

Estimation of electron temperature in heated metallic nanoparticle

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A method is proposed for determining the temperature of hot electrons in a metallic nanoparticle embedded in a dielectric matrix under ultrashort laser pulses irradiation. The amplitude and power of the longitudinal spherical acoustic oscillations as functions of density and elastic properties of the medium, the laser pulse duration, the electron temperature, radii of particles, and the electron-phonon coupling constant are obtained. The efficiency of the electron energy transfer from heated noble nanoparticles to a surrounding environment is estimated for different electron temperatures.

Key words: *electron temperature, metal nanoparticle, ultrashort laser pulses*

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1. Introduction

Metallic nanoparticles (MNs) are mainly studied due to their unique optical properties [1] and extensive practical applications [2–5]. Over the last decade, the acoustic oscillations of MNs excited by ultrashort laser pulses have been under intense study [1–9]. This is due to the availability of important data relating to the elastic properties of these particles and their mechanical coupling to the surrounding medium, which forms the foundation for the design of elasticity sensors in the nanometer range [10]. The generation of sound waves by MNs was originally observed experimentally for noble nanoparticles [2–4].

When MNs are excited by ultrashort pulses, the energy pulse is initially transferred to the gas of free electrons which collide with one another as well as with the lattice vibrations, and redistribute this energy (being thermalized) over a short time (of the order of tens of picoseconds in a 100 Å particle [3]). The electron gas in the particle is immediately heated. Due to a low heat capacity of the electron gas (compared to the lattice), there originates a short (but strong) electron pressure burst [11]. It is equivalent to a short mechanical impact upon the particle surface over an infinitely small time interval and can generate spherical sound waves in the medium surrounding the particle [12, 13].

In this article, we proposed a method for determining the temperature of hot electron gas in a metallic nanoparticle embedded in a dielectric matrix under ultrashort laser pulses irradiation. To accomplish this, the amplitude and power of the longitudinal sound wave generated by the excess pressure of an electron gas inside the MN driven by ultrashort laser pulses are calculated. So far, this problem has been little studied theoretically.

2. Initial principles and model

Let an ultrashort laser pulse of duration τ_0 be incident on a spherical MN embedded in an infinite, elastically isotropic, dielectric medium. Energy transfer from the laser to the lattice of a nanoparticle depends much on the relationship between the particle sizes, the electron mean free path in the particle, and the Debye length $l_D = \pi v_F / \omega_D$, which plays an important role in the energy exchange between the hot

electrons and the lattice [14]. Its values for several metals are listed below in table 2. We shall consider here the particles with radii $R < l_D/2$.

The additional pressure that develops during heating of the electron gas by the laser pulse can produce oscillations of the nanoparticle surface, which in turn, generate longitudinal acoustic waves in the elastically isotropic space around the particle.

The equation for the propagation of longitudinal acoustic oscillations in this space is given by [15]

$$\nabla^2 \mathbf{u}_L(t) - \frac{1}{s_L^2} \frac{\partial^2}{\partial t^2} \mathbf{u}_L(t) = 0, \quad (2.1)$$

where s_L is the longitudinal sound speed in the medium. Elastic displacement waves \mathbf{u}_L are accompanied by bulk compressions and expansions of the surrounding medium.

As boundary conditions for equation (2.1), we take a condition that all forces applied to the particle surface ($r = R$) are equal to zero. This equality can be presented as [15]

$$\left[\sigma_{rr} - \frac{2\mathcal{E}_s}{R^2} u_r(t) \right] \Big|_{r=R} = -\delta p(t). \quad (2.2)$$

Here, $u_r = |\mathbf{u}_r|$ is the radial component of the displacement, R is the particle radius, \mathcal{E}_s is the surface energy density, $\delta p(t)$ is the time dependent additional pressure of the hot electrons, and σ_{rr} is the stress tensor, whose components are expressed through the moduli K , μ of the uniform compression and rigidity, which one can find elsewhere (e.g., in [15]). For spherical acoustic oscillations, one can assume that $u_L(t) = u_r(t)$.

The pressure of the electron gas on the surface of the nanoparticle can be written as the sum

$$p(t) = p_0 + \delta p(t). \quad (2.3)$$

Here, the first term represents the pressure of a degenerate Fermi gas at $T_e = 0$ K, $p_0 = \frac{2}{5} n \mu_0$, where $n = N/V$ is the electron density, V is the particle volume, and μ_0 is the limiting value of the chemical potential at $T_e = 0$ K ($\mu_0 = \varepsilon_F$, where ε_F is the Fermi energy). The second term, $\delta p(t)$, represents the additional time dependent gas pressure owing to the electron mobility at temperatures $T_e > 0$. It can be written as [16]

$$\delta p(t) = \alpha T_e^2(t), \quad \alpha = \frac{\pi^2}{6} n \frac{k_B^2}{\mu_0}. \quad (2.4)$$

3. Amplitude of the sound wave

To solve equation (2.1) with the boundary condition (2.2), we use the potential method [15] $\mathbf{u}(\mathbf{r}, t) \equiv \nabla \varphi(\mathbf{r}, t)$. Then, representing of function $\varphi(\mathbf{r}, t)$ in the form of spherical waves expanding from particle center $\varphi(\mathbf{r}, t) = \phi(t - (r - R)/s_L)/r$, where ϕ is an arbitrary, twice differentiable function and using the boundary condition (2.2) at the nanoparticle surface, we found for ϕ an inhomogeneous differential equation

$$\frac{\partial^2}{\partial t^2} \phi(t) + 2\gamma \frac{\partial}{\partial t} \phi(t) + \omega_0^2 \phi(t) = -\delta p(t) \frac{R}{\rho}, \quad (3.1)$$

in which ρ is the mass density of the medium surrounding the nanoparticle, and

$$\gamma = \frac{1}{s_L R} \left(2s_T^2 + \frac{\mathcal{E}_s}{\rho R} \right), \quad \omega_0^2 = \frac{2}{R^2} \left(2s_T^2 + \frac{\mathcal{E}_s}{\rho R} \right). \quad (3.2)$$

Here, s_T is a transverse sound speed [15]. The parameters ω_0 and γ are the oscillator eigenfrequency in the absence of friction and the damping decrement, respectively. Note that when a solid medium surrounds the nanoparticle, we must set $\mathcal{E}_s = 0$ in expressions (3.2). However, if the nanoparticle is embedded in a liquid medium, then only the second terms in expressions (3.2) must be retained. The quantity $1/\gamma$ specifies the time over which the oscillations are completely damped. It depends on the nanoparticle radius [17].

Equation (3.1) formally describes the motion of the oscillator under the action of an external driving force of the form $\delta p(t)R/\rho$. We will consider that there are no oscillator eigenmodes without an external force, and that they are generated only by an external force. If the driving force acting on the oscillator, for example, is δ -shaped

$$\delta p(t) = p_{\text{im}}\delta(t - \tau_0), \quad (3.3)$$

then the solution of equation (3.1) with the initial conditions $\phi = 0$ and $\phi' = -Rp_{\text{im}}/\rho$ for $t = \tau_0$ is given by

$$\phi(t) = -e^{-\gamma(t-\tau_0)} \sin[\omega(t - \tau_0)] Rp_{\text{im}}/\rho\omega, \quad (3.4)$$

where $\omega = \sqrt{\omega_0^2 - \gamma_0^2}$, provided that $\omega_0 > \gamma$.

When the time dependence of the function $T_e(t)$ is known, the value of p_{im} can be determined explicitly. Comparing expressions (2.4) and (3.3), we find

$$\alpha \int_0^{\infty} T_e^2(t - \tau_0) dt = p_{\text{im}}. \quad (3.5)$$

The variation of the electron temperature with time can be modelled or determined analytically.

Let us suppose that initially (at $t = \tau_0$) the oscillator is not displaced, i.e., is at rest, and obeys the initial conditions $\phi(\tau_0) = 0$ and $\phi'(\tau_0) = 0$. Then, the solution of equation (3.1) for an arbitrary form of the driving force can be presented in an integral form [18]

$$\phi(t) = -\frac{Re^{-\gamma t}}{\rho\omega} \int_{\tau_0}^t \delta p(\tau) e^{\gamma\tau} \sin[\omega(t - \tau)] d\tau, \quad (3.6)$$

with the conditions that $\delta p(\tau) \neq 0$ and $t > 0$.

Equation (3.6) easily answers the question of how the oscillator behaves after an excess pressure $\delta p(\tau)$ has acted on it over a short time interval (τ_0, τ_1) . The upper limit in equation (3.6) for this case, evidently, will be the quantity τ_1 . Then, using the mean-value theorem in equation (3.6), we obtain

$$\phi(t) = -RIe^{-\gamma t} e^{\gamma\xi\tau_1} \sin(\omega t)/\rho\omega, \quad (3.7)$$

where $\tau_0/\tau_1 < \xi < 1$, while $\int_{\tau_0}^{\tau_1} \delta p(\tau) d\tau \equiv I$ presents the impulse of the excess pressure of the electron gas. If $\omega_0\tau_1\xi \ll 2\pi$, then the explicit form of $\delta p(\tau)$ is not essential; an important point is only the value of I . When $\omega_0 \gg \gamma$, the maximum displacement u_{max} caused by the excess pressure ‘‘impact’’, with using potential method and equation (3.7), will be

$$u_{\text{max}} = I/\rho\omega_0 R. \quad (3.8)$$

Taking into account here equation (3.2) for the case $\mathcal{E}_s = 0$, we obtain

$$u_{\text{max}, \mathcal{E}_s=0} = I/2\sqrt{\mu\rho}, \quad (3.9)$$

i.e., the magnitude of $u_{\text{max}, \mathcal{E}_s=0}$ is independent of the particle radius (and is more independent since the rigidity modulus and mass density of the nanoparticle are smaller). On the other hand, if the $\mathcal{E}_s > 0$, then a dependence on the particle radius shows up, and the magnitude of u_{max} is decreased as R is reduced. The maximum displacement over the time $\tau_1 - \tau_0$ with account for the finite damping, has the form:

$$u_{\text{max}, \mathcal{E}_s=0} = \frac{I(1 - R\gamma/s_L)}{2\sqrt{\mu\rho}\sqrt{1 - (s_T/s_L)^2}} e^{-\gamma(\tau_1 - \tau_0)}. \quad (3.10)$$

Therefore, it is possible to determine the pressure impulse I by measuring the maximum displacement of the physical object.

4. Power radiated by nanoparticle surface

A certain part of the accumulated by MN's energy is carried away from the particle into the surrounding matrix by sound waves. The instantaneous elastic energy flux from the surface of the sphere (for the case of $\mathcal{E}_s = 0$) can be calculated using a formula [19]

$$\mathcal{W}(t) = S \delta p(t) v_r(R, t). \quad (4.1)$$

Here, S is the area of the particle surface and v_r is the radial or vibrational velocity.

In terms of time, conventionally, two stages in this process can be identified: one is an impetuous rise in δp during an ultrashort time spell ($t \sim \tau_0$) and another one is a slower decrease (under $t > \tau_0$) owing to a transfer of the electron energy to the surrounding matrix. The dependence of the $\delta p(t)$ within the second time spell, can be well described by expression (2.4).

In order to take an explicit account of the behavior of the pressure in the initial time spell, the function $\delta p(t)$ can be presented as the product of two functions,

$$\delta p(t) \rightarrow \theta(t - \tau_0) \cdot \alpha T_e^2(t), \quad (4.2)$$

where we use equation (2.4), while $\theta(t - \tau_0)$ is the Heaviside unit-step function that specifies the pressure behavior when $t \rightarrow \tau_0$. Let us also suppose that the electron temperature varies with time in accordance with the rule

$$T_e(t) = T_e(\tau_0) e^{-\beta(t-\tau_0)}, \quad \beta = \frac{g_R}{C_e}, \quad C_e = 3\alpha T_e, \quad (4.3)$$

where $C_e \equiv C_e(T_e(\tau_0))$ is the heat capacity of the electron gas, which depends on the electron temperature taken at the instant of time $t = \tau_0$. The electron-phonon coupling constant in equation (4.3) can be computed from the formula

$$g_R = \frac{27}{16} \frac{n}{m} \frac{\alpha}{k_B} \frac{1}{\rho_p R} \left(\frac{\pi \hbar}{a} \right)^3 \left(\frac{U_1}{A} \right)^2. \quad (4.4)$$

Here, a is the lattice constant for the MN, ρ_p denotes the mass density of the MN, U_1 is the energy required to detach the first electron from the neutral unexcited atom, and A refers to the electron work function of the metal. Formula (4.4) follows from equation (115) of the work [20] using the equation (2.4) and the following relationships

$$\omega_D \approx \sqrt{\frac{\sigma}{\rho_p} \left(\frac{\pi}{a} \right)^3}, \quad v_F = \frac{\hbar}{m} (3n\pi^2)^{1/3}, \quad (4.5)$$

where ω_D is the Debye frequency and σ is the surface energy density.

Taking into account equations (4.2) and (4.3), after integrating with respect to τ within limits $0 < \tau < t$ and some transformations, we finally obtain

$$\begin{aligned} \mathcal{W}(t) = & \frac{4\pi R^2}{\rho_{sL}} \frac{\delta p(t) \delta p_m(t)}{\omega_0^2 + 4\beta(\beta - \gamma)} \left(-2\beta \left(\frac{s_L}{R} - 2\beta \right) + e^{(2\beta - \gamma)t_0} \left\{ \left[\omega_0^2 + 2\beta \left(\frac{s_L}{R} - 2\gamma \right) \right] \cos(\omega t_0) \right. \right. \\ & \left. \left. + \left[(\omega_0^2 - 2\beta\gamma) \left(\frac{s_L}{R} - \gamma \right) - 2\beta\omega^2 \right] \frac{\sin(\omega t_0)}{\omega} \right\} \right), \end{aligned} \quad (4.6)$$

where $t_0 = t - \tau_0$. Equation (4.6) gives the energy per unit time carried out by spherical sound waves from the nanoparticle into the surrounding medium.

The analysis show that the power of the sonic signal $\mathcal{W}(t)$ is of the form of damped oscillations. The number of oscillations depends much on the density of the matrix material. The amplitude of the sound power is increased with an increase in the radius of MN, and it is fully damped over longer times. For different metals (but with the same radius) the amplitude of $\mathcal{W}(t)$ will be greater for MNs with a higher coupling constant g_R .

The maximum power of the acoustic signal is reached at the time instant $t_0 = 0$. From equation (4.6), it is equal to

$$\mathcal{W}_{\max}|_{t_0=0} = 4\pi R^2 [\delta p(\tau_0)]^2 / \rho_{sL}. \quad (4.7)$$

The temperature decrease of the electron gas is proportional to the product βT_e , which characterizes the cooling rate of the electron gas. Since the fall of the electron temperature occurs much more slowly than the electron's heating, we can assume that the influence of this mechanism on the sound power is negligible in the first approximation. Formally, this allows us to direct $\beta \rightarrow 0$ in equation (4.6). In this case, the sound signal will correspond to exponentially damped oscillations.

The time dependence of the excess pressure in the case of $\beta \rightarrow 0$, can also be estimated using the equation

$$\delta p(t)|_{\beta \rightarrow 0} \approx -2g_R t T_e(\tau_0)/3, \quad (4.8)$$

which follows from the energy balance equation for the electrons. The maximum power of the signal [equation (4.7)], with the account for approximation (4.8), is

$$\mathcal{W}_{\max}|_{t=\tau_0, \beta \rightarrow 0} = \frac{16}{9} \frac{\pi}{\rho_{sL}} [g_R \tau_0 R T_e(\tau_0)]^2, \quad (4.9)$$

where the dependence on the laser pulse duration appears explicitly.

5. Total energy of the oscillations of the metal nanoparticle

We now determine the total energy transferred from the pulsating spherical surface to the surrounding matrix. We will consider the case $\beta \neq 0$, when the power $\mathcal{W}(t)$ is given by equation (4.6). Integrating equation (4.6) with respect to time over the interval $0 < t < \infty$, in view of expressions (2.4) and (3.2) at $\mathcal{E}_s = 0$, we obtain

$$E = \int_{\tau_0}^{\infty} \mathcal{W}(t) dt = \frac{2\pi R}{\rho_{sL}} \frac{s_L + 2\beta R}{\omega_0^2 + 4\beta(\beta + \gamma)} \alpha^2 T_e^4. \quad (5.1)$$

Equation (5.1) shows that the total energy is proportional to the fourth power of the electron temperature. In the case $\beta = 0$, it is easy to reveal that this energy depends on the volume of the particle and is reciprocal to the rigidity modulus of the medium.

On the other hand, the work necessary to shift the spherical surface at the maximal distance u_{\max} [defined by equation (3.10)], is

$$E = 8\pi R u_{\max}^2 \alpha T_e^2. \quad (5.2)$$

Comparing it with equation (5.1), one finds

$$T_e = 2u_{\max} \sqrt{\frac{\rho_{sL}}{\alpha} \frac{\omega_0^2 + 4\beta(\beta + \gamma)}{s_L + 2\beta R}}. \quad (5.3)$$

Another way to determine T_e is as follows. The total energy obtained by the electron gas in the MN is

$$E_{\text{tot}} = 3V\alpha T_e^2/2. \quad (5.4)$$

The efficiency of the energy transfer η from the pulsating spherical MN to the sound oscillations can be estimated taking the ratio of equations (5.1) and (5.4). It has the simplest form in the case $\beta \rightarrow 0$

$$\eta(T) \equiv E|_{\beta \rightarrow 0}/E_{\text{tot}} = \alpha T_e^2/4\mu. \quad (5.5)$$

The temperature dependence of $\eta(T)$ for noble nanoparticles embedded in a plexiglass matrix is plotted in figure 1. The calculations were done using equations (5.1) and (5.4), with account for the characteristics both of the matrix and the MNs given in tables 1 and 2. The efficiency increases as the square of the temperature. Actually, it increases if the rigidity modulus of the medium in which the nanoparticle is embedded gets smaller. So, knowing from experiment the efficiency η , it is easy to estimate the electron temperature in MN. The highest possible η_T for noble metal nanoparticles with $R = 100 \text{ \AA}$ embedded in a plexiglass matrix are presented in table 2 at $T_e = 10^4$.

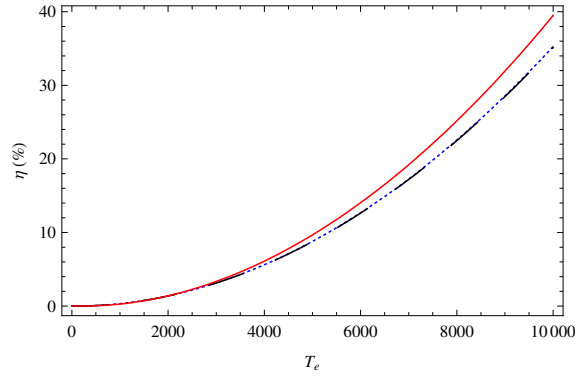


Figure 1. (Color online) Dependence of the sound signal power efficiency on electron temperature for Cu nanoparticle (solid curve), Ag (dashed curve), and Au (dotted curve), embedded in plexiglass.

Table 1. Constants for the plexiglass [21].

Medium	K (dyn/cm ²)	μ (dyn/cm ²)	ρ (g/cm ³)	s_L (cm/s)	s_T (cm/s)
Plexiglass	$5.83 \cdot 10^{10}$	$1.48 \cdot 10^{10}$	1.18	$2.57 \cdot 10^5$	$1.12 \cdot 10^5$

Table 2. Physical parameters of the noble MNs.

Metal	l_D (Å)	α $\left(\frac{\text{erg}}{\text{cm}^3 \text{K}^2}\right)$	n (cm ⁻³) [22]	η_{10^4} (%)
Cu	1197	235.5	$8.45 \cdot 10^{22}$	39.5
Ag	1552	208.35	$5.85 \cdot 10^{22}$	35.15
Au	1968	208.9	$5.90 \cdot 10^{22}$	35.3

6. Conclusions

The method has been proposed for the estimation of hot electron temperature in metallic nanoparticles embedded in a dielectric matrix under irradiation of ultrashort laser pulses. Analytic expressions have been derived for the amplitude and power of longitudinal spherical sound oscillations as function of the density and elastic properties of the medium, the laser pulse duration, electron temperature, nanoparticle radius, and electron-phonon coupling constants.

The maximal displacement of any oscillator in the MN's material is due to the "impact" of the excess pressure of the electron gas. The magnitude of the sound signal power at the moment of the laser pulse ending can be used to estimate the maximal electron temperature in the MN. The latter is determined by the cooling rate of the electron gas and the rate of its pressure change.

The efficiency with which the energy of the hot electrons is carried away by sound oscillations has been examined for the noble metals. It has been shown that the sound energy transfer efficiency is considerably higher in the medium with smaller rigidity moduli.

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Визначення температури електронів у підігрітій металевій наночастинці

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Запропоновано метод визначення температури гарячих електронів в металевій наночастинці, що знаходиться в середовищі під дією ультракоротких лазерних імпульсів. Одержані амплітуда і потужність поздовжніх сферичних акустичних коливань як функція густини і пружних властивостей середовища, тривалості лазерного імпульсу, радіусу частинки, електрон-фононної константи зв'язку та електронної температури. Зроблено оцінку ефективності передачі енергії електронів від підігрітих благородних наночастинок в оточуюче їх середовище за різних температур електронів.

Ключові слова: електронна температура, металева наночастинка, ультракороткі лазерні імпульси
