

Contributions of Inter- and Intraband Excitations to Electron Heat Capacity and Electron-Phonon Coupling in Noble Metals

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This work examines the effects of photonically induced interband excitations from the d-band to states at the Fermi energy on thermophysical properties in noble metals. The change in the electron population in the d-band and the conduction band causes a change in electron heat capacity and electron-phonon coupling factor, which in turn impacts the evolution of the temperature after pulse absorption and electron thermalization. Expressions for heat capacity and electron-phonon coupling factor are derived for electrons undergoing both inter- and intraband transitions. In noble metals, due to the large d-band to Fermi energy separation, the contributions to electron heat capacity and electron-phonon coupling factor of intra- and interband transitions can be separated. At high absorbed laser fluences and pulse energies greater than the interband transition threshold, the interband and intraband contributions to thermophysical properties differ.
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1 Introduction

The well known two temperature model (TTM) [1], which describes the rate of energy transfer from a hot thermalized electron system to a cooler phonon system, has been used to predict temperature changes and to deduce thermophysical properties in a wide array of studies, including ablation of metal targets [2–4], electron-phonon heat transfer in thin films [5–7], effects of microstructural disorder on electron-phonon scattering [8,9], excitations in nanoparticles [10–12], and electron-interface heat transfer [13–17]. In all of these aforementioned studies, including original derivation of the TTM by Anisimov et al. [1], a source term from an optical excitation is considered. However, this source term is only considered as a thermal excitation in heat transfer analyses. In the case of an incident optical excitation, such as that delivered by an ultrashort laser pulse, the incident photon energy will cause the electrons in the metal to undergo various inter- and intraband transitions. In the case of interband transitions, the population in the electronic bands participating in thermal processes will change, which will affect the electron heat capacity and electron-phonon coupling factor [18,19], subsequently affecting the predicted temperature change after the optical excitation [20].

In this technical brief, the effects of optically and thermally excited interband transitions from the d-band (below the Fermi surface) to the Fermi surface on the electron heat capacity and

electron-phonon coupling factor responses are considered and compared with the effects of intraband excitations. Both thermal transitions from Fermi smearing [18,19] and optical excitations from incident photon energies [20] cause a change in the electronic density of states, which in turn affects the thermal properties and temperature changes after the excitation. Based on changes in the electronic population in the conduction band and lower d-band, expressions are derived for heat capacity and electron-phonon coupling factors in the case of inter- and intraband transitions. Specific calculations of electronic heat capacity and electron-phonon coupling factor are presented for Au. Also, the intra- and interband contributions to the thermal properties are separated.

2 Separating Intra- and Interband Transitions

The electronic heat capacity, $C_e(T_e)$, and the electron-phonon coupling factor, $G(T_e)$, are dependent on the population of the electron bands within a few $k_B T_e$ of the Fermi surface [9], where k_B is Boltzmann's constant and T_e is the temperature of the electron system. Therefore, the density of states and the electronic distribution around the Fermi surface will dictate $C_e(T_e)$ and $G(T_e)$ [9]. In the case of intraband transitions, the population in the electron bands does not change, so $C_e(T_e)$ and $G(T_e)$ are governed by classical low temperature solid state theory. However, interband transitions increase/decrease the electron populations at various energies depending on the nature of the excitation, and therefore the density of states of the various bands below the Fermi surface must be taken into account.

In this development, an isotropic dispersion is assumed for the electron bands. Implications of this assumption are discussed and some calculations are compared with previous results using ab initio calculations for the density of states [19]. For the specific Au calculations involving interband excitations, only the outermost d-bands ($5d^{10}$) are considered, since for electron temperatures and photon energies of interest (0–10,000 K and 0–2.4 eV), this is the only band that will contribute excited electrons to the Fermi level. Excitations from the Fermi level to higher energy bands will not be considered since those higher energy bands are initially empty and, therefore, there is no net change in electron population above the Fermi level from excitations originating at the Fermi level. In addition, the $5d^{10}$ band to Fermi surface transitions is the dominant population changing transition in Au, as shown in its thermomodulation spectra [21]. This transition, often called the interband transition threshold (ITT) [7], is 2.4 eV in Au [22] and assumed constant with wavevector in this work due to the assumption of an isotropic dispersion.

An intraband transition occurs from an excitation with energy less than the interband transition threshold energy, ε_{ITT} . If the thermal excitation of energy $k_B T_e$ or optical excitation of energy $h\nu$, where h is Planck's constant and ν is the photon frequency, is less than ε_{ITT} , then the thermal properties are only subject to intraband effects. Intraband transitions would occur within the conduction band only as electrons are thermally or optically excited. At these low energies (i.e., excitation energies less than ε_{ITT}), a thermal or optical excitation does not excite any electrons from the d-bands. Therefore, the electron heat capacity is calculated by

$$C_e(T_e) = \int_{-\infty}^{\infty} \varepsilon D_C(\varepsilon) \frac{\partial f(\varepsilon, \mu(T_e), T_e)}{\partial T_e} d\varepsilon \quad (1)$$

where ε is the electron energy, $D_C(\varepsilon)$ is the conduction band density of states (in the case of Au, the $6s^1$ band), and f is the Fermi–Dirac distribution function with $\mu(T_e)$ being the chemical potential, which is a function of electron temperature. In the low temperature limit, $\mu(T_e)$ is approximately equal to the Fermi energy, ε_F , and Eq. (1) can be expressed as $C_e(T_e) = \gamma T_e$, where γ is commonly called the Sommerfeld coefficient, which is theoretic-

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cally $62.9 \text{ J m}^{-3} \text{ K}^{-2}$ [23]. The reduction of Eq. (1) to $C_e(T_e) = \gamma T_e$ also assumes that only electrons at the Fermi energy participate in energy storage, that is, $C_e(T_e) \propto D_C(\varepsilon_F)$.

The general form for the electron-phonon coupling factor is given by [24]

$$G(T_e) = \pi \hbar k_B \lambda \langle \omega^2 \rangle \int_{-\infty}^{\infty} \frac{(D_T(\varepsilon))^2}{D_T(\varepsilon_F)} \left(- \frac{\partial f(\varepsilon, \mu(T_e), T_e)}{\partial \varepsilon} \right) d\varepsilon \quad (2)$$

where \hbar is the reduced Planck's constant, λ is the dimensionless electron-phonon mass enhancement parameter [25], $\langle \omega^2 \rangle$ is the second moment of the phonon spectrum [26], and $D_T(\varepsilon)$ is density of states of all the electron bands modified by the excitation. For Au, $\lambda \langle \omega^2 \rangle = 23 \text{ meV}^2 / \hbar^2$ [27]. In the case of only intraband transitions in the conduction band, Eq. (2) is given by

$$G(T_e) = \pi \hbar k_B \lambda \langle \omega^2 \rangle \int_{-\infty}^{\infty} \frac{(D_C(\varepsilon))^2}{D_C(\varepsilon_F)} \left(- \frac{\partial f(\varepsilon, \mu(T_e), T_e)}{\partial \varepsilon} \right) d\varepsilon \quad (3)$$

At relatively low temperatures, $\partial f(\varepsilon, \mu(T_e), T_e) \partial \varepsilon \approx \delta(\varepsilon - \mu(T_e)) \approx \delta(\varepsilon)$ and Eq. (3) reduces to $G_0 = \pi \hbar k_B \lambda \langle \omega^2 \rangle D_C(\varepsilon_F)$, which is the original expression derived by Allen [28].

An interband transition occurs from an excitation with energy greater than ε_{IT} . In this case, electrons in the d-band are excited to empty states around the Fermi energy and must be considered in thermal processes. Therefore, the total electron heat capacity taking into account the conduction band and d-band is given by

$$C_e(T_e) = \int_{-\infty}^{\infty} \varepsilon D_T(\varepsilon) \frac{\partial f(\varepsilon, \mu(T_e), T_e)}{\partial T_e} d\varepsilon \quad (4)$$

and the electron-phonon coupling factor is given by Eq. (2). However, in the event that an electron moves from a filled state in a d-band to an empty state near the Fermi level through an optical excitation, the density of states of the conduction band and the d-band will be changed without a change in Fermi smearing and, therefore, calculations of $C_e(T_e)$ and $G(T_e)$ will be altered. In this case, the number density of the electrons in each band will change. The number of empty states in the conduction band for which there is sufficient photon energy to excite an electron is given by [20]

$$n_{\text{available}} = \int_{-\infty}^{\infty} D_C(\varepsilon) (1 - f(\varepsilon, \mu(T_e), T_e)) (1 - H[\varepsilon - (\mu(T_e) - (\varepsilon_{IT} - h\nu))]) d\varepsilon \quad (5)$$

where $H[\dots]$ is the Heaviside function. Equation (5) will affect the density of states calculations. Note that this expression for available states is more physical than the expression used previously [20] since Eq. (5) does not put any stipulation on the width of the Fermi surface that is available for electronic excitation, in addition to accounting for the maximum number of electrons that can fill the bands, as opposed to arbitrarily increasing the number density.

Separation of the interband contribution to electron heat capacity is straightforward recognizing that any excitation involving the d-band electrons is an interband contribution to electron heat capacity. Therefore, since heat capacity is an additive property, the interband contribution to electron heat capacity is given by

$$C_{e,\text{inter}}(T_e) = \int_{-\infty}^{\infty} \varepsilon D_D(\varepsilon) \frac{\partial f(\varepsilon, \mu(T_e), T_e)}{\partial T_e} d\varepsilon \quad (6)$$

where $D_D(\varepsilon)$ is density of states of the d-band and $D_T(\varepsilon) = D_C(\varepsilon) + D_D(\varepsilon)$ and, therefore, $C_{e,\text{intra}}(T_e)$ is defined by Eq. (1), which can be rewritten as

$$C_{e,\text{intra}}(T_e) = \int_{-\infty}^{\infty} \varepsilon D_C(\varepsilon) \frac{\partial f(\varepsilon, \mu(T_e), T_e)}{\partial T_e} d\varepsilon \quad (7)$$

The separation of the transitions on electron-phonon coupling factor is dependent on the band structure, however. Equation (2) can be rewritten as

$$G(T_e) = \pi \hbar k_B \lambda \langle \omega^2 \rangle \int_{-\infty}^{\infty} \frac{(D_C(\varepsilon) + D_D(\varepsilon))^2}{D_C(\varepsilon_F) + D_D(\varepsilon_F)} \left(- \frac{\partial f(\varepsilon, \mu(T_e), T_e)}{\partial \varepsilon} \right) d\varepsilon \quad (8)$$

In noble metals (Au, Ag, and Cu), the density of states of the d-band at the Fermi level is zero, that is, $D_D(\varepsilon_F) = 0$. Therefore, Eq. (8) reduces to

$$G(T_e) = \pi \hbar k_B \lambda \langle \omega^2 \rangle \int_{-\infty}^{\infty} \frac{(D_C(\varepsilon) + D_D(\varepsilon))^2}{D_C(\varepsilon_F)} \left(- \frac{\partial f(\varepsilon, \mu(T_e), T_e)}{\partial \varepsilon} \right) d\varepsilon \quad (9)$$

Now considering only intraband transitions, Eq. (9) reduces to Eq. (3). In noble metals, the density of states of the d-band is so much greater than that of the conduction band that the conduction band density of states is relatively constant with energy (this is especially true in Au as evident from ab initio electronic structure calculations [19,29]) and, therefore, Eq. (3) reduces to the expression for G_0 derived by Allen, even at high temperature. Thus, the interband contribution is given by

$$G_{\text{inter}}(T_e) = \pi \hbar k_B \lambda \langle \omega^2 \rangle \int_{-\infty}^{\infty} \frac{(D_D(\varepsilon))^2}{D_C(\varepsilon_F)} \left(- \frac{\partial f(\varepsilon, \mu(T_e), T_e)}{\partial \varepsilon} \right) d\varepsilon \quad (10)$$

The reader is cautioned that Eq. (10) is only valid for noble metals due to the large d-band separation from the Fermi energy. Since the intraband contribution to G reduces to G_0 for noble metals

$$G_{\text{total}}(T_e) = G_0 + G_{\text{inter}}(T_e) \quad (11)$$

showing that, for noble metals, the inter- and intraband contributions to the electron-phonon coupling factor are additive, similar to heat capacity.

3 Effects of Optical Excitations

The key to evaluating $C_e(T_e)$ and $G(T_e)$ lies in determining $\mu(T_e)$, which, when only considering intraband transitions, can be approximated by the Sommerfeld expansion [30], but when taking into account d-band excitations must be calculated by conservation of electron number density by evaluating

$$n_C + n_D = \int_{-\infty}^{\infty} (D_C(\varepsilon) + D_D(\varepsilon)) f(\varepsilon, \mu(T_e), T_e) d\varepsilon \quad (12)$$

where $n_C + n_D$ is a constant and $\mu(T_e)$ is iterated for each temperature. For Au, n_C is $5.9 \times 10^{28} \text{ m}^{-3}$ and n_D is $5.9 \times 10^{29} \text{ m}^{-3}$, which is estimated by the atomic density [23] and the number of electrons in the $6s^1$ and $5d^{10}$ bands, respectively [31]. The conduction band density of states is estimated by $D_C = 3n_{C,\text{total}} \varepsilon^{1/2} / (2\varepsilon_F^{3/2})$, where the Fermi energy of Au is 5.53 eV [23], and $n_{C,\text{total}}$ is the total number of electrons in the conduction band after the excitation, which is given by $n_{C,\text{total}} = n_C + n_{\text{excited}}$. Here, n_{excited} is the number of electrons excited from the d-band to the conduction band from incident photons, which is a function of the number of photons absorbed by the material. If there are enough photons absorbed by the metal to excite all the electrons in the d-band up to available states in the conduction band, then $n_{\text{excited}} = n_{\text{available}}$, where $n_{\text{available}}$ is defined by Eq. (5), and therefore $n_{C,\text{total}} = n_C + n_{\text{available}}$. For this condition to be true, then $n_{\text{available}} \leq n_{\text{photons}}$, where n_{photons} is the number of photons per unit

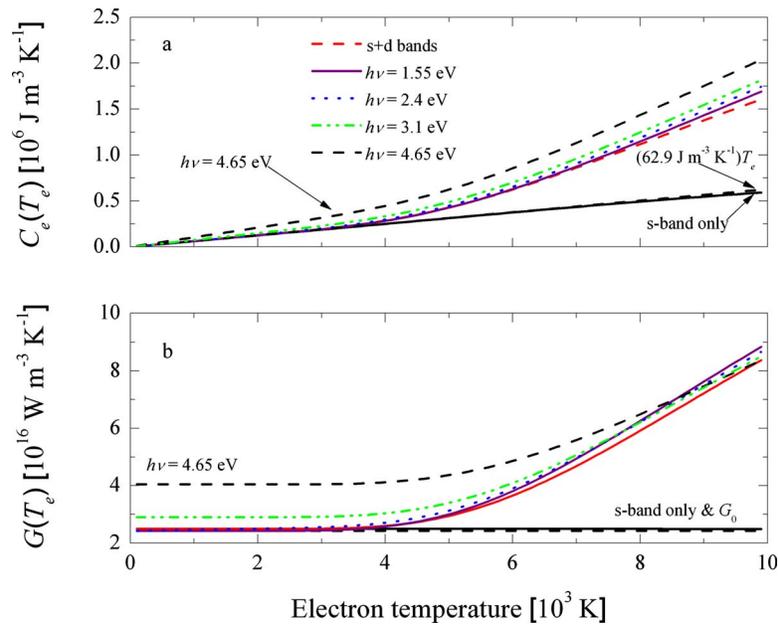


Fig. 1 Predictions of (a) electron heat capacity and (b) electron-phonon coupling factor in Au. The numerical calculations are performed using only the s-band density of states (s-band only), the s- and d-band density of states with no photonic excitation (s+d bands), and the s- and d-band density of states with a photonic excitation, $h\nu$ ($h\nu=1.55$ eV, $h\nu=2.4$ eV, $h\nu=3.1$ eV, and $h\nu=4.65$ eV). The s-band density of states calculations show close agreement to low temperature theory throughout the temperature range $T_e=0-10,000$ K.

volume in the absorbed laser pulse. The number of photons per volume can be estimated as $n_{\text{photons}}=A/(h\nu\delta)$, where A is the absorbed fluence and δ is the optical penetration depth at $h\nu$. In the case n_{photons} is less than $n_{\text{available}}$, then only $n_{\text{photons}}/n_{\text{available}}$ of the empty states in the conduction band will be filled by electrons undergoing interband transitions.

Assuming that all empty states in the conduction band below the Fermi level are filled by interband excited electrons, that is $n_{\text{available}} \leq n_{\text{photons}}$ and $n_{C,\text{total}}=n_C+n_{\text{available}}$, the number of states in the d-band after photonically induced interband transitions is given by $n_{D,\text{total}}=n_D-n_{\text{available}}$. The density of states of the $5d^{10}$ band in Au can be approximated by a square function with a width of 5.28 eV [32,33] and the high energy edge of the square function 2.4 eV below the Fermi energy, giving rise to the ITT energy in Au, so that $D_D(\epsilon)=n_{D,\text{total}}/5.28(-H[\epsilon-3.13]+H[\epsilon+2.15])$. Note the calculation of temperature dependent thermophysical properties using this approximate band structure has shown close agreement with calculations of thermophysical properties using exact ab initio calculations for electronic band structure [29].

Figure 1 shows the calculations for (a) the electronic heat capacity, $C_e(T_e)$, and (b) electron-phonon coupling factor, $G(T_e)$, as a function of temperature in Au for photonic excitations of various energies, $h\nu=1.55, 2.40, 3.10,$ and 4.65 eV. 1.55 eV is the fundamental output of Ti:Al₂O₃ oscillators, which are typically used to examine electron-phonon coupling; 3.10 eV and 4.65 eV are the frequency doubled and tripled components of the fundamental Ti:Al₂O₃ frequency; 2.40 eV is the interband transition threshold of Au. Figure 1(a) shows predictions for the electron heat capacity for the different band excitation cases presented in Fig. 1(a). When only considering the 6s band in the numerical calculation, the results are almost identical to the theoretical value for heat capacity predicted by $C_e(T_e)=\gamma T_e$, where $\gamma=62.9$ J m⁻³ K⁻². When considering the 5d excitation, $C_e(T_e)$ increases due to Fermi smearing and thermal excitation of the d-band electrons. This is intuitive since, when considering d-band electrons at elevated

temperatures, there are more electrons available to “store” heat. Photonic excitations that completely fill empty states in the conduction band increase the electron heat capacity; the increasing trend in heat capacity increases as more empty states in the conduction band are filled.

Figure 1(b) shows the calculations for the electron-phonon coupling factor for the different excitation cases discussed. The s-band only calculations show a constant trend in G with temperature and are very close to the predictions of G_0 . This further supports Eq. (12) where the constant G is the intraband contribution. The difference between the two values most likely lies in the assumption in the numerical calculation of a parabolic density of states with energy and error in the numerical integration method. However, the difference between the predicted G_0 from theory (2.43×10^{16} W m⁻³ K⁻¹) and the constant G predicted from the numerical calculations (2.49×10^{16} W m⁻³ K⁻¹) is negligible when comparing to G with d-band excitations. Including d-band excitations to empty states above the Fermi surface noticeably increases the electron-phonon coupling factor at low temperatures. In both Figs. 1(a) and 1(b), when the photon energy is greater than ϵ_{ITT} , the values and trends of the thermophysical properties change due to a change in number density in the electron bands around the Fermi energy which affects the electron density of states.

Figures 1(a) and 1(b) show the total $C_e(T_e)$ and $G(T_e)$, which include both intra- and interband contributions. The intraband contribution in each figure is depicted by the “s-band only” data. Therefore, the interband contribution is determined by subtracting the s-band only data from the total $C_e(T_e)$ and $G(T_e)$ predictions. The intraband contribution to the heat capacity is given by $C_{e,\text{intra}}(T_e)=62.9T_e$ over the entire temperature range. The intraband contribution to electron-phonon coupling is simply an offset of $G_{\text{intra}}=G_0 \approx 2.49 \times 10^{16}$ W m⁻³ K⁻¹.

4 Conclusions

This work examines the effects of photonic induced interband excitations from the d-band to states at the Fermi energy on thermophysical properties in noble metals. The change in the electron population in the d-band and the conduction band causes a change in electron heat capacity and electron-phonon coupling factor, which in turn impacts the evolution of the temperature after pulse absorption and electron thermalization. Expressions for heat capacity, electron-phonon coupling factor, and transient temperature change are derived for electrons undergoing both inter- and intraband transitions. In noble metals, due to the large d-band to Fermi energy separation, the contributions to electron heat capacity and electron-phonon coupling factor of intra- and interband transitions can be separated. At high absorbed laser fluences and pulse energies greater than the interband transition threshold, the interband and intraband contributions to thermophysical properties differ.

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Nomenclature

C_e	= electron heat capacity, $\text{J m}^{-3} \text{K}^{-1}$
D	= electron spectral density of states per unit volume, $\text{m}^{-3} \text{eV}^{-1}$
f	= Fermi-Dirac distribution function
G	= electron-phonon coupling factor, $\text{W m}^{-3} \text{K}^{-1}$
H	= Heaviside function
h	= Planck's constant, J s
\hbar	= Planck's constant divided by 2π , J s
k_B	= Boltzmann constant, J K^{-1}
n	= number density, m^{-3}
T_e	= electron temperature, K

Greek Symbols

ϵ	= electron energy, eV
γ	= Sommerfeld coefficient (linear coefficient to heat capacity), $\text{J m}^{-3} \text{K}^{-2}$
λ	= electron-phonon mass enhancement parameter
$\langle \omega^2 \rangle$	= second moment of the phonon spectrum
μ	= chemical potential, eV
ν	= photon frequency, Hz

Subscripts

C	= conduction band
D	= d-band
F	= Fermi
inter	= interband transition
intra	= intraband transition
T	= total

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