of the zeroes and poles of $\varepsilon_1(\omega)$ and $\varepsilon_2(\omega)$. There are only three distinct configurations (we assume without loss of generality $\omega_{T1} < \omega_{T2}$) [7],

A: $\omega_{T1} < \omega_{L1} < \omega_{T2} < \omega_{L2}$,

B: $\omega_{T1} < \omega_{T2} < \omega_{L1} < \omega_{L2}$,

C: $\omega_{T1} < \omega_{T2} < \omega_{L2} < \omega_{L1}$.

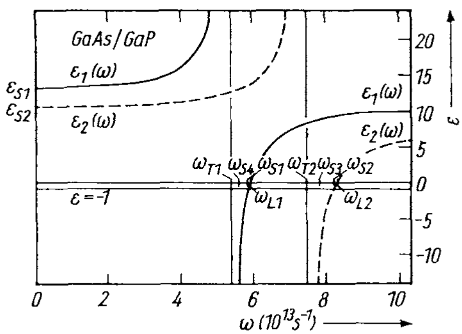


Fig. 2. The dielectric functions $\varepsilon_1(\omega)$ (heavy lines) and $\varepsilon_2(\omega)$ (dashed lines) of GaAs and GaP realizing case A. The frequencies ω_{S1}, ω_{S2}, ω_{S3}, and ω_{S4} are the asymptotic frequencies for large values of the wave vector as described in the text

Equation (45) with the dielectric functions given in (47) yields the asymptotic frequencies ω_{S1}, ω_{S2}, ω_{S3}, and ω_{S4} for large wave vectors

$$\omega_{S1} = \left(\frac{\varepsilon_{\infty1}\omega_{L1}^2 + \omega_{T1}^2}{\varepsilon_{\infty1} + 1}\right)^{1/2}, \tag{51}$$

$$\omega_{S2} = \left(\frac{\varepsilon_{\infty2}\omega_{L2}^2 + \omega_{T2}^2}{\varepsilon_{\infty2} + 1}\right)^{1/2}, \tag{52}$$

$$\omega_{S3,4} = \left\{\frac{1}{2(\varepsilon_{\infty1} + \varepsilon_{\infty2})} \left(\varepsilon_{\infty1}(\omega_{L1}^2 + \omega_{T2}^2) + \varepsilon_{\infty2}(\omega_{L2}^2 + \omega_{T1}^2) \pm \right.\right.$$
$$\pm \left[(\varepsilon_{\infty1}(\omega_{L1}^2 + \omega_{T2}^2) + \varepsilon_{\infty2}(\omega_{L2}^2 + \omega_{T1}^2))^2 - \right.$$
$$\left.\left.- 4(\varepsilon_{\infty1} + \varepsilon_{\infty2})\,(\varepsilon_{\infty1}\omega_{L1}^2\omega_{T2}^2 + \varepsilon_{\infty2}\omega_{L2}^2\omega_{T1}^2)\right]^{1/2}\right)\right\}^{1/2}. \tag{53}$$

A bilayer system composed of two media with dielectric functions given by (47) alway supports exactly four surface phonon modes, each of the media supports two modes (see also Fig. 2). For numerical work we have chosen a GaAs/GaP bilayer system (optical constants see [6]) realizing case A. In Fig. 2 the dielectric function of these two media are given and the four asymptotic frequencies are indicated. To cases B and C there is no essential difference [6, 7]. Fig. 3 and 4 show the dispersion curves of surface phonons for various thicknesses of the two layers. The dispersion curves start at $q_{\|} = 0$ and $\omega = \omega_{T1}$, ω_{L1}, ω_{T2}, or ω_{L2}, respectively. For large values of the wave vector $\boldsymbol{q}_{\|}$ the four dispersion curves approach ω_{S1}, ω_{S2}, ω_{S3}, or ω_{S4}, respectively, which are given by (51) to (53).

The normalization constant in (42) and (43) is given with (29) by

$$C_S^s = \sqrt{q_{\|}} \left\{\frac{\theta_1}{\omega_{p1}^2} \left[F_1^{s^2}(e^{2q_{\|}a} - 1) - G_1^{s^2}(e^{-2q_{\|}a} - 1)\right] + \right.$$
$$\left. + \frac{\theta_2}{\omega_{p2}^2} \left[F_2^{s^2}(1 - e^{-2q_{\|}b}) - G_2^{s^2}(1 - e^{2q_{\|}b})\right]\right\}^{-1/2}. \tag{54}$$

The surface phonon modes are not connected with bulk polarization charges (div $\boldsymbol{P} = 0$) but they are accompanied by the appearance of surface charges at the boundaries of the bilayer system according to

$$\eta_S^{\mathrm{pol},s} = C_S^s \begin{cases} F_1^s\, e^{q_{\|}a} + G_1^s\, e^{-q_{\|}a}; & x_3 = a\,, \\ (F_2^s - F_1^s) + (G_2^s - G_1^s); & x_3 = 0\,, \\ -F_2^s\, e^{-q_{\|}b} - G_2^s\, e^{q_{\|}b}; & x_3 = -b\,. \end{cases} \tag{55}$$

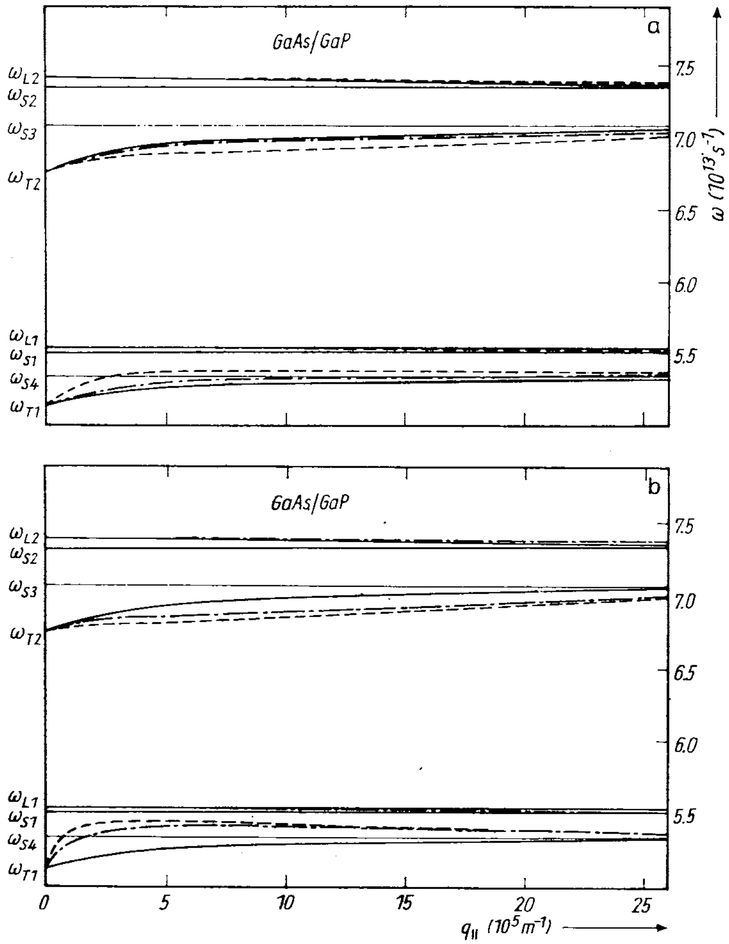


Fig. 3. Dispersion relation of long-wave optical surface phonons of a GaAs/GaP bilayer system for various thicknesses of the GaAs layer. a) ——— $a = 0.3 \times 10^{-6}$ m, $b = 0.3 \times 10^{-6}$ m, —·— $a = 0.4 \times 10^{-6}$ m, – – – – $a = 0.9 \times 10^{-6}$ m; b) ——— $a = 0.3 \times 10^{-6}$ m, $b = 0.3 \times 10^{-6}$ m, —·— $a = 1.5 \times 10^{-6}$ m, – – – – $a = 3.0 \times 10^{-6}$ m

2.1.2 *p-polarization: LO phonons*

For LO phonons we have $\varepsilon_n(\omega_{\mathrm{L}n}) = 0$ and $\varepsilon_p(\omega_{\mathrm{L}n}) \neq 0$, $p \neq n$ and hence for $\omega = \omega_{\mathrm{L}n}$

$$\boldsymbol{D}_{\mathrm{L}n} = 0 \quad \text{and} \quad \boldsymbol{E}_{\mathrm{L}n} = \left(-\frac{1}{\varepsilon_0}\right) \boldsymbol{P}_{\mathrm{L}n}\,.$$

From the differential equations (39) and (40) we obtain in the layer n

$$\frac{\mathrm{d}}{\mathrm{d}x_3}\, P^m_{q_{\parallel}\mathrm{L}n}(\boldsymbol{q}_{\parallel}, x_3) = iq_{\parallel} P^m_{3\mathrm{L}n}(\boldsymbol{q}_{\parallel}, x_3) \tag{56}$$

and in the layer p

$$\frac{\mathrm{d}}{\mathrm{d}x_3}\, P_{q_{\parallel}\mathrm{L}p}(\boldsymbol{q}_{\parallel}, x_3) = q_{\parallel}^2 P_{q_{\parallel}\mathrm{L}p}(\boldsymbol{q}_{\parallel}, x_3) \tag{57}$$

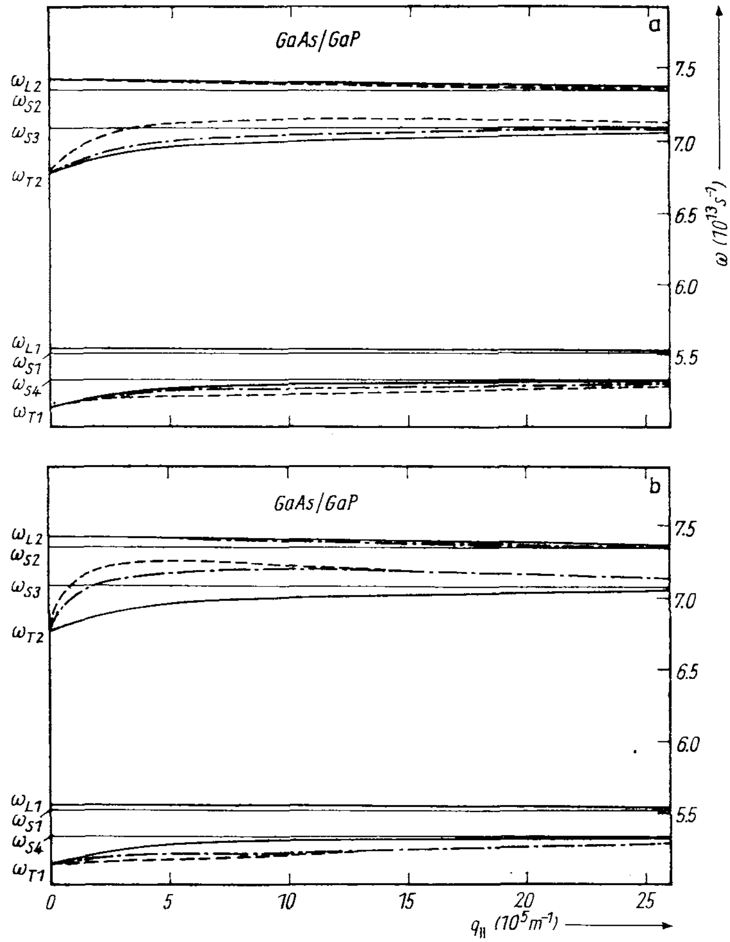


Fig. 4. Dispersion relation of long-wave optical surface phonons of a GaAs/GaP bilayer system for various thicknesses of the GaP layer. a) ——— $a = 0.3 \times 10^{-6}$ m, $b = 0.3 \times 10^{-6}$ m, —·— $b = 0.4 \times 10^{-6}$ m, - - - - $b = 0.9 \times 10^{-6}$ m; b) ——— $a = 0.3 \times 10^{-6}$ m, $b = 0.3 \times 10^{-6}$ m, —·— $b = 1.5 \times 10^{-6}$ m, - - - - $b = 3.0 \times 10^{-6}$ m

The solution of (57) is given by (42) and (43). The amplitudes F_p and G_p are determined by the boundary conditions for the fields $\boldsymbol{E}$ and $\boldsymbol{D}$ at the surfaces of the layer p. These conditions yield $F_p = G_p = 0$ so that $\boldsymbol{P}$, $\boldsymbol{E}$, and $\boldsymbol{D}$ are zero outside the layer n. Inside the layer n (56) and the boundary conditions result in

$$\boldsymbol{P}^m_{\mathrm{L}n}(\boldsymbol{q}_{\|}, x_3) = C^m_{\mathrm{L}n} \left[i \sin\left(q^m_n x_3\right) \boldsymbol{e}_{q_{\|}} + \frac{q^m_n}{q_{\|}} \cos\left(q^m_n x_3\right) \boldsymbol{e}_3 \right] \tag{58}$$

with

$$q^m_n = \frac{\pi}{a_n}\, m; \qquad m = 1, 2, 3 \ldots; \qquad a_1 = a; \qquad a_2 = b\,.$$

The LO phonon modes of the layer n are highly degenerated because all of them have the same frequency ω_{Ln}. The normalization constant is given by

$$C^m_{Ln} = \frac{\omega_{pn}}{\theta_n^{1/2}} \sqrt{\frac{2}{a_n}} \frac{q_{||}}{\sqrt{q_{||}^2 + (q_n^m)^2}} . \tag{59}$$

The LO phonons are connected with both bulk polarization charges

$$\varrho^{\mathrm{pol},m}_{Ln} = C^m_{Ln} \left(q_{||} + \frac{(q_n^m)^2}{q_{||}}\right) \sin (q_n^m x_3) \tag{60}$$

and surface polarization charges

$$\eta^{\mathrm{pol},m}_{L1} = C^m_{L1} \frac{q_1^m}{q_{||}} \begin{cases} (-1)^m; & x_3 = a , \\ -1; & x_3 = 0 , \end{cases}$$

$$\eta^{\mathrm{pol},m}_{L2} = -C^m_{L2} \frac{q_2^m}{q_{||}} \begin{cases} (-1)^m; & x_3 = -b , \\ -1; & x_3 = 0 . \end{cases} \tag{61}$$

2.1.3 *p-polarization: TO phonons*

For TO phonons we have

$$\chi_n^{-1}(\omega_{Tn}) = 0 \quad \text{and} \quad \chi_p^{-1}(\omega_{Tn}) \neq 0, \qquad p \neq n$$

and hence for $\omega = \omega_{Tn}$

$$\boldsymbol{D}_{Tn} = \boldsymbol{P}_{Tn} \quad \text{and} \quad \boldsymbol{E}_{Tn} = 0 .$$

Analogous to the case of LO phonon modes we obtain

$$\boldsymbol{P}^m_{Tn}(\boldsymbol{q}_{||}, x_3) = C^m_{Tn} \left[i \frac{q_n^m}{q_{||}} \cos (q_n^m x_3)\, \boldsymbol{e}_{q_{||}} + \sin (q_n^m x_3)\, \boldsymbol{e}_3 \right]. \tag{62}$$

The TO phonon modes of the layer n are also highly degenerated. The normalization constant is given by

$$C^m_{Tn} = \frac{\omega_{pn}}{\theta_n^{1/2}} \sqrt{\frac{2}{a_n}} \frac{q_{||}}{\sqrt{q_{||}^2 + (q_n^m)^2}} . \tag{63}$$

The TO phonon modes are not accompanied by the appearance either of surface polarization charges or of bulk ones.

2.1.4 *s-polarization*

The s-polarized modes are solutions of (36). For nontrivial solutions of (36) $\chi_n^{-1}(\omega) = 0$ must be used. That means only for $\omega = \omega_{T1}$ or $\omega = \omega_{T2}$ s-polarized TO phonon modes exist. Analogous to the case of p-polarized TO phonon modes we obtain for $\omega = \omega_{Tn}$ $D_{sTn} = P_{sTn}$ and $E_{sTn} = 0$ (in the whole space).

The eigenfunctions can be arbitrary functions of x_3, and we may expand it in a complete set of orthonormal eigenfunctions,

$$P^{(+)j}_{sTn} (\boldsymbol{q}_{||}, x_3) = A^j_{Tn} \sin \left(\frac{\pi}{a_n} j x_3\right); \qquad j = 1, 2, 3 \dots , \tag{64}$$

$$P^{(-)j}_{sTn} (\boldsymbol{q}_{||}, x_3) = B^j_{Tn} \cos \left(\frac{\pi}{a_n} j x_3\right); \qquad j = 0, 1, 2 \dots . \tag{65}$$

The s-polarized TO phonon modes are degenerate, and they are not accompanied by the appearance either of a surface polarization charge or a bulk one.

The p-polarized modes are completely decoupled from the s-polarized modes, and both polarization eigenmode systems satisfy their own closure relation

$$\sum_i \boldsymbol{P}_{\mathrm{p}}^{i*}(\boldsymbol{q}_{||}, x_3)\, \boldsymbol{P}_{\mathrm{p}}^{i}(\boldsymbol{q}_{||}, x_3') = \frac{\omega_{\mathrm{p}n}^2}{\theta_n}\, \delta(x_3 - x_3')\,, \tag{66}$$

where p denotes the p-polarization and i runs over all p-polarized polarization eigenmodes, and

$$\sum_j P_{\mathrm{sT}n}^{j*}(\boldsymbol{q}_{||}, x_3)\, P_{\mathrm{sT}n}^{j}(\boldsymbol{q}_{||}, x_3') = \frac{\omega_{\mathrm{p}n}^2}{\theta_n}\, \delta(x_3 - x_3')\,. \tag{67}$$

2.2 Hamiltonian of the polarization eigenmodes

According to the equation of motion (3) we find the Hamiltonian density

$$\mathscr{H}_{\mathrm{p}} = \frac{1}{2} \sum_\alpha \left\{\frac{1}{n_n\mu_n}\, \Pi_\alpha \Pi_\alpha + n_n\mu_n\omega_{0n}^2 u_\alpha u_\alpha - n_n e_n^* u_\alpha E_\alpha^{\mathrm{loc}}\right\}, \tag{68}$$

where

$$\boldsymbol{\Pi} = n_n\mu_n\dot{\boldsymbol{u}}$$

is the canonical momentum function. The Hamiltonian function H_{p} is given by the space integral over $\mathscr{H}_{\mathrm{p}}$,

$$H_{\mathrm{p}} = \frac{1}{2} \sum_\alpha \int \mathrm{d}^3x \left\{\frac{1}{n_n\mu_n}\, \Pi_\alpha \Pi_\alpha + n_n\mu_n\omega_{0n}^2 u_\alpha u_\alpha - n_n e_n^* u_\alpha E_\alpha^{\mathrm{loc}}\right\}. \tag{69}$$

With the classical Poisson brackets [8] for field functions,

$$\dot{u}_\alpha = \{u_\alpha, H_{\mathrm{p}}\}; \qquad \dot{\Pi}_\alpha = \{\Pi_\alpha, H_{\mathrm{p}}\}\,, \tag{70}$$

the Hamiltonian function (69) yields the equation of motion (3). Using the symmetry of the bilayer system by introducing a two-dimensional Fourier transform according (17) we can write

$$H_{\mathrm{p}} = \frac{1}{8\pi^2} \sum_\alpha \int \mathrm{d}^2q_{||} \int \mathrm{d}x_3 \left\{\frac{1}{n_n\mu_n}\, \Pi_\alpha^*(\boldsymbol{q}_{||}, x_3)\, \Pi_\alpha(\boldsymbol{q}_{||}, x_3) + {} \right.$$
$$\left. {} + n_n\mu_n\omega_{0n}^2 u_\alpha^*(\boldsymbol{q}_{||}, x_3)\, u_\alpha(\boldsymbol{q}_{||}, x_3) - n_n e_n^* u_\alpha^*(\boldsymbol{q}_{||}, x_3)\, E_\alpha^{\mathrm{loc}}(\boldsymbol{q}_{||}, x_3)\right\}, \tag{71}$$

where $\boldsymbol{\Pi}$, $\boldsymbol{u}$, and $\boldsymbol{E}^{\mathrm{loc}}$ are time-dependent. For the local field we obtain

$$\boldsymbol{E}^{\mathrm{loc}}(\boldsymbol{q}_{||}, x_3) = \Phi_n \boldsymbol{P}(\boldsymbol{q}_{||}, x_3) \tag{72}$$

with

$$\Phi_n = \frac{\lambda_{0n} - \lambda_n}{1 + n_n\alpha_n(\lambda_{0n} - \lambda_n)}\,. \tag{73}$$

The relation between the displacement $\boldsymbol{u}$ and the polarization $\boldsymbol{P}$ is given by

$$\boldsymbol{u}(\boldsymbol{q}_{||}, x_3) = \frac{\theta_n^{1/2}}{n_n e_n^*}\, \boldsymbol{P}(\boldsymbol{q}_{||}, x_3)\,. \tag{74}$$

With (72) and (73) we can write the Hamiltonian function (71) in the form

$$H_{\rm p} = \frac{1}{2A} \sum_{\boldsymbol{q}_{||}} \sum_{\alpha} \int \mathrm{d}x_3 \, \frac{\theta_n}{\varepsilon_0 \omega_{\mathrm{p}n}^2} \, \{\dot{P}_\alpha^*(\boldsymbol{q}_{||}, x_3) \, \dot{P}_\alpha(\boldsymbol{q}_{||}, x_3) + $$
$$+ \omega^2 P_\alpha^*(\boldsymbol{q}_{||}, x_3) \, P_\alpha(\boldsymbol{q}_{||}, x_3)\} \,, \tag{75}$$

where we have introduced Born- von Kármán periodic boundary conditions in the x_1–x_2 plane according to

$$\int \mathrm{d}^2 q_{||} \to \frac{(2\pi)^2}{A} \sum_{\boldsymbol{q}_{||}} \quad \text{and} \quad \delta(\boldsymbol{q}_{||} - \boldsymbol{q}_{||}') \to \frac{A}{(2\pi)^2} \, \delta_{\boldsymbol{q}_{||}\boldsymbol{q}_{||}'} \,,$$

where A is the unit area of the bilayer system in the x_1–x_2 plane. Proceeding in the standard manner the polarization $\boldsymbol{P}(\boldsymbol{q}_{||}, x_3)$ is represented in terms of the complete set of orthonormal polarization eigenmodes $\boldsymbol{P}^i(\boldsymbol{q}_{||}, x_3)$,

$$\hat{\boldsymbol{P}}(\boldsymbol{q}_{||}, x_3) = \sum_i \left(\frac{\hbar \varepsilon_0 A}{2\omega_i(\boldsymbol{q}_{||})}\right)^{1/2} \left(\hat{a}_i(\boldsymbol{q}_{||}) + \hat{a}_i^+(-\boldsymbol{q}_{||})\right) \boldsymbol{P}^i(\boldsymbol{q}_{||}, x_3) \,, \tag{76}$$

$$\hat{\dot{\boldsymbol{P}}}(\boldsymbol{q}_{||}, x_3) = i \sum_i \left(\frac{\hbar \varepsilon_0 A \omega_i(\boldsymbol{q}_{||})}{2}\right)^{1/2} \left(\hat{a}_i^+(-\boldsymbol{q}_{||}) - \hat{a}_i(\boldsymbol{q}_{||})\right) \boldsymbol{P}^i(\boldsymbol{q}_{||}, x_3) \,. \tag{77}$$

In (76) and (77) the vectors $\hat{\boldsymbol{P}}$ and $\hat{\dot{\boldsymbol{P}}}$ are now considered as quantum field operators denoted by the cap. The following commutation relations are valid:

$$[\hat{\dot{P}}_\alpha \, (\boldsymbol{q}_{||}, x_3), \hat{P}_\beta(\boldsymbol{q}_{||}', x_3')] = \frac{\hbar}{i} \, \frac{\varepsilon_0 \omega_{\mathrm{p}n}^2 A}{\theta_n} \, \delta_{\boldsymbol{q}_{||}, -\boldsymbol{q}_{||}'} \, \delta(x_3 - x_3') \, \delta_{\alpha\beta} \,, \tag{78}$$

$$[\hat{\dot{P}}_\alpha(\boldsymbol{q}_{||}, x_3), \hat{\dot{P}}_\beta(\boldsymbol{q}_{||}', x_3')] = [\hat{P}_\alpha(\boldsymbol{q}_{||}, x_3), \hat{P}_\beta(\boldsymbol{q}_{||}', x_3')] = 0 \,.$$

These commutation relations yield the well-known commutation relation for the operators $\hat{a}_i^+$ and $\hat{a}_i$,

$$[\hat{a}_i(\boldsymbol{q}_{||}), \hat{a}_j^+(\boldsymbol{q}_{||}')] = \delta_{\boldsymbol{q}_{||}\boldsymbol{q}_{||}'} \delta_{ij} \,,$$
$$[\hat{a}_i^+(\boldsymbol{q}_{||}), \hat{a}_j^+(\boldsymbol{q}_{||}')] = [\hat{a}_i(\boldsymbol{q}_{||}), \hat{a}_j(\boldsymbol{q}_{||})] = 0 \,. \tag{79}$$

The operator $\hat{a}_i^+$ is the *creation* and $\hat{a}_i$ the *annihilation operator of long-wave optical phonons of the bilayer system.* Using the relations (79) and the orthonormalization relation (29) we obtain the *Hamiltonian of the polarization eigenmodes* in the usual diagonalized form,

$$\hat{H}_{\rm p} = \hat{H}_{\rm p}^{\rm p} + \hat{H}_{\rm p}^{\rm s} \,, \tag{80}$$

$$\hat{H}_{\rm p}^{\rm p} = \sum_i \sum_{\boldsymbol{q}_{||}} \hbar \omega_i(\boldsymbol{q}_{||}) \, [\hat{a}_{\mathrm{p}i}^+(\boldsymbol{q}_{||}) \, \hat{a}_{\mathrm{p}i}(\boldsymbol{q}_{||}) + \tfrac{1}{2}] \tag{81}$$

and

$$\hat{H}_{\rm p}^{\rm s} = \sum_j \sum_{\boldsymbol{q}_{||}} \hbar \omega_{\mathrm{T}n} \, [\hat{a}_{\mathrm{s}j}^+(\boldsymbol{q}_{||}) \, \hat{a}_{\mathrm{s}j}(\boldsymbol{q}_{||}) + \tfrac{1}{2}] \,. \tag{82}$$

The Hamiltonian $\hat{H}_{\rm p}^{\rm s}$ of the s-polarized long-wave optical phonons of the bilayer system and the corresponding Hamiltonian $\hat{H}_{\rm p}^{\rm p}$ of the p-polarization are completely decoupled from each other. In $\hat{H}_p^p$ the summation extends over all p-polarized polarization eigenmodes of the bilayer system: long-wave optical surface, LO and TO phonons.

Since $\hat{H}_p$ is the Hamiltonian of long-wave optical phonons of the bilayer system, the summation over $\boldsymbol{q}_{||}$ is limited by the continuum approximation. The summation over $\boldsymbol{q}_{||}$ should be performed up to the limit $q < q_c$ where $\boldsymbol{q}_c$ is the cut-off wave vector of the continuum approximation and $\boldsymbol{q} = \boldsymbol{q}_{||} + \boldsymbol{q}_n^m \boldsymbol{e}_3$ (for surface modes is $q_n^m = 0$). We can roughly estimate q_c to be in the order of 10^8 m^{-1}.

The Hamiltonian (80) to (82) together with the polarization eigenmodes and eigenfrequencies defined in Section 2.1 gives a complete description of the phonon modes of a bilayer system in the long-wave approximation.

3. Electron-Phonon Interaction

The interaction energy of an electron (charge $-e$) at the position $\boldsymbol{x}^e$ with a polarizable medium is

$$H_{ep} = -e\Phi(\boldsymbol{x}^e, t) . \tag{83}$$

Using the expression for the scalar potential $\Phi(\boldsymbol{x}^e, t)$ (6) and the symmetry of the bilayer system by introducing the two-dimensional Fourier transform, we obtain in a straightforward way

$$H_{ep} = \frac{e}{2A\varepsilon_0} \sum_{\boldsymbol{q}_{||}} \int_{-b}^{a} dx_3\, e^{i\boldsymbol{q}_{||}\boldsymbol{x}_{||}^e}\, e^{-q_{||}|x_3 - x_3^e|} \times \times [i\boldsymbol{e}_{q_{||}} + \operatorname{sgn}(x_3 - x_3^e)\, \boldsymbol{e}_3]\, \boldsymbol{P}(\boldsymbol{q}_{||}, x_3, t) . \tag{84}$$

Making use of the representation of $\boldsymbol{P}(\boldsymbol{q}_{||}, x_3, t)$ in terms of polarization eigenmodes the Hamiltonian of the electron–phonon interaction has finally the form

$$\hat{H}_{ep} = \sum_i \sum_{\boldsymbol{q}_{||}} e^{i\boldsymbol{q}_{||}\boldsymbol{x}_{||}^e}\, \Gamma_i(\boldsymbol{q}_{||}, x_3^e)\, (\hat{a}_i(\boldsymbol{q}_{||}) + \hat{a}_i^+(-\boldsymbol{q}_{||})), \tag{85}$$

where the coupling functions Γ_i are defined by

$$\Gamma_i(\boldsymbol{q}_{||}, x_3^e) = \left(\frac{\hbar e^2}{8A\varepsilon_0\omega_i(\boldsymbol{q}_{||})}\right)^{1/2} \int_{-b}^{a} dx_3\, e^{-q_{||}|x_3 - x_3^e|} \times \times [i\boldsymbol{e}_{q_{||}} + \operatorname{sgn}(x_3 - x_3^e)\, \boldsymbol{e}_3]\, \boldsymbol{P}^{i*}(\boldsymbol{q}_{||}, x_3) . \tag{86}$$

The coupling functions Γ_i can be expressed by the macroscopic electric field. Starting with

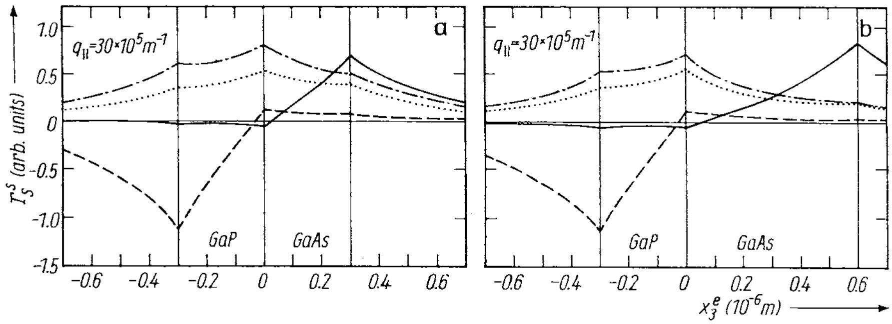
$$E_{q_{||}}(\boldsymbol{q}_{||}, x_3) = -iq_{||}\Phi(\boldsymbol{q}_{||}, x_3) \tag{87}$$

we obtain with (83) and (84) an expression equivalent to (86),

$$\Gamma_i(\boldsymbol{q}_{||}, x_3^e) = -\left(\frac{\hbar e^2\varepsilon_0}{2A\omega_i(\boldsymbol{q}_{||})}\right)^{1/2} \frac{i}{q_{||}}\, E_{q_{||}}^i(\boldsymbol{q}_{||}, x_3^e) . \tag{88}$$

Inserting in (86) the various polarization eigenmodes from Section 2.1 one finds that the electron does not couple to TO phonon modes of p- and s-polarization. The reason for this is that TO phonon modes are not accompanied by the appearance either of a surface polarization charge or a bulk one.

The numerical work has been done for a GaAs/GaP bilayer system. We have calculated the coupling functions Γ_i (86) for the interaction of the electron with surface phonon modes (42), (43) and with LO phonon modes (58). Since the media are isotropic



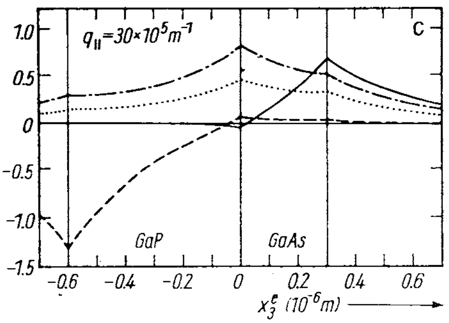


Fig. 5. Spatial dependence of the coupling functions $\Gamma_S^s(q_{\|}, x_3^e)$ of the electron–(long-wave optical) surface phonon interaction for various thicknesses of the two layers of the GaAs/GaP bilayer system ($q_{\|} = 30 \times 10^5$ m^{-1}). ——— Γ_S^1, – – – – Γ_S^2, —·— Γ_S^3, and ······ Γ_S^4. a) $a = 0.3 \times 10^{-6}$ m, $b = 0.3 \times 10^{-6}$ m, b) $a = 0.6 \times 10^{-6}$ m, $b = 0.3 \times 10^{-6}$ m, c) $a = 0.3 \times 10^{-6}$ m, $b = 0.6 \times 10^{-6}$ m

Γ_i is only a function of $q_{\|} = |\boldsymbol{q}_{\|}|$ and not of the direction of $\boldsymbol{q}_{\|}$. Fig. 5 to 8 show the spatial dependence of the coupling functions $\Gamma_S^s(q_{\|}, x_3^e)$ of the electron coupling with the four surface phonon modes ($s = 1, 2, 3, 4$) for the GaAs/GaP bilayer system.

As can be seen from (88) the spatial dependence of the coupling functions Γ_i is strongly correlated to the spatical dependence of the electric fields associated with the phonons [6]. The coupling function of the electron–surface phonon interaction is mainly localized at the surfaces and the interface of the bilayer system (see Fig. 5 to 7). Furthermore an interaction takes place even when the electron is outside the bilayer system close to one of its surfaces. This effect is used in EELS measurements. Surface mode 1 (2) is supported by the GaAs(GaP) layer and consequently this mode gives the main contribution to the interaction if the electron is localized close to the GaAs (GaP) surface of the bilayer system.The behaviour is analogous in case of the interface modes 3 and 4, they are dominant if the electron is localized close to the interface between the GaAs and the GaP layer. It is illustrated that the localization of the coupling function Γ_S^s at the surfaces increases with larger values of the wave vector $q_{\|}$ (see Fig. 5a and 6). Fig. 7 shows the coupling functions Γ_S^s that are calculated

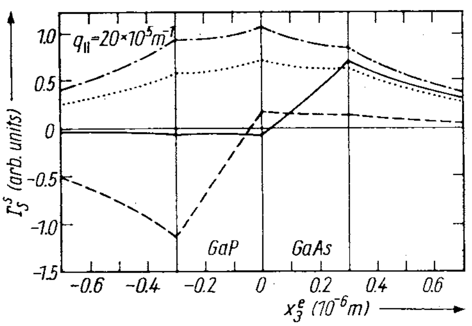


Fig. 6. Spatial dependence of the coupling functions $\Gamma_S^s(q_{\|}, x_3^e)$ of the electron–(long-wave optical) surface phonon interaction of the GaAs/GaP bilayer system ($q_{\|} = 20 \times 10^5$ m^{-1}). ——— Γ_S^1, – – – – Γ_S^2, —·— Γ_S^3, ······· Γ_S^4

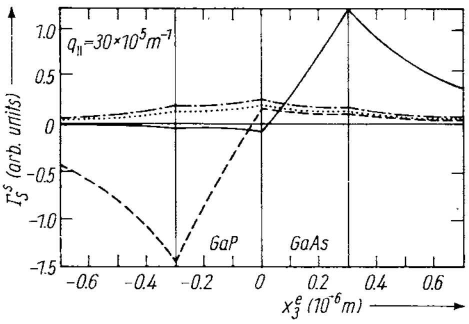


Fig. 7. Spatial dependence of the coupling functions $\Gamma_S^s(q_{\|}, x_3^e)$ of the electron–(long-wave optical) surface phonon interaction by neglecting the contribution of the electronic polarizability ($\theta_n = 1$) of the GaAs/GaP bilayer system ($q_{\|} = 30 \times \times 10^5$ m^{-1}). ——— Γ_S^1, - - - Γ_S^2, —·—·— Γ_S^3, · · · · · · Γ_S^4

with $\theta_n = 1$ in (86), which corresponds to an insuffient contribution of the electronic polarizability as it is often done by other autors. A comparison of the results given in Fig. 5a and 7 shows that the electronic polarizability plays an important role in the electron–surface phonon interaction. The difference in the strength of the interaction is certainly a non-negligible effect, especially for the technologically important $A^{III}B^{V}$ semiconductors.

If the electron is inside the GaAs (GaP) layer it interacts also with the LO phonon modes of these layers. As is seen in Fig. 8 the interaction strength of the electron with the LO phonon modes vanishes outside the bilayer system and at the surfaces, and inside the layer it is small near the surface because of the factor sin $(q_n^m x_3)$ appearing in (86) with (58).

Thus, when the electron is close to the surface but inside the layer the main contribution to the strength of the electron–phonon interaction arises from that with surface modes. In this case the electron and the surface phonon mode form a new surface elementary excitation called *surface polaron*. The shift in energy ΔE of the electron caused by the interaction $\hat{H}_{ep}$ can be estimated by second-order pertubation theory to be proportional to $|\Gamma|^2$. In this process described by this second-order correction the electron emits a virtual phonon of wave vector $q_{\|}$ and then reabsorbs it. Thus, when the electron is close to the surface but inside the bilayer system it will be attracted to the surface just as if it is outside. If the electron is in one of both layers and close to

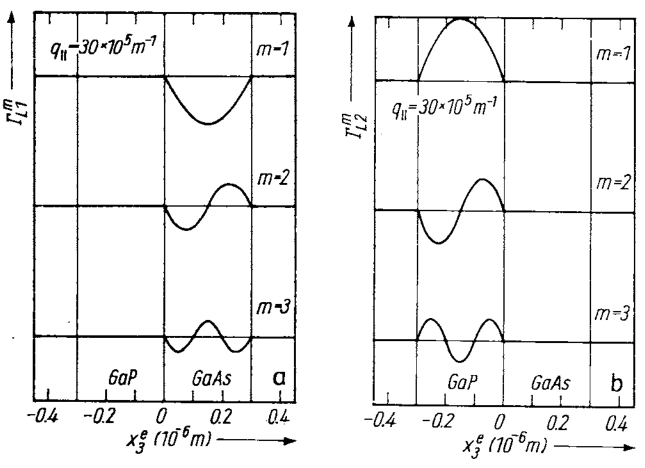


Fig. 8. Spatial dependence of the coupling functions $\Gamma_{Ln}^m(q_{\|}, x_3^e)$ of the electron–LO phonon interaction of the GaAs/GaP bilayer system ($q_{\|} = 30 \times 10^5$ m^{-1}). a) $\Gamma_{L1}^m(q_{\|}, x_3^e)$, b) $\Gamma_{L2}^m(q_{\|}, x_3^e)$

the interface it will be attracted to the interface. This result is completely opposite to that obtained from electrostatic considerations. From the theory of the classical image charges one obtains that the image charge is of the same sign as the electron inside the layer, so the electron is repelled from the surface. If the electron is far away from the bilayer system classical theory of image charge and second-order pertubation theory with the Hamiltonian (85) lead to the same results for ΔE.

4. Concluding Remarks

In this paper a quantum-mechanical theory of electron–(long-wave optical) phonon interaction in bilayer systems has been developed. Many experiments have been performed with bi- and multilayer systems, or with a single layer on a substrate, and they have stimulated the theoretical study of such systems. For instance the physical processes occurring in EELS for electrons reflected or transmitted by a bilayer system or XPS from bilayer systems are successfully treated in the present formalism. Typical quantum-mechanical features, such as multiple-excitation processes in EELS are naturally incorporated in the treatment here given. Since we have used both microscopic and macroscopic relations it is easy to generalize the results to an arbitrary dielectric function $\varepsilon(\omega)$. We must insert this dielectric function in (28) which yields an altered normalization relation, and can use (86) to calculate the coupling functions Γ_i. For example, this generalization enables us to study also metallic bilayer systems (or the combinations dielectric/metal, dielectric/n-type semiconductor) where electrons behave like those of a quasi-free electron gas.

The theory developed is also applicable to the problem of a conduction electron in a bilayer system (polar semiconductors) interacting with the long-wave optical phonon modes [9]. In this case the Hamiltonian derived here for the bilayer system is analoguous to the well-known Fröhlich polaron problem in the bulk. There is currently a revival of interest in the study of this problem [9 to 15]. In modern microelectronic devices based for example on $GaAs_x/Ga_xAl_{1-x}As$ heterostructures the contribution from the electron–phonon interaction influences the electronic properties noticeably [16]. The electron–phonon interaction is an important scattering mechanism limiting the mobility of the free carriers.

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