Microscopic Screening and Phonon Dispersion of Silicon: Moment Expansion for the Polarizability

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A phonon dispersion curve of Si is calculated using the microscopic theory of dielectric screening. The electron energies and wave functions are calculated from a crystal pseudopotential which is self-consistent with the electron-ion pseudopotential. For the polarizability matrix a continued fraction expansion including the zeroth and first moment is used. The first moment is treated as a constant. The results are sensitive to both the self-consistency condition and the closure approximation.

In the microscopic theory of lattice vibrations the effect of the electrons on the forces between the ions is taken explicitly into account. This leads to two contributions to the dynamical matrix: a direct ion-ion term and an ion-electron-ion term which describes that part of the interionic interaction mediated by the electrons. The direct ion-ion term is well known and is calculated in the usual way by an Ewald procedure. The indirect ion-electron-ion term is given by the following expression[1]:

$$D_{ij}^{en}(\vec{q}; ab) = [\sum_{\vec{G}, \vec{G}} X_{ij}(ab; \vec{q}, \vec{G}, \vec{G}') - \delta_{a,b}\sum_{c} X_{ij}(ac; 0, \vec{G}, \vec{G}')] \tag{1}$$

with

$$X_{ij}(ab; \vec{q}, \vec{G}, \vec{G}') = \frac{1}{(M_a M_b)^{1/2}} \frac{1}{v_c} [(\vec{q}+\vec{G})_i \, V_a^*(\vec{q}+\vec{G}) \exp(i\vec{G}\cdot\vec{R}_a) \, \chi(\vec{q}, \vec{G}, \vec{G}')$$
$$\times \exp(-i\vec{G}'\cdot\vec{R}_b) V_b(\vec{q}+\vec{G}')(\vec{q}+\vec{G}')_j] , \tag{2}$$

where i and j refer to the Cartesian coordinates, and a, b, and c refer to the atoms in the unit cell. $\vec{q}$ is the phonon wave vector and $\vec{G}$ and $\vec{G}'$ are reciprocal-lattice vectors. M_a and M_b are the masses of atoms a and b and v_c is the volume of the unit cell. V_a and V_b are the form factors of the electron-ion potential. χ is the density response matrix which describes the linear response of the electrons to an external potential. This matrix is related to the inverse dielectric matrix ϵ^{-1} and the polarizability matrix by the following relations:

$$\epsilon = 1 - v\tilde{\chi}$$

and

$$\epsilon^{-1} = 1 + v\chi ,$$

where v is the electron-electron potential.

In the Hartree approximation, the polarizability matrix is given by[2]

$$\tilde{\chi}(\vec{q}, \vec{G}, \vec{G}') = \frac{1}{\Omega} \sum_{k,l}' \frac{\eta_k}{E_k - E_l} (\langle k | e^{-i(\vec{q}+\vec{G})\cdot\vec{r}} | l \rangle \langle l | e^{i(\vec{q}+\vec{G}')\cdot\vec{r}} | k \rangle + \langle l | e^{-i(\vec{q}+\vec{G})\cdot\vec{r}} | k \rangle \langle k | e^{i(\vec{q}+\vec{G}')\cdot\vec{r}} | l \rangle) . \tag{3}$$

The indices k and l refer to the electron quantum numbers, i.e., the wave vector and band number. η_k and η_l are the occupation numbers of the electron states $|k\rangle$ and $|l\rangle$. The prime on the summation excludes states with the same indices.

Calculations of the phonon dispersion by the present authors based on Eq. (3) have shown that for a direct summation of up to forty conduction bands the square of the TA mode remains negative for all wave vectors in the Δ direction, including the X point.[3] It is also found that the convergence of the phonon frequencies with respect to the number of conduction bands is rather slow. The first term in a Taylor expansion of the wave function with respect to the displacement of the ions is represented by a perturbation expansion. Sinha[4] showed before that such an expansion does not converge well at all in the case of a displaced hydrogen atom. A similar slow convergence of the number of conduction bands has been reported for the elements of the polarizability matrix.[5]

This convergence problem is inherent to Eq. (3) since in practice only a limited number of conduction states can be taken into account. In order to investigate the effect of the higher conduction bands,

the following approach has been adopted. The polarizability matrix is expanded in a continued-fraction series[6] with respect to the summation index l. Using such an expansion, the summation over the index l can be eliminated in each moment via the completeness relations, so that closure over all the states is obtained.

In the present calculation only the zeroth and first moments are retained. The polarizability matrix can be written as follows:

$$\chi(\vec{q}, \vec{G}, \vec{G}') = \frac{2}{\Omega} \sum_k \frac{\eta_k \, \mu_0(k)}{E_k - \mu_1(k)/\mu_0(k)}. \tag{4}$$

The zeroth moment is equal to

$$\mu_0(k) = \langle k | \exp[i(\vec{G}' - \vec{G}) \cdot \vec{r}] | k \rangle - \sum_l \eta_l \langle k | \exp[-i(\vec{q} + \vec{G}) \cdot \vec{r}] | l \rangle \langle l | \exp[i(\vec{q} + \vec{G}') \cdot \vec{r}] | k \rangle \tag{5}$$

and the first moment

$$\mu_1(k) = \langle k | \exp[-i(\vec{q} + \vec{G}) \cdot \vec{r}] H_0 \exp[i(\vec{q} + \vec{G}') \cdot \vec{r}] | k \rangle$$

$$- \sum_l \eta_l \langle k | \exp[-i(\vec{q} + \vec{G}) \cdot \vec{r}] | l \rangle \langle l | H_0 \exp[i(\vec{q} + \vec{G}') \cdot \vec{r}] | k \rangle, \tag{6}$$

where H_0 is the pseudopotential Hamiltonian.

As an additional approximation, μ_1/μ_0 in the denominator of Eq. (4) has been treated as a constant, E_0. This constant is used as a parameter.

The formula for the polarizability in Eq. (4) is a refinement of the expression reported earlier by the present authors[7] where the energy denominator [Eq. (3)] is approximated by a constant and set equal to the Phillips's band gap.[8]

It should be noted that in both Eqs. (5) and (6) the sum over l includes all the valence states. The first term of μ_0, which is the Fourier transform of the charge density, is independent of $\vec{q}$. For small values of the wave vector $\vec{q}$, the second term of μ_0 cancels the first term and gives an additional contribution proportional to q^2. This means that in the limit as $\vec{q}$ goes to zero, χ is also proportional to q^2, as it should be for a semiconductor. If only one state $l = k$ had been subtracted from the first term on the right-hand side of Eq. (5), χ would have been a constant for small values of $\vec{q}$.

As a result of treating μ_1 as a constant, the diagonal of the polarizability is also a constant for large values of $\vec{G}$ and $\vec{G}'$. Instead, it should behave as $1/G^2$.

The one-electron energies and wave functions needed in Eq. (5) are calculated in the pseudopotential method. The crystal pseudopotential, which enters in the Hamiltonian, is calculated iteratively to self-consistency,[9] starting from a Topp-Hopfield electron-ion potential:

$$V_a(\vec{r}) = V_0 \cos\beta r + V_1, \quad r \leqslant r_c$$

$$V_a(\vec{r}) = -Z_a/r, \quad r > r_c, \tag{7}$$

where $r_c = 1.6$ a.u. and $\beta = 2.2156$ (a.u.)$^{-1}$. The

constants V_0 and V_1 are determined by requiring that the potential and the first derivative are continuous at $r = r_c$.

The self-consistency between the crystal and electron-ion pseudopotential plays an essential role in obtaining positive transverse-acoustical frequencies. This self-consistency makes the wave functions in χ and the electron-ion pseudopotentials V_a and V_b, in Eq. (1), compatible. Because of this compatibility, the generalized acoustic sum rule, which gives a relationship between the electron density response matrix χ and the charge density, is automatically satisfied.[10] It should be emphasized that the constants r_c and β are not used as parameters to fit the phonon frequencies.

In the present calculation, the origin has been chosen halfway in between the atoms of the unit cell. Consequently all matrices—Hamiltonian as well as dielectric matrix—are real. The lattice constant is 5.417 Å. A dimension of 59 has been taken for the Hamiltonian matrix in the Γ point. The dimension of the polarizability matrix is 27 for $\vec{q} = 0$.

The resulting phonon dispersion for a calculation with the parameter $E_0 = 10.88$ eV is shown in Fig. 1. This value of E_0 has been chosen in the following way: As a first approximation, the average over the Brillouin zone of the denominator of Eq. (4) is set equal to minus the average of the energy difference between the lowest conduction and highest valence bands. This average is also equal to the Phillips band gap, ΔE. For Si, $\Delta E = 4.08$ eV and the average of the valence-band energies is 6.8 eV. As a result, one obtains overall meaningful dispersion curves.
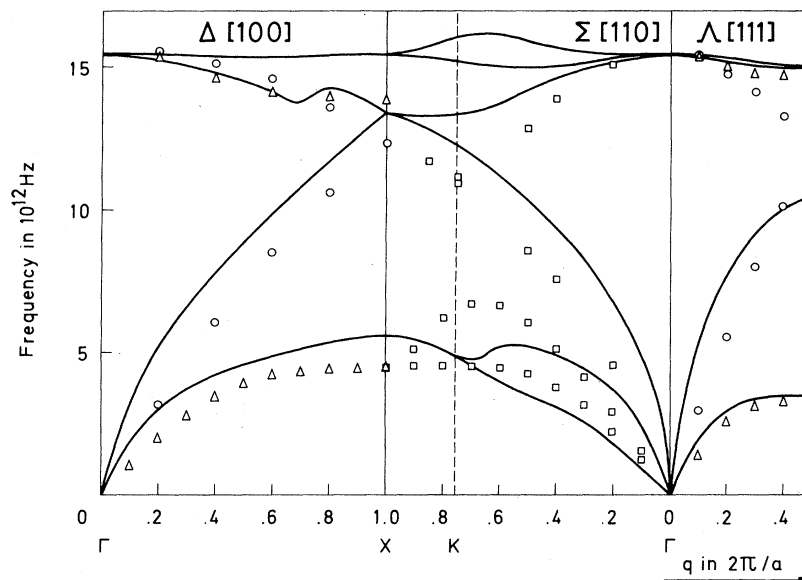
FIG. 1. Phonon frequencies obtained in the closure approximation with the crystal pseudopotential which is self-consistent with the Topp-Hopfield electron-ion potential (solid line). For comparison, the experimental data points (Ref. 11) are shown (circle, longitudinal modes; triangle, transverse modes).

The TO mode in the Δ direction still increases slightly in contradiction with experiment. The TA mode in the X point is within 17% of the experimental value. In the approximations used here, it is found that the determinants of $\bar{\chi}$ and χ, are exactly equal to zero, for all the values of $\vec{q}$, in the same way as these determinants are zero for a direct summation in Eq. (3) of up to forty conduction bands.

The TA mode in the Δ direction is very sensitive to the details of the calculation. For a change of the parameter E_0 by 25% to a value of 13.6 eV, the squares of all the TA modes are negative, i.e., the crystal becomes unstable against shear. For smaller changes of E_0 by 10% to a value of 11.42 eV, the TA mode in X is positive but the square of the TA mode in the Σ direction becomes negative. If the crystal pseudopotential is kept constant, but the first node of the electron-ion pseudopotential in Eq. (7) is changed by 5%, then the squares of the TA mode become negative again. It is therefore concluded that the self-consistency between crystal and electron-ion pseudopotential is essential in order to get meaningful phonon frequencies.

It should be noted that negative squares of the TA mode are also obtained if the polarizability matrix is calculated by direct summation over the unoccupied states, as in Eq. (3), for up to forty conduction bands.

In summary, the phonon curves presented in this paper are obtained by using the self-consistency between crystal and electron-ion pseudopotential, and closure over all the conduction states via a moment expansion of the polarizability. The results are sensitive to the choice of the pseudopotential as well as the value of the parameter used for the first moment.

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Effective Renormalization Behavior of Ag Films Backed by Superconducting Pb

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When the superconducting half of a superconductor–normal-metal film sandwich is a
strong-coupling superconductor, phonon structure due to the superconductor (Pb)—but
observed by tunneling into the normal metal (Ag)—provides a direct probe of the effec-
tive renormalization interaction in the normal metal. Unexpectedly, even fairly thick
Ag films (≈ 350 Å) backed by Pb exhibit effective renormalizations approaching that of Pb,
both in overall strength and energy dependence.

When the mean free path (mfp) l_N in a normal-
state (N) metal becomes comparable to the film
thickness d_N, thin metal films N backed with a
strong-coupling superconductor S yield ac tunnel-
ing characteristics containing phonon structure
attributable to S.[1-4] Here, N denotes a metal
which remains normal in bulk form, but may be-
come superconductive in film form because of
the proximity effect (PE). The energy-dependent
amplitude of such structure—reckoned relative to
that observed when N is absent—oscillates with
d_N and with energy ω roughly as $\exp[i\varphi(\omega)]$, where
the complex phase

$$\varphi(\omega) \simeq 2\pi[2d_N Z_N(\omega)/h v_{FN}{}^0]\epsilon_N(\omega) \tag{1}$$

depends on the bare band velocity $v_{FN}{}^0$, effective
renormalization $Z_N(\omega)$, and induced energy gap
$\Delta_N(\omega) = (\omega^2 - \epsilon_N{}^2)^{1/2}$. As Rowell and McMillan[1]
point out in their original study of Ag-Pb, such
structure stems from Andreev reflections[5] (elec-
tron $\rightleftharpoons$ hole) at the N-S boundary, and should per-
mit direct determination of the dressed Fermi
velocity $v_{FN}{}^* = v_{FN}{}^0/Z_N(\omega)$ from observed phases.
Since their preliminary estimate[1] $v_{FN}{}^* = 1.1 \pm 0.2$
(Ref. 6) falls near the free-electron value $v_F{}^0$(Ag)
$= 1.39$,[6] one might infer that such experiments ba-
sically measure the intrinsic (bulk) electron-
phonon renormalization in N.

Our more extensive measurements, acquired

with thinner Ag films, suggest that the PE can
cause substantial and progressive enhancements
of effective renormalizations as d_N(Ag) decreases
from 1 000 to 100 Å. Both the size and energy de-
pendence of Re[$Z_N(\omega)$] observed with our two thin-
nest films suggest behavior approaching that of
the Pb backing ($d_S = 4 000 \pm 500$ Å). These results
run counter to current PE theories such as those
of McMillan[7] and Arnold,[8] which predict nearly
bulk behavior even as d_N approaches zero. De-
spite this difficulty, we will continue to describe
our results in terms of effective renormaliza-
tions, leaving open the possibility that other fac-
tors may actually be at work.

All junctions studied utilized Al counterelec-
trodes (CE), deposited on glass substrates (T
$\simeq 300$ K), and natural oxide barriers grown by ex-
posure to air. Pressures remained in the mid-
10^{-7}-Torr range and little time elapsed (~ 20 sec)
between Ag and Pb evaporations. Formvar var-
nish prevented tunneling from all film edges.
Final film thicknesses were determined by Tolan-
sky interferometry.

Wolfram's model theory[9] for quasiparticle in-
terference (QPI) effects in M_1-M_2 sandwiches
provides the basic framework for interpreting
our observations. The theory is founded on exact
solution of Gor'kov's equations for two spatially
uniform energy gaps $\Delta_{1,2} = \Delta_{S,N}(\omega)$ which may be-