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Isotope scattering of phonons in superlattices

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The elastic scattering rate of acoustic phonons in periodic superlattices due to isotopic atoms is calculated in the isotropic, continuum approximation. Unlike the case in bulk solids, the scattering rate depends on both the propagation direction and mode of phonons. Numerical examples are given for AlAs/GaAs and Al/W superlattices for propagation along the normal to the layer interfaces. We find that the scattering of longitudinal phonons is much weaker than that of transverse phonons for wide ranges of frequencies.

\[ H_I = \int d^3x \Delta \rho(x)\left[\mathbf{P}(x)\right]^2/2[\rho_0(z)]^2, \]

where \( \mathbf{P}(x) \) is the momentum operator canonically conjugate to the displacement field of the phonons, \( \rho_0 \) either represents the discontinuous mass density distribution with a periodic parapet profile along the growth direction \( z \) (the interfaces are parallel to the \( x \) plane) or takes the averaged mass density in each constituent layer, and \( \Delta \rho \) is the mass density fluctuation around \( \rho_0 \). In a superlattice consisting of two neighboring layers \( A \) (thickness \( d_A \)) and \( B \) (thickness \( d_B \)), \( \mathbf{P} \) takes the form

\[ \mathbf{P}(x) = -i \sum_\lambda \left[ \frac{\hbar \omega_\lambda \rho_0(z)}{2NS} \right] (c_\lambda - c_\lambda^\dagger) W_\lambda(z)e^{i\mathbf{q}_\lambda \cdot \mathbf{x}}, \]

where \( \mathbf{q}_\lambda \) is the momentum operator canonically conjugate to the mass density \( \rho_0 \) in \( \lambda \)-th layer, and \( W_\lambda(z) = \sum_n \mathbf{e}_{\lambda,n}^A \mathbf{e}_{\lambda,n}^B e^{i\mathbf{q}_\lambda \cdot \mathbf{x}} \).
is the averaged mass density of the A (B) layer, $\Theta$ is the unit
step function, $t_{\lambda,i}$ and $r_{\lambda,i}$ are the transmission and reflection
coefficients, $e_{\lambda,i}$ and $\tilde{e}_{\lambda,i}$ are the unit-polarization vectors,
and $k'_{\lambda,i}(I = A, B)$ is the wave number in a layer along the
growth direction. It should be remarked here that the wave
vector $Q$ and angular frequency $\omega_\lambda$ define the superlattice
phonon dispersion, whereas the wave vector $K_{\lambda,i}^I = (q_i, k'_{\lambda,i})$
and $\omega_\lambda$ for $I = A$ and $B$ define the bulk phonon dispersion
relations of the constituent materials. Equation (2) satisfies
Bloch’s theorem required for a periodic system. The magnitudes
of $t_{\lambda,i}$ and $r_{\lambda,i}$ are normalized according to
$$ f_0^D |u_{\lambda}(z)|^2dz = D \quad \text{or, more generally,} $$

$$ \int_0^D [u_{\lambda,-q,j,m}(z)]^* u_{\lambda,-q,j,m}(z)dz = D \delta_{j,j'} \delta_{m,m}. $$

(7)

With these equations the interaction Hamiltonian becomes

$$ H_I = \frac{\hbar}{4L_{\lambda,i}} \sum_{\lambda,i} (\omega_\lambda \omega_\lambda)^{1/2} C^{1/2} \Phi_{\lambda,\lambda', -q_i + q_j + \text{H.c.}}, $$

(8)

where $L = ND$ is the length of the superlattice,

$$ \Phi_{\lambda,\lambda', -q_i} = \int dz \frac{\Delta \rho_\lambda(z)}{\rho(z)} \mathbf{W}_\lambda(z) \cdot \mathbf{W}_{\lambda', z}, $$

and $\Delta \rho_\lambda$ is defined by

$$ \Delta \rho(x) = \sum_k \Delta \rho_k(z) e^{ik_\lambda \cdot x} $$

or

$$ \Delta \rho_k(z) = \frac{1}{S} \int d^2x \Delta \rho(x) e^{-ik_\lambda \cdot x}. $$

(9)

Applying the golden rule for the transition rates, we obtain
the following equation for the deviation $\Delta n_\lambda$ of the phonon
occupation number from the thermal equilibrium value:

$$ \Delta n_\lambda = \frac{1}{\rho_\lambda} \delta_n \omega_\lambda^2 \sum_{\lambda'} \delta(\omega_\lambda - \omega_\lambda') \Phi_{\lambda,\lambda', -q_i + q_j + q_i} |^2. $$

(10)

Now we take the configuration average over mass density fluctuations

$$ \langle \Delta \rho(x, z + nD) \Delta \rho(x', z' + n'D) \rangle $$

$$ = \frac{[\Delta M_I]^2}{V_0^I} \partial_{n,n'} \delta(x - x') \delta(z - z'), $$

(13)

where $I = A$ if $0 < z < d_A$, $I = B$ if $d_A < z < D$, and $V_0^A$ and $V_0^B$
are the unit-cell volumes of $A$ and $B$ materials. In Eq. (13)

$$ [\Delta M_I]^2 = \sum_{I = A,B} f_{I,i}, (M_{I,i} - \bar{M}_I)^2, \quad I = A,B, $$

measures the mass fluctuation due to the isotope distributions,
where $n_I$ is the number of isotopes, $f_{I,i}$ and $M_{I,i}$ are
the fraction and mass of $I$ material due to $i$th isotope, and $\bar{M}_I$
the average mass, i.e., $M_I = \rho_0 V_0$. With the above averaging procedure we have

$$ \langle \Phi_{\lambda,\lambda', -q_i + q_j} \rangle^2 = \frac{N}{S} \int dA \mathbf{B}_\lambda(z) \cdot \mathbf{B}_{\lambda', z} |^2dz $$

$$ + \int dA \mathbf{B}_{\lambda}(z) \cdot \mathbf{B}_{\lambda}(z) |^2dz, $$

(15)

where $g_i = [\Delta M_I]^2/(\bar{M}_I)^2$.

For a numerical calculation we consider the AlAs/GaAs and
Al/W superlattices, whose phonon band structures have been studied extensively. Specifically, the Al/W superlattice exhibits large-frequency gaps in its phonon dispersion relations
due to large acoustic mismatch between the constituent materials (see Table I). Although aluminum ($\bar{M}_{Al} = 26.98$
amu) and arsenic ($\bar{M}_{As} = 74.92$ amu) are isotopically pure, natural gallium consists of two isotopes $^{69}$Ga and $^{71}$Ga (the
relative abundances are 0.602 and 0.398, $\bar{M}_{Ga} = 69.72$ amu,
and $g_{Ga} = 1.97 \times 10^{-4}$). Hence the mass-difference scattering in the GaAs superlattice occurs only in the GaAs layers with
$g_{GaAs} = g_{Ga}(\bar{M}_{Ga})^2/(\bar{M}_{Ga} + \bar{M}_{As})^2 = 4.58 \times 10^{-5}$. A similar situation holds for the Al/W superlattice, where tungsten
consists of five isotopes ($\bar{M}_{W} = 183.8$ amu and $g_{W} = 7.01 \times 10^{-5}$). Thus the following formula is applied to calculate numerically the isotope scattering rate in these superlattices with A the GaAs or tungsten:

$$ \tau_{\lambda}^{-1} = \frac{\nu_A^B g_{AlW} \omega_\lambda^2}{16 \pi^2 L} \int dJ dJ' dJ'' dJ''' \mathbf{A}_\lambda(z) \cdot \mathbf{A}_{\lambda'}(z) |^2dz, $$

(16)

with $\nu_\lambda = |d\omega_\lambda/dq_z|$, or, more explicitly,

$$ \tau_{\lambda}^{-1} = \frac{\nu_A^B dA}{4 \pi D^3} g_A v_A I_A(\omega_\lambda), $$

(17)

where

<table>
<thead>
<tr>
<th>$I$</th>
<th>$\rho_0$ (cm$^{-3}$)</th>
<th>$v_l$ (10$^5$cm/s)</th>
<th>$v_t$ (10$^5$cm/s)</th>
<th>$g_I$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al</td>
<td>2.73</td>
<td>6.25</td>
<td>3.21</td>
<td>0.0</td>
</tr>
<tr>
<td>W</td>
<td>19.32</td>
<td>5.21</td>
<td>2.88</td>
<td>7.01 $\times$ 10$^{-5}$</td>
</tr>
<tr>
<td>GaAs</td>
<td>5.32</td>
<td>5.03</td>
<td>3.03</td>
<td>4.58 $\times$ 10$^{-5}$</td>
</tr>
<tr>
<td>AlAs</td>
<td>3.76</td>
<td>5.98</td>
<td>3.60</td>
<td>0.0</td>
</tr>
</tbody>
</table>

TABLE I. Employed numerical values of the averaged mass density $\rho_0$, bulk longitudinal and transverse sound velocities $v_l$ and $v_t$, and $g_I$ measuring the mass-density fluctuation for each material. For GaAs the value of $g_{Ga}(\bar{M}_{Ga})^2/(\bar{M}_{Ga} + \bar{M}_{As})^2$ is given in the column of $g_I$. |
The simultaneous bands of the longitudinal (L) and transverse (T) modes are filled in black. In the above equations the numerical values of the sound velocities used are 5.03 × 10^5 and 5.98 × 10^5 cm s⁻¹ (L modes) and 3.03 × 10^5 and 3.60 × 10^5 cm s⁻¹ (T modes) for GaAs and AlAs, respectively, and the mass densities are 5.32 g cm⁻³ for GaAs and 3.76 g cm⁻³ for AlAs. \( v_0 \) is the transverse sound velocity (3.03 × 10^5 cm s⁻¹) in GaAs.

\[
I_A(\omega_\lambda) = \left( \frac{\omega_\lambda D}{v_0} \right)^2 \times \sum_{j'} \int_{a_{\omega_\lambda}}^{a_{\omega_\lambda+1}} \frac{d^2(q_j'|D)}{2\pi} \left| \frac{d(\omega_\lambda, D/v_0)}{d(q_j'|D)} \right|^{-1} J^A_{\lambda,\lambda'} \tag{18}
\]

and

\[
J^A_{\lambda,\lambda'} = \sum_{ijmm} a_{\lambda,i}^* a_{\lambda',j} a_{\lambda,n} a_{\lambda',m} \left( e_{\lambda,i}^A * e_{\lambda',j}^A \right) \left( e_{\lambda,m}^A * e_{\lambda',n}^A \right)
\]

\[
\times \frac{\exp[i(k_{\lambda,i}^A - k_{\lambda,j}^A - k_{\lambda,m}^A + k_{\lambda,n}^A)d_{ij}^A] - 1}{i(k_{\lambda,i}^A - k_{\lambda,j}^A - k_{\lambda,m}^A + k_{\lambda,n}^A)d_{ij}^A}. \tag{19}
\]

In the above equations \( I_A(\omega_\lambda) \) is a dimensionless quantity, \( a_{\lambda,i} \) and \( a_{\lambda,i+3}(i=1-3) \) stand for the transmission and reflection coefficients \( t_{\lambda,i}^A \) and \( r_{\lambda,i}^A \), respectively, \( e_{\lambda,i}^A \), \( k_{\lambda,i}^A = -k_{\lambda,-i}^A \), and \( v_0 \) is a velocity used for the normalization. If the materials \( A \) and \( B \) are the same, Eq. (17) reduces to 1/2 of the isotopic scattering rate of phonons in the bulk \( A \) material, i.e., \( \tau_{\text{bulk}} = V_A^A v_A^A \omega^2 / 4 \pi(v_A^3) \), where \( v_A^A \) is the sound velocity and \( \omega \) means the average over three bulk phonon modes.

Figures 1(a) and 1(b) depict the band structures of both the sagittal (SG) and pure transverse SH phonons in the AlAs/GaAs superlattice with \( d_{\text{AlAs}} = d_{\text{GaAs}} \). Due to the small acoustic mismatch between AlAs and GaAs, the frequency gaps are rather small. It should be noted that the lowest band edges of both the SG and SH modes, i.e., the edges of transverse phonons, coincide with each other.

Figure 2 also shows the band structure of the AI/W superlattice with \( d_{\text{Al}} = d_{\text{W}} \). For the Al/W superlattice the frequency gaps are much wider than those of the AlAs/GaAs superlattice, so we have plotted the frequency bands of the SG and SH modes simultaneously. An interesting feature of the band structure of the Al/W superlattice is the fact that there exists an allowed frequency band below the lowest SH band, though no such frequency band is present for the AlAs/GaAs superlattice. This extra band in the Al/W superlattice (surrounded by the solid lines in Fig. 2) stems from the phonons localized at the interfaces between the tungsten and aluminum, i.e., \( k_{\lambda,i}^A \) and \( k_{\lambda,j}^A \) are complex numbers. Owing to the mutual overlapping of the localized lattice displacements along the growth direction due to the finite thicknesses of the constituent layers, these phonons are extended over the superlattice and forms a frequency band. In the following we call this frequency band the Stoneley (ST) band. This is also the origin of the enhanced phonon density of states (DOS) in the Al/W system at low frequencies, which can be seen below. Note that the phonons localized at the layer interfaces are reduced to the Stoneley interface phonons when the layer thicknesses become infinite, which is characteristic of the special combinations of the elastic constants and mass densities of the constituent materials. No such localized mode exists at the interfaces between bulk GaAs and AlAs.
The DOS of superlattice phonons

\[ G(\omega) = \sum_{\lambda} \delta(\omega - \omega_{\lambda}), \]

which reflects the band structures together with those of bulk materials, are plotted in Figs. 3 and 4 for the AlAs/GaAs and Al/W superlattices, respectively. At low frequencies the DOS grows in proportion to \( \omega^2 \) as in the bulk materials and for the AlAs/GaAs system the magnitude takes almost the averaged value of the DOS's of bulk AlAs and GaAs (circles). This means that the magnitude of DOS in the low-frequency region of the AlAs/GaAs superlattice is consistent with the one estimated from the formula for the bulk phonons with the mass and elastic constants averaged over GaAs and AlAs. However, for the Al/W superlattice the magnitude of DOS at low frequencies is much larger than those of the bulk aluminum (squares) and tungsten (circles). This enhanced DOS of the Al/W superlattice is due to the existence of the extra ST band below the lowest band of the transverse phonons as stated above.

At high frequencies the situation is somewhat different. In particular, we see in Fig. 4 for the Al/W superlattice that the DOS increases linearly with \( \omega \) in certain frequency ranges and also exhibits various structures. (The frequencies at which singularities are found in the DOS correspond well to the local maxima and minima of the frequency bands shown in Fig. 2.) This linear behavior of the DOS reflects the localized (low-dimensional) nature of the phonons in the superlattice, which is shown as narrow lines in the band structures of Fig. 2. Quite unlike the DOS in the Al/W superlattice, the DOS in the AlAs/GaAs superlattice exhibits almost \( \omega^2 \) behavior below \( \omega D/v_0 < 3.5 \) except for the tiny structures around \( \omega D/v_0 = 3.5 \) and 10.

In Fig. 5 we have plotted the isotope scattering rates of \( L \) and \( T \) phonons propagating along the growth direction of the AlAs/GaAs superlattice together with the corresponding phonon dispersion relations. (The scattering does not exist in the frequency gaps where phonon propagation is prohibited.) Due to the structural anisotropy of the superlattice, the scattering rate depends on both the phonon mode and propagation direction. For comparison, the isotope scattering rate (proportional to \( \omega^4 \)) in bulk GaAs, which is independent of the phonon polarization and propagation direction, is represented by the dotted line. As expected, the scattering rates are smaller than that in the bulk GaAs because the scattering does not occur in the AlAs layers. We also see that the scattering rate of the \( L \) phonons is smaller than that of the \( T \) phonons. This can be understood as follows. For phonons in a frequency band the propagation angles in each layer measured from the growth direction are allowed from 0° to 90° for the \( L \) mode but 0° to some angle smaller than 90° for the \( T \) mode. Hence the inner products of the polarization vectors between the initial (propagating at 0°) and a scattered sagittal phonons [see Eq. (16)] extends from 1 to 0 for the \( L \) mode but from 1 to some finite value for the \( T \) mode, leading to the enhanced scattering for the transverse phonons.

The similarly enhanced scattering rate of \( T \) phonon can be seen in Fig. 6 for the Al/W superlattice at the low-frequency
region below the lowest-frequency gap. However, the reason for the suppressed $L$ phonon scattering rate is somewhat different from that for the AlAs/GaAs superlattice. The $L$ phonon in this superlattice is scattered predominantly into the ST phonons, but the $T$ phonon is scattered into both the SG and SH phonons, which have larger DOS than that of $T$ phonons. This is also the reason that we cannot see discernible structures in the scattering rate of $L$ phonon at about $\omega D/v_0 = 2.4$. At higher frequencies very complicated band structures of phonons in the Al/W superlattice makes it hard to tell which scattering rate is larger. Also we note that the scattering rate of low-frequency $T$ phonons is close to the scattering rate of phonons in bulk tungsten. This enhancement is again due to the existence of the ST band below the SH band.

Here we should note that the Al/W superlattice is metallic and hence the electron-phonon scattering should dominate the isotope scattering of phonons. The relaxation times of phonons in the bulk tungsten and aluminum due to electron scattering have been reported at frequencies higher than 0.1 GHz where the attenuation depends linearly on the frequency. In this regime the longitudinal phonon relaxation rate in the [001] direction is $\tau_{el}^{-1}_{\text{ph},L} = \alpha_1 v$, where $\alpha_1 = 3.3 \times 10^{-3}$ for tungsten$^{10,11}$ and $\alpha_{Al} = 2.7 \times 10^{-2}$ for aluminum.$^{12}$ An enhanced attenuation in a metallic superlattice has also been reported.$^{13}$ These results indeed indicate much larger phonon attenuation due to the scattering with electrons in the gigahertz frequency range. The reason that we have given the numerical calculation for the Al/W superlattice is, as remarked repeatedly, to emphasize the existence of the ST band and its effect on the isotope scattering of phonons. Such a ST band and the associated enhancement of the isotope scattering should be also found in nonmetallic (possibly strained) superlattices with a large acoustic mismatch.

To summarize, we have derived a formula for the isotope scattering of phonons in superlattices and also given numerical results for the AlAs/GaAs and Al/W superlattices. For phonons propagating normal to the layer interfaces, the scattering of the longitudinal phonons in the AlAs/GaAs superlattice (also at the low-frequency region of the Al/W superlattice) is smaller than that of the transverse phonons by a factor of 2 or more. Thus, as far as the lattice thermal conductivity in the elastic scattering dominated regime is concerned, we expect that the longitudinal phonons play a more important role in superlattices than in bulk materials.

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