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Relaxation between electrons and surface phonons of a homogeneously photoexcited metal film

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Abstract. The energy relaxation between the hot degenerate electrons of a homogeneously photoexcited metal film and the surface phonons (phonon wave vectors in two dimensions) is considered under Debye approximation. The state of electrons and phonons is described by equilibrium Fermi and Bose functions with different temperatures. Two cases for electron scattering by the metal surface, namely specular and diffuse scattering, are considered.

Keywords. Hot degenerate electron gas; electron surface scattering; electron surface phonon coupling.

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1. Introduction

In nanoscale metallic systems such as island metal films used in microelectronics [1], the phenomenon of hot electron scattering by surface phonons is quite important. One important problem in this field is to calculate the energy transfer between excited hot electrons and the lattice bath. The present paper is devoted to the calculation of energy transfer rate from degenerate hot electrons to surface phonons. We consider the case of a homogeneously photoexcited (no spatial diffusion) nanoscale metal film, in which the electron mean free path is more than the film thickness (as in case of metals, in which even at high temperatures, the electron mean free path is several hundred angstroms). So the electrons will be scattered by the film surface. The excited metal can be thought of as consisting of two subsystems, namely, degenerate electronic subsystem at temperature T_{e} and the surface phonon bath at temperature T ($T_{\mathrm{e}} > T$). The condition for degenerate hot electron subsystem is justified, because, the time required to establish equilibrium in the electron gas (strong electron–electron interactions) is much less than the time for achieving equilibrium between the electrons and the phonons, $\tau_{\mathrm{e-e}} \ll \tau_{\mathrm{e-p}} \gg \tau_{\mathrm{p-p}}$. In other words, the electron–electron and the phonon–phonon relaxation processes are fast enough to maintain the electron and the phonon

distributions in their respective equilibrium conditions, i.e. Fermi–Dirac and Bose distribution functions respectively. Thus, one calculates the energy transfer from the degenerate electron subsystem at the elevated temperature T_{e} to the phonon subsystem at lower temperature T [2]. In the present work we consider the case of electron energy relaxation from higher-lying (energy-wise) electron subsystem to the lower-lying surface phonon subsystem under Debye approximation. It is assumed that electron surface phonon coupling constant is the same as electron bulk phonon coupling constant.

2. Theory

Case 1. The incident and scattered electron wave vectors are in the same plane of incidence.

Consider an electron gas of volume V' bounded by x–y plane. The electrons scatter from the surface and transfer their energy to surface phonon modes. Initial temperature of degenerate electron gas is T_{e} and that of phonon bath is T ($T_{\mathrm{e}} > T$). We calculate the energy transfer rate U_{surface} from electron subsystem to surface phonon subsystem. The equilibrium distributions of electron gas and the surface phonon bath is

$$N_k = 1/\{\exp(\beta_{\mathrm{e}}[\varepsilon - \varepsilon_0]) + 1\}, \quad \beta_{\mathrm{e}} = 1/KT_{\mathrm{e}},$$

$$N_p = 1/\{\exp(\beta\hbar\omega) - 1\}, \quad \beta = 1/KT, \quad T_{\mathrm{e}} > T,$$

K is the Boltzmann constant.

Conservation of energy and momentum gives

$$\begin{aligned} &\varepsilon_{k'} - \varepsilon_k = \hbar\omega, \quad \omega = sf, \\ &k'_{||} - k_{||} = f, \qquad (1) \\ &k' \sin\theta - k \sin\theta' = f, \\ &k' \cos\theta = -k \cos\theta'. \qquad (2) \end{aligned}$$

Consider that the linear dispersion relation holds good for surface phonons, where f is the phonon wave vector and s is the speed of sound at the surface of the metal film. The mentioned process will always happen, as from eq. (1), $\sin\theta \approx f/2k'$ and $f \gg 2ms/\hbar$, which holds good in a metal. The rate of phonon generation can be written as [2]

$$\begin{aligned} \dot{N}_p = \int\!\!\int\!\!\int_{f_{\min}} \alpha\omega[&(N_p + 1)N_{k'}(1 - N_k) \\ &- N_p N_k(1 - N_{k'})]\mathrm{d}^3k \frac{2V'}{(2\pi)^3}\delta(\varepsilon_{k'} - \varepsilon_k - \hbar\omega), \qquad (3) \end{aligned}$$

where $\alpha = \pi U^2/\rho V s^2$ is having the dimension of energy. Here U is the electron phonon interaction constant, ρ is the density of the metal and V is the lattice volume.

Equation (3) will reduce to

$$\dot{N}_p = \left(\frac{\alpha m\omega}{4\pi^2\hbar^2}\right)\left\{\frac{\mathrm{e}^{\beta\hbar\omega} - \mathrm{e}^{\beta_\mathrm{e}\hbar\omega}}{\mathrm{e}^{\beta\hbar\omega} - 1}\right\}\int_{f_{\min}}^{\infty}\frac{\mathrm{e}^{\beta_\mathrm{e}(\varepsilon_{k'}-\varepsilon_0)}}{(\mathrm{e}^{\beta_\mathrm{e}(\varepsilon_{k'}-\varepsilon_k)}+1)^2}\mathrm{d}k'. \tag{4}$$

Expanding the integral near the Fermi surface, the integral in eq. (4) will be

$$\int_{k_l=k_f-\Delta k/2}^{k_u=k_f+\Delta k/s}\left(1+(k'-k_f)\frac{\beta_\mathrm{e}\hbar^2k_f}{m}\right)\Big/\left(2+(k'-k_f)\frac{\beta_\mathrm{e}\hbar^2k_f}{m}\right)^2\mathrm{d}k'.$$

$$\dot{N}_p = \frac{\alpha m^2\omega^2[\ln 5/3 - 4/15]}{4\pi^2\hbar^3k_0}\left\{\frac{\mathrm{e}^{\beta\hbar\omega} - \mathrm{e}^{\beta_\mathrm{e}\hbar\omega}}{(\mathrm{e}^{\beta\hbar\omega}-1)(\mathrm{e}^{\beta_\mathrm{e}\hbar\omega}-1)}\right\}. \tag{5}$$

The energy transferred by the electrons to the surface modes per unit volume per unit time is

$$U_{\text{surface}} = \int \dot{N}_p\hbar\omega\frac{(\text{area of unit cell})}{4\pi^2}2\pi f\,\mathrm{d}f.$$

On comparing with the bulk case [2], we get

$$\frac{U_{\text{surface}}}{U_{\text{bulk}}} = \frac{\pi^{1/3}(\ln 5/3 - 4/15)}{2.3^{1/3}an^{1/3}} = 0.08.$$

For $T \gg T_0$ and $T_\mathrm{e} - T \ll T$, the energy transfer rate will reduce to [2],

$$U_{\text{surface}} = \left[\frac{\pi^{7/3}ms^2n^{2/3}(\ln 5/3 - \ln 4/15)}{3^{4/3}a\tau(T)T}\right]\{T_\mathrm{e} - T\}. \tag{6}$$

Here n is the electron number density, a is the lattice constant and $\tau(T)$ is the electron flight time at temperature T, which is inversely proportional to T. Thus the electron phonon coupling constant (the expression in square brackets) is temperature independent.

Case 2. Diffuse scattering (when the incident and scattered electron waves are not in the same plane of incidence). The scattered phonon can go in any direction in x–y plane (figure 1).

In line with Case 1, conservation of energy and momentum equations are

$$\varepsilon_{k'} - \varepsilon_k = \hbar\omega,$$

$$\frac{\hbar^2}{2m}[k_x'^2 - (k_x' - f_x)^2 + k_y'^2 - (k_y' - f_y)^2] = \hbar sf.$$

Scattered phonon can go in any direction in x–y plane.

The rate of phonon generation is

$$\begin{aligned}\dot{N}_p = \iiint_{f_{\min}} &\alpha\omega[(N_p+1)N_{k'}(1-N_k)\\ &-N_pN_k(1-N_{k'})]k'^2\sin\theta\,\mathrm{d}\theta\,\mathrm{d}k'\,\mathrm{d}\phi\,\frac{4mV'}{(2\pi)^3\hbar^2f}\\ &\times\delta(2k'\sin\theta\cos(\phi-\phi') - f).\end{aligned}$$

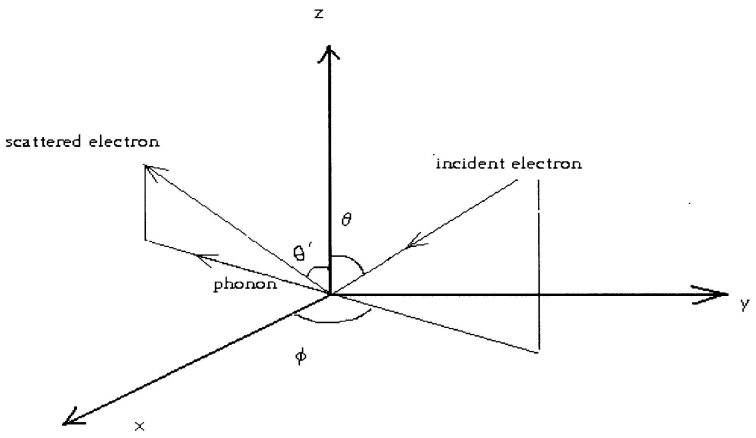

Figure 1. Scattering of an electron from x–y plane. The emitted phonon is confined in the plane of incidence.

$$\dot{N}_p = \frac{\alpha m^2 \omega s V'}{(2\pi)^2 \hbar^3} \left\{ \frac{\mathrm{e}^{\beta\hbar\omega} - \mathrm{e}^{\beta_\mathrm{e}\hbar\omega}}{(\mathrm{e}^{\beta\hbar\omega} - 1)(\mathrm{e}^{\beta_\mathrm{e}\hbar\omega} - 1)} \right\}.$$

When T, $T_\mathrm{e} \ll T_0$,

$$U'_{\mathrm{surface}} = \left(\frac{\pi U^2 m^2}{(2\pi)^3 \hbar^2 \rho a s^3} \right) \left(\frac{KT_0}{\hbar} \right)^4 \left[\frac{T_\mathrm{e}^4 - T^4}{T_0^4} \right] \int_0^\infty \frac{x^3}{\mathrm{e}^x - 1} \mathrm{d}x.$$

For $T \gg T_0$ and $T_\mathrm{e} - T \ll T$, the energy transfer rate will reduce to

$$U'_{\mathrm{surface}} = \left(\frac{\pi m^2 U^2 (KT_0)^4}{3(2\pi)^3 \hbar^6 \rho a s^3} \right) \left(\frac{T_\mathrm{e} - T}{T_0} \right). \tag{7}$$

Now, [2]

$$U_{\mathrm{bulk}} = \left(\frac{m^2 U^2 (KT_0)^5}{2\hbar^7 \rho s^4 (2\pi)^3} \right) \left(\frac{T_\mathrm{e} - T}{T_0} \right).$$

$$\frac{U'_{\mathrm{surface}}}{U_{\mathrm{bulk}}} = \eta \left[\frac{s^5 n^{4/3}}{a\omega_0^5} \right] \cong 0.14.$$

Here ω_0 is the Debye frequency. $\eta = 2^7\pi^3/3$. The calculation is done for gold.

3. Conclusion

The expressions for electron energy loss rate to surface phonons (U_{surface}) are obtained in case of a nano-metric metal film. We found that in low temperature regime ($T, T_\mathrm{e} \ll T_0$), U_{surface} is proportional to the fourth power of the electron temperature, whereas, in the bulk case [2], it is proportional to the fifth power of

electron temperature. The ratio of energy transferred by hot electrons to surface phonons, with that of bulk case is calculated for high temperature regime ($T \gg T_0$ and $T_e - T \ll T$). We show that, in Case 1 where the emitted phonon is confined in the plane of incidence (less freedom), this ratio $\lfloor U_{\text{surface}}/U_{\text{bulk}} \rfloor$ of energy transferred by hot electrons is about 8%. The same ratio is about 14% in Case 2 (more freedom for phonon direction), i.e., the diffuse scattering case.

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