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The dynamic process of non-equilibrium phonons irradiated by laser pulse

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Abstract. An attempt has been made to carry out an overall calculation of the various damping rates of phonons in order to highlight the dynamic picture of non-equilibrium phonons arising in a metal film excited by laser. The calculations are compared with the experimental results and found to be consistent.

1. Introduction

Since Bron and Grill [1] investigated the evolution of the phonon pulse with a phonon spectrometer, studies of the dynamics of non-equilibrium phonon in films excited by a laser have received wide attention. Experimentally, it was pointed out [2–4] that, while the laser beam irradiates the film surface, it is first absorbed by electrons and makes their temperature much higher than that of the lattice. The electrons thermalize rapidly and then cool by transferring energy to the lattice via electron–phonon scattering. Theoretically, Klemens [5] calculated the lifetime of phonon in terms of the Grüneisen parameter. Recently, calculations of the phonon lifetime in crystals of cubic symmetry have been reported [6–8]. They are all based on non-linear elasticity theory including anisotropy, with dispersion neglected. Calculation results show that the lifetime is proportional to the inverse fifth power of phonon frequency. These calculations can be used to analyse the intrinsic damping of phonons governed by the spontaneous decay via cubic anharmonicity, especially for the damping in different propagation directions. However, we find that only a few attempts have been made so far to calculate the decay of high-frequency phonons due to electron and phonon interaction. Based on the generalized Kadanoff–Baym equation of the non-equilibrium Green function [12], this paper presents an attempt to carry out an overall calculation of various damping rates of phonon in order to highlight the dynamic picture of non-equilibrium phonons arising in a metal film excited by a laser. Making use of the experimental parameters in [1], we compare our calculation with the experimental results and find it to be consistent.

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2. Phonon Boltzmann equation

As can be seen from the previous experimental work [4, 10] a laser pulse of picosecond duration created a non-equilibrium conduction electron distribution. Then, during the relaxation processes of these excited electrons, non-equilibrium phonons with high frequencies were observed to be created as a result of the non-radiative transition of electrons. In this section, we shall investigate the dynamics of these non-equilibrium phonons after excitation by the laser pulse. Obviously, it is a system of electron–phonon interaction and the Boltzmann equations describe the dynamics of electrons and phonons coupling with each other. To simplify the problem, we may note that the electron–electron scattering time is expected to be about 2.1×10^{-13} s [9] in metal Cu at low temperatures. On the other hand, the theoretical calculations [11] indicated that the electron–phonon scattering times in Cu, Mg and Au are in the range $(2\text{–}10) \times 10^{-10}$ s ($T = 2$ K). This implies that, during the period of interaction between an electron and a phonon, the interactions between electrons will happen thousands of times. In other words, the electron system seems to be always in an equilibrium state corresponding to the dynamics of interaction system between electrons and phonons. Thus, we shall treat the electron system as a heat bath without internal interaction, and the dynamics of the interaction system are determined by the phonon system including the interaction between electrons and phonons. Under this assumption, the coupled set of transport equations can be reduced to a single-phonon Boltzmann equation.

The pulse length used in the experiment [1] is about 10^{-8} s which is longer than the time scale of interest. So we can assume that the external disturbance varies slowly in space and time. According to the Kadanoff–Baym [12] equation, we have

$$(\partial/\partial T + \nabla_q \omega \cdot \nabla R) D_{jj}^{<}(\boldsymbol{q}, \omega; \boldsymbol{R}, T) = -D_{jj}^{<}(\boldsymbol{q}, \omega; \boldsymbol{R}, T)\Pi^{>}(\boldsymbol{q}, \omega; \boldsymbol{R}, T) + D_{jj}^{>}(\boldsymbol{q}, \omega; \boldsymbol{R}, T)\Pi^{<}(\boldsymbol{q}, \omega; \boldsymbol{R}, T) \tag{2.1}$$

where $D_{jj}^{<}$ and $D_{jj}^{>}$ are the correlation functions of the jth branch phonon with wavevector $\boldsymbol{q}$ and frequency ω, and $\boldsymbol{R}$ and T are the macroscopic variables in space and time. The self-energies $\Pi^{>}$ and $\Pi^{<}$ of phonons come from two terms. If we consider only the first-order approximation, one of these results from the contribution Π_e from the electron–phonon interaction, and the other is due to the contribution Π_{ph} from the phonon–phonon interaction:

$$\Pi^{\gtrless} = \Pi_e^{\gtrless} + \Pi_{ph}^{\gtrless}. \tag{2.2}$$

If we make use of the spectral function $A_{jj}(\omega)$, the correlation function of phonon can be described as

$$D_{jj}^{<}(\boldsymbol{q}, \omega; \boldsymbol{R}, T) = A_{jj}(\omega)N(\boldsymbol{q}, \omega; \boldsymbol{R}, T)$$
$$D_{jj}^{>}(\boldsymbol{q}, \omega; \boldsymbol{R}, T) = A_{jj}(\omega)[1 + N(\boldsymbol{q}, \omega; \boldsymbol{R}, T)] \tag{2.3}$$

where $N(\boldsymbol{q}, \omega; \boldsymbol{R}, T)$ is the distribution function of phonons.

Kadanoff and Baym [12] assumed that, in the case of weak non-equilibrium, the spectral functions have a δ-like character:

$$A_{jj}(\omega) = 2\pi\delta(\omega - \omega_{jq}). \tag{2.4}$$

Substituting equation (2.3) and (2.4) into equation (2.1) and integrating the equation over ω from 0 to ∞, we obtain

$$(\partial/\partial T + \nabla_q \omega \cdot \nabla R) N_j(\omega) = -\Pi^{>}(\boldsymbol{q}, \omega_{jq}; \boldsymbol{R}, T)N_j(\omega) + \Pi^{<}(\boldsymbol{q}, \omega_{jq}; \boldsymbol{R}, T)[1 + N_j(\omega)] \tag{2.5}$$

where $N_j(\omega) = N(\boldsymbol{q}, \omega_{jq}; \boldsymbol{R}, T)$.

In the first-order approximations, the self-energies $\Pi_e^>$ and $\Pi_e^<$ take the following form [13]:

$$\Pi_e^>(\boldsymbol{q}, \omega_{jq}; \boldsymbol{R}, T) = \Pi_e^<(-\boldsymbol{q}, -\omega_{jq}; \boldsymbol{R}, T) = 2\int \frac{\mathrm{d}\boldsymbol{k}\,\mathrm{d}E}{(2\pi)^4} |V_q|^2$$
$$\times G^>(\boldsymbol{k}+\boldsymbol{q}, E+\omega_{jq}; \boldsymbol{R}, T) G^<(\boldsymbol{k}, E; \boldsymbol{R}, T) \tag{2.6}$$

in which V_q is the electron-phonon interaction matrix element, $G^>$ and $G^<$ are the correlation functions of the electron with the wavevector $\boldsymbol{k}$ and energy E. In the situation discussed above, they can be written as

$$G^<(\boldsymbol{k}, E; \boldsymbol{R}, T) = 2\pi\delta(E - E_k) f(\boldsymbol{k}, E; \boldsymbol{R}, T)$$
$$G^>(\boldsymbol{k}, E; \boldsymbol{R}, T) = 2\pi\delta(E - E_k)[1 - f(\boldsymbol{k}, E; \boldsymbol{R}, T)] \tag{2.7}$$

with $f(\boldsymbol{k}, E; \boldsymbol{R}, T)$ the distribution function of the electrons.

If we consider only the lowest-order approximation of phonon decay in three-phonon anharmonic process due to cubic anharmonicity, the self-energies $\Pi_{ph}^>$ and $\Pi_{ph}^<$ can be written as [14]

$$\Pi_{ph}^{\gtrless}(\boldsymbol{q}, \omega_{jq}; \boldsymbol{R}, T) = \sum_{j_1 j_2} \int \frac{\mathrm{d}\boldsymbol{q}_2\,\mathrm{d}\omega_2}{(2\pi)^4} |V_3(-\boldsymbol{q}j, \boldsymbol{q}_1 j_1, \boldsymbol{q}_2 j_2)|^2$$
$$\times D_{j_1 j_1}^{\gtrless}(\boldsymbol{q}-\boldsymbol{q}_2, \omega_{jq}-\omega_2; \boldsymbol{R}, T) D_{j_2 j_2}^{\gtrless}(\boldsymbol{q}_2, \omega_2; \boldsymbol{R}, T) \tag{2.8}$$

in which $V_3(-\boldsymbol{q}j, \boldsymbol{q}_1 j_1, \boldsymbol{q}_2 j_2)$ are the three-phonon interaction matrix elements. In terms of equations (2.3), (2.7), (2.6) and (2.8) and after some integration, we obtain the phonon Boltzmann equation

$$(\partial/\partial T + \nabla_q \omega \cdot \nabla \boldsymbol{R}) N_j(\omega) = 4\pi \int \frac{\mathrm{d}\boldsymbol{k}}{(2\pi)^3} |V_q|^2 \delta(E_k - E_{k-q} - \omega_q)$$
$$\times \{[1 - f(E_k)] f(E_{k-q}) N_j(\omega) - f(E_k)[1 - f(E_{k-q})][1 + N_j(\omega)]\}$$
$$+ 2\pi \sum_{j_1 j_2} \int \frac{\mathrm{d}\boldsymbol{q}_1}{(2\pi)^3} |V_3(-\boldsymbol{q}j, \boldsymbol{q}_1 j_1, \boldsymbol{q}_2 j_2)|^2 \delta(\omega_{jq} - \omega_{j_1 q_1} - \omega_{j_2 q_2})$$
$$\times \{(1 + N_{j_1}(\omega))(1 + N_{j_2}(\omega)) N_j(\omega) - N_{j_1}(\omega) N_{j_2}(\omega)[1 + N_j(\omega)]\} \tag{2.9}$$

where $f(E_k) = f(\hat{\boldsymbol{k}}, E_k; \boldsymbol{R}, T)$.

3. Numerical calculation and discussion

In order to find out the damping rates, we assume that

$$N_j(\boldsymbol{q}\omega; \boldsymbol{R}, T) = N_j^0(\omega) + \delta N_j \qquad f(\hat{\boldsymbol{k}}, E_k; \boldsymbol{R}, T) = f^0(E_k) + \delta f_k \tag{3.1}$$

where $\delta N_j \equiv \delta N_j(\boldsymbol{q}\omega; \boldsymbol{R}, T)$ and $\delta f_k \equiv \delta f(\boldsymbol{k}E_k; \boldsymbol{R}, T)$. $N_j^0(\omega)$ and $f^0(E_k)$ are the Fermi and Bose–Einstein distribution functions, respectively, $N_{j_1}(\boldsymbol{q}\omega; \boldsymbol{R}, T)$ and $N_{j_2}(\boldsymbol{q}\omega; \boldsymbol{R}, T)$ are in thermal equilibrium (e.g. $N_{j_1}(\boldsymbol{q}\omega; \boldsymbol{R}, T) = N_{j_1}^0(\omega_1)$,

$N_{j_2}(q\omega, R, T) = N^0_{j_2}(\omega_2)$). Substituting equation (3.1) into equation (2.9), if we retain only the linear terms, we have

$$\frac{\partial N_j}{\partial t} + \nabla_q \omega \nabla_R \delta N_j = 4\pi \int \frac{dk}{(2\pi)^3} |V_q|^2 \delta(E_k - E_{k-q} - \omega_q)$$
$$\times \{\delta N_j [f^0(E_k) - f^0(E_{k-q})] - \delta f_{k-q}[N^0_j(\omega) + f^0(E_k)]$$
$$+ \delta f_k [1 + N^0_j(\omega) - f^0(E_{k-q})]\} - 2\pi \sum_{j_1 j_2} \frac{dq_1}{(2\pi)^3} |V_3(-qj, q_1 j_1, q_2 j_2)|^2$$
$$\times \delta(\omega_{jq} - \omega_{j_1 q_1} - \omega_{j_2 q_2})\{\delta N_j [1 + N^0_{j_1}(\omega_1) + N^0_{j_2}(\omega_2)]\}. \tag{3.2}$$

The damping rates of the modes of diffusing particles can be obtained from the isotropic part of the Boltzmann equation with all gradients neglected [15]. The functions δN_j, δf_{k-q} and δf_k are now independent of the momentum direction. Thus, the only angle that enters the collision operator in equation (3.2) is the angle between k and q; we can write

$$\int_0^\infty \frac{k^2\, dk}{2\pi^2} \int_{-\infty}^{\infty} \frac{dq}{(2\pi)^3} \rightarrow \int_{-\infty}^{\infty} \frac{dk}{(2\pi)^3} \int_{-\infty}^{\infty} \frac{dq}{(2\pi)^3}.$$

Consequently, we can make the substitution $k \rightarrow k - q$. Integrating equation (3.2) over the magnitude of the momentum, we have the damping rates of phonons in different processes:

$$\Gamma^1_P(\omega) = 4\pi \int \frac{dk}{(2\pi)^3} |V_q|^2 \delta(E_k - E_{k-q} - \omega_q)[f^0(E_{k-q}) - f^0(E_k)] \tag{3.3}$$

$$\Gamma^1_Q(E) = 4\pi \int \frac{dq}{(2\pi)^3} |V_q|^2 \delta(E_k - E_{k-q} - \omega_q)[1 + N^0(\omega_q) - f^0(E_{k-q})] \tag{3.4}$$

$$\Gamma^2_Q(E) = 4\pi \int \frac{dq}{(2\pi)^3} |V_q|^2 \delta(E_{k+q} - E_k - \omega_q)[N^0(\omega_q) + f^0(E_{k+q})] \tag{3.5}$$

$$\Gamma^2_P(\omega) = 2\pi \sum_{j_1 j_2} \int \frac{dq_1}{(2\pi)^3} |V_3(-qj, q_1 j_1, q_2 j_2)|^2 \delta(\omega_{jq} - \omega_{j_1 q_1} - \omega_{j_2 q_2})$$
$$\times (1 + N^0_{j_1}(\omega_1) + N^0_{j_2}(\omega_2)). \tag{3.6}$$

A diagrammatic representation of each of the damping rates is given in figure 1.

In order to obtain equation (3.5), we have made a substitution of $k - q \rightarrow k$.

If we consider the simple electron–phonon interaction model, the square of the electron–phonon interaction matrix element is given by [16]

$$|V_q|^2 = \delta^2 q^2 / 2\omega_q \rho_m. \tag{3.7}$$

Here, δ is the electron–phonon coupling constant. If we consider only phonons of long wavelength compared with the lattice spacing, the deformation potential model can be used and α takes the value $-\frac{2}{3} E_F$ [17], where E_F is the Fermi energy. ρ_m is the mass density and $\omega_q = V_L q$ where V_L the longitudinal phonon velocity and q the wavevector

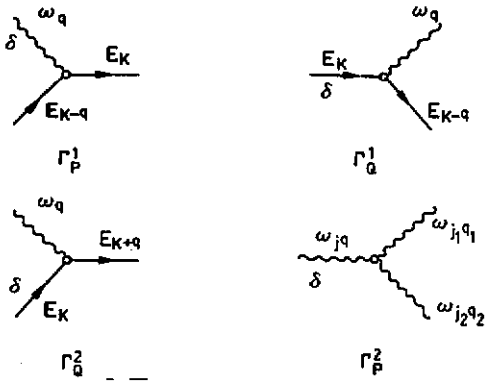

Figure 1. Various decay processes considered in this work: ——, electrons; ∿∿∿, phonons. The mark δ beside the line denotes non-equilibrium occupation probability.

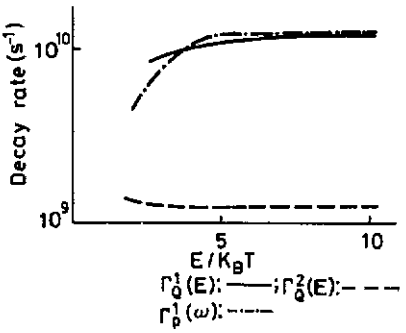

Figure 2. Decay rate at different electron energies E: ——, $\Gamma^1_Q(E)$; – – –, $\Gamma^2_Q(E)$; —·—, $\Gamma^1_P(\omega)$.

of the phonon. We calculate equations (3.3)–(3.5) in terms of equation (3.7). The final results are

$$\Gamma^1_P(\omega) = (\delta^2 m^2 T k_B / 2\pi V_L \rho_m \hbar^4) \ln \tfrac{1}{2}[1 + \exp(E/k_B T)/\{\exp[(E - \hbar\omega)/k_B T] + 1\}] \tag{3.8}$$

$$\Gamma^1_Q(E) = (\delta^2 m / 2\pi V^4_L \rho_m k_{max} \hbar^2)\{\omega^3/3 + 2.4(k_B T/\hbar)^3 - \omega^2(k_B T/\hbar) \times \exp[(-E + \hbar\omega)/k_B T]\} \tag{3.9}$$

$$\Gamma^2_Q(E) = (1.2\delta^2 m / \pi V^4_L \rho_m k_{max} \hbar^2)(k_B T/\hbar)^3[1 + \exp(-E/k_B T)]. \tag{3.10}$$

Klemens [5] calculated the decay rate of the longitudinal phonon based on the perturbative approaches in which the anharmonic interactions are described in terms of the Grüneisen parameter. We use the same form of the coupling coefficient as that used by Klemens. Thus,

$$|V_3(-qj, q_1 j_1, q_2 j_2)|^2 = 4\hbar\gamma^2 a^3 / 3MV^2_L(\omega_{jq}\omega_{j_1 q_1}\omega_{j_2 q_2}) \tag{3.11}$$

in which γ is the Grüneisen parameter, M is the atomic mass and a^3 is the atomic volume. In the isotropic case, three-phonon anharmonic decay will occur only for longitudinal phonons [18], i.e. L → L + T or L → T + T. Hot electrons can also lose energy through the emission of LO phonons. However, Orbach [19] has shown that the occupation number of the acoustic phonons created will be strongly enhanced over the occupation number of $k = 0$ optical phonons in the spontaneous decay of LO phonons into two LA phonons, which is the major process of optical phonon relaxation. The collinear process L → L + L is also possible, but from a final-density-of-states argument the decay rate is small compared with other processes. Hence, we consider here the dominant decay channels of LA phonons, LA → TA + LA and LA → TA + TA.

Substituting equation (3.11) into equation (3.6) and carrying out some calculations, we obtain

$$\Gamma^2_{P_1}(\omega) = \Gamma_{LA\to TA+TA} = (16\pi\hbar r^2 / mV^2_T \omega^3_D)(V_L/V_T)^2 f(\omega, T) \tag{3.12}$$

$$\Gamma^2_{P_2}(\omega) = \Gamma_{LA\to LA+TA} = (8\pi\hbar r^2 / mV^2_T \omega^3_D) f(\omega, T) \tag{3.13}$$

Table 1. Parameters used for the quantitative calculation.

Parameter	V_L	V_T	k_{max}	ρ_m	α	T
Data	5.24 km s^{-1}	2.64 km s^{-1}	1.72×10^{10} m^{-1}	8.80 g cm^{-3}	−7.0 eV	10 K
Source	[1]	[1]	[1]	[1]		[1]

Table 2. The calculation results.

Mode	Γ_P^1	Γ_Q^1	Γ_Q^2	$\Gamma_{P_1}^2$	$\Gamma_{P_2}^2$
Decay rate (s^{-1})	1.20×10^{10}	1.20×10^{10}	1.76×10^{9}	3.49×10^{7}	4.40×10^{6}
Lifetime (s)	8.30×10^{-11}	8.30×10^{-11}	5.68×10^{-10}	2.87×10^{-8}	2.28×10^{-7}

with

$$f(\omega, T) = \omega^5/60 + 2.4(k_B T/\hbar)^3\omega^2 - 12.8(k_B T/\hbar)^4\omega + 24.8(k_B T/\hbar)^5 \qquad (\hbar\omega > k_B T) \tag{3.14}$$

where $\omega = \omega_q$, V_T is the transverse velocity of sound and ω_D is the longitudinal Debye frequency of phonon.

The result is different slightly from that obtained by Klemens [5], because we consider the condition $\hbar\omega > k_B T$, instead of $k_B T > \frac{1}{2}\hbar\omega$ in [5]. In addition, we do not first choose $\omega_{j_1 q_1} = \omega_{j_2 q_2} = \frac{1}{2}\omega_j q$. Under the condition $\hbar\omega \gg k_B T$, only the first term remains in equation (3.14) and $\Gamma_P^2 \propto \omega^5$ which is similar to the results of other workers [5–8]. However, if $\hbar\omega$ is only slightly larger than $k_B T$, this ω^5-dependence of decay rates will break down; this has been verified experimentally [20]. Obviously, we have $V_L > V_T$ (typically $\alpha = (V_L/V_T) \simeq \frac{8}{3}$ [5]), so the process LA → TA + TA dominates over the process LA → LA + TA by a factor of 4.5, which is compatible with the calculation made by Tamura [21].

Quantitative calculations of the damping rates of phonons due to the spontaneous decay were performed for constantan. The parameters needed for calculation are listed in table 1. Figure 2 shows the decay rates for various energies of electrons at $\omega =$ 6.28 THz. In our situation, the results can be fitted to those listed in table 2.

The experimental results of electron–phonon interaction time which were found to be 3×10^{-11} s $< \tau_{ep} < 8 \times 10^{-10}$ s [22] are in agreement with our calculations.

From table 2, we can see that Γ_Q^1/Γ_Q^2 is equal to 6.8. This implies that hot electrons emit phonons seven times faster than equilibrium phonons excite hot electrons to a higher energy level. It can be understood physically that the great mismatch between a hot-electron energy and a phonon energy requires several collisions before the excited electrons reach equilibrium with the lattice. Comparing equation (3.9) with equation (3.10), we can see that this difference appears only under the condition $\hbar\omega > k_B T$. It is the very effect of the interaction between electrons and high-frequency phonons excited by an ultrashort laser pulse. In the circumstance of normal thermal excitation, the frequency of the phonons is about 10^9 Hz; so $\hbar\omega \ll k_B T$ and the decay rates Γ_Q^1 and Γ_Q^2 will have the same form (as can be seen from figure 2, the term $\exp(-E/k_B T)$ can be neglected); then the difference will vanish. We also find that $\Gamma_Q^1/\Gamma_{P_1}^2 \approx 50.4$ in table 2.

This may illustrate that the lifetime of high-frequency phonons will delay to the time of the electron–phonon interaction. In other words, in our case, the phonon thermalization may delay in time to that of the electron–phonon interaction process, because the phonon–phonon collision is responsible for the thermalization of the phonon gas.

It is worth mentioning that there is much research into the non-equilibrium process in semiconductors. Because of different aims and study time scales, these investigations all placed an emphasis on the relaxation of hot electrons. In contrast with them, we study the dynamics of phonons in metal films, which may be useful for investigating some transient behaviour related to the dynamics of non-equilibrium phonons. Zhou Benlian [23] has observed a delay in thermal expansion under rapid heating. It may be explained in terms of the thermalization and propagation of the phonon gas and further work is in progress.

4. Conclusion

In conclusion, we believe that our calculation is helpful in clarifying the picture of the dynamics of non-equilibrium phonons in a metal film excited by a laser and can confirm quantitatively the phenomenological analyses [2, 3].

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