Ultrafast dynamics of non-equilibrium electrons and strain generation under femtosecond laser irradiation of Nickel

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Abstract
We present a theoretical study of the ultrafast electron dynamics in transition metals of large electron–phonon coupling constant using ultrashort pulsed laser beams. The significant influence of the dynamics of produced nonthermal electrons to electron thermalisation and electron–phonon interaction is thoroughly investigated for various values of the pulse duration (i.e., from 10 fs to 2.3 ps). The model correlates the role of nonthermal electrons, relaxation processes and induced stress–strain fields. Simulations are presented by choosing Nickel (Ni) as a test material to compute electron–phonon relaxation time due to its large electron–phonon coupling constant. We demonstrate that the consideration of the aforementioned factors leads to significant changes compared to the results the traditional two-temperature model provides. The proposed model predicts a substantially (~33%) smaller damage threshold and a large increase of the stress (~20%, at early times) which first underlines the role of the nonthermal electron interactions and second enhances its importance with respect to the precise determination of laser specifications in material micromachining techniques.

1 Introduction
Material processing with ultra-short pulsed lasers has received considerable attention over the past decades due to its important technological applications, in particular in industry and medicine [1–9]. These abundant applications require a thorough knowledge of the fundamentals of laser interaction with the target material for enhanced controllability of the resulting modification of the target relief. Physical mechanisms that lead to surface modification have been extensively explored both theoretically and experimentally [10–23].

It is well-known that after irradiation, the initial electron population is highly nonthermal. Experimental observations related to the non-equilibrium dynamics of electron systems in metals shows nonthermalised electron distribution leads to a faster electron–phonon relaxation [24, 25]. Hence, to discriminate nonthermal electron-generated electron–phonon relaxation, it is necessary to investigate thoroughly the dynamics of nonthermal electron gas in these conditions. Regarding modelling of the laser-matter interaction, it is known that the traditional two temperature model (TTM) [26] assumes a rapid (instantaneous) thermalisation of the electronic distribution. Therefore, TTM yields an overestimation of the electronic temperature which has been also confirmed by pump–probe experiments [27]. To overcome the limitations of the TTM, analysis based on the Boltzmann’s transport equations [28] or revised versions of the TTM [27, 29–31] have been proposed. Those works presented the necessity for the inclusion of the initial nonthermal (NTH) electron population. More specifically, Sun at al. used simultaneously a three-coupled equation-based model and Boltzmann’s transport equation, where heating of the electron gas by the initial NTH electrons and the lattice are included [31]. On the other hand, Lisowksi et al. proposed an improved version of the TTM by presenting a three-temperature model where a temperature for the NTH electrons was introduced [27]. Although these approaches successfully predicted results from pump–probe experiments for noble metals [31] or Ruthenium [27], they were characterised by the necessity of including fitting parameters [29].
By contrast, in a recent approach presented by Carpene, an extension of the TTM was introduced by incorporating the electron thermalisation dynamics into the source term, a potentially direct energy transfer from NTH electrons to the lattice and the consideration of a very-low-density NTH electron distribution [29]. The approach allows a consistent calculation of the resulting (thermalized) electron temperature assuming the contribution of both the thermalized (TH) and NTH electron distributions. The incorporation of the influence of both TH and NTH electron dynamics, also, overcomes the weakness of the classical TTM related to the non-equilibrium state of the NTH, and thereby, inability to define an “electron temperature” in the early stages. Validation of the model through pump–probe experiments illustrated that it provides an accurate description of ultrafast dynamics after irradiation of noble metals with ultrashort laser pulses [32–36].

Nevertheless, there are still some open questions regarding the model presented by Carpene. It was assumed that the electron density of states (DOS) is constant around the Fermi energy [29]. This appears to constitute a sufficient approximation for noble metals such as Au, Cu, Ag, however, for transition metals with large electron–phonon coupling strength such as nickel (Ni) or titanium (Ti) it is a rather crude approximation that expects to lead to inaccurate results [37]; hence, a more rigorous approach is required to determine the combined influence of the NTH electrons and DOS. On the other hand, experimental observations related to the nonequilibrium dynamics of electron systems in metals shows NTH electron distribution leads to a faster electron–phonon relaxation [24, 25]. It is important, thus, to elaborate on how metals characterised by a large electron–phonon coupling strength \( G_{el} \) influence relaxation processes.

The elucidation of the aforementioned issues is of paramount importance not only to understand further the underlying physical mechanisms of laser-matter interactions and ultrafast electron dynamics but also to associate the resulting thermal effects with the surface response which can be used to process systematically the material. Therefore, there is a growing interest to reveal the physics of the underlying processes from both a fundamental and application point of view.

To proceed with the influence of electronic excitation on the morphological changes, one aspect that has yet to be explored is the correlation of the pulse width, energy deposition and structural changes. In principle, morphological surface changes at low excitation levels are strongly related to stress generation as well as whether lattice temperatures induce large stresses (i.e., that exceeds the yield stress) [37]. It is known that although NTH interactions with TH electrons and the lattice yield remarkably smaller maximum electron temperature they induce smaller maximum lattice temperature variations due to the large lattice heat capacity [29]. Nevertheless, it is important to investigate whether a material with a large electron–phonon coupling constant and a complex DOS around the Fermi energy behaves differently and thereby, significant lattice temperature changes occur which, in turn, is also reflected on an enhanced mechanical response.

To this end, we present an extension of the model proposed by Carpene which comprises: (1) the consideration of a nonconstant DOS around the Fermi energy, and (2) a thermomechanical component that describes the mechanical response of the material due to material heating. To highlight the significance of the nonthermal electrons for materials with large and temperature-dependent electron–lattice coupling constant, \( G_{el} \), Ni is used. For the sake of simplicity, the investigation has been focused on single shot while a similar approach could be pursued in case of repetitive irradiation. Low laser fluences have been used primarily in this work to distinguish the role of nonthermal electron contribution and electron–phonon relaxation processes; this is due to the fact that at larger energies, more complex effects such as ablation or melting occur. In that case, hydrodynamical models or atomistic simulations are required to be incorporated into a multiscale theoretical framework which could hinder the significance of the aforementioned factors while possible major morphological changes might also be attributed to other effects [10, 38, 39]. Nevertheless, prediction for the electronic and lattice temperatures at larger fluences were also performed to estimate the energy fluence at which damage occurs (i.e., when lattice temperature exceeds the melting point of the material).

In the following section, we present the theoretical framework used to describe the physical mechanism and the components of the revised TTM (rTTM). Section 3 explains the numerical algorithm and the adaptation of the model to Ni. A systematic analysis of the results and the role of the nonthermal electron contribution are presented in Sect. 4 while the energy exchange between electrons and lattice for small excitations is also investigated. We determine the thermalisation time of the laser-excited electron gas as a function of the pulse duration. The thermomechanical response is correlated with the pulse duration through the computation of strain fields and displacements. A parametric study is followed in which the role of pulse duration variation in both thermal (evolution of electron and lattice temperatures) and mechanical response of the material (stress/strain propagation) is investigated. All theoretical results are tested against the traditional TTM to highlight the discrepancies. Furthermore, simulations yield the magnitude of change of the optical characteristics within the laser heating time that potentially affects laser energy absorption. Fluence dependence of the thermomechanical response and determination of
ablation thresholds are also explored. Concluding remarks follow in Sect. 5.

2 Theoretical model

2.1 Laser beam profile and non-equilibrium electrons

The laser pulse at time \( t' \) is described by an energy flux (in a three-dimensional space characterised by the Cartesian coordinates \( x,y,z \)), \( I(t,x,y,z) \) provided by the following expressions

\[
- \frac{\partial I(t',x,y,z)}{\partial z} = \alpha(t',x,y,z)I(t',x,y,z) = W(t',x,y,z),
\]

(1)

where \( W(t',x,y,z) \) corresponds to the absorbed laser power density at time \( t' \), \( \alpha \) is the absorption coefficient, \( \tau_p \) is the pulse duration, \( E_p \) is the fluence and \( R_0 \) stands for the Gaussian beam radius (i.e., distance from the beam axis where the optical intensity drops to \( 1/e^2 \)). For the sake of simplicity, it is assumed that the ballistic length of electrons for Ni is small, and therefore, it is neglected [40]. Based on a previous work by Carpene [29], it is assumed that within an infinitesimal time \( \Delta t' \) laser photons will interact with electrons lying in occupied states below Fermi energy \( \epsilon_F \), causing their excitation to previously unoccupied states above \( \epsilon_F \). As a result, an infinitesimal nonthermal change \( \Delta f \) to the (Fermi–Dirac) electronic distribution \( f(\epsilon) \) will be produced which has the form [29, 31]

\[
\Delta f(\epsilon, t', x, y, z) = \frac{\delta_0(\epsilon, t', x, y, z)(f(\epsilon - h\nu) - f(\epsilon)[1 - f(\epsilon + h\nu)])}{[1 - f(\epsilon)] - f(\epsilon)[1 - f(\epsilon + h\nu)]},
\]

(3)

where \( f(\epsilon) = \{1 + \exp[(\epsilon - \mu)/k_B T_\epsilon]\}^{-1} \) is the Fermi–Dirac distribution for electrons with energies \( \epsilon \) and temperature \( T_\epsilon \), \( h\nu \) is the laser photon energy (~1.55 eV for laser beam wavelength \( \lambda_L = 800 \) nm), \( \mu \) is the chemical potential that is equal to \( \epsilon_F \) at \( T_\epsilon = 0 \) K and \( k_B \) is the Boltzmann constant. We note that comparing the underlying physics with the proposed processes in the case of noble metals with small coupling constant [29], some significant changes have to be performed to describe nonthermal electron distribution and their influence in excitation, thermalisation and electron–phonon relaxation processes for transition materials of such as Ni or Ti:

(a) First, it is evident that at fluences large enough to produce very energetic electrons (Fig. 1), the variation of the Fermi–Dirac distribution (Eq. 5) cannot be described by a step-like function for energies in the range \([\epsilon_F - \hbar\nu, \epsilon_F + \hbar\nu]\) [29]. It is evident that at larger temperatures a different shape of \( \Delta f \) contains a substantially large tail that becomes even bigger with increasing \( T_\epsilon \). According to Fig. 1, the non-zero change of the Fermi–Dirac distribution in a larger range of energies (characteristic for larger electron temperatures) requires consideration of the complete expression of the nonthermal electron distribution to estimate the correlation of the energy density associated with the nonthermal electrons and the absorbed laser power density. The size of \( \Delta f \) is computed by the following expression

\[
\int_{\epsilon_F - h\nu}^{\epsilon_F + h\nu} \Delta f(\epsilon, t', x, y, z)N(\epsilon)\epsilon d\epsilon = W(t', x, y, z)dt',
\]

(4)

which relates the absorbed laser power density within the interval \( \Delta t' \) with the energy density of the NTH electrons for energies in \([\epsilon_F - \hbar\nu, \epsilon_F + \hbar\nu]\).

(b) Second, (which is more important), results related to Fig. 1 assume a constant DOS that can be used for energies in the range \([\epsilon_F - \hbar\nu, \epsilon_F + \hbar\nu]\) for noble metals such as Au, Cu, Ag. [41]. By contrast, it is rather a crude

![Fig. 1 Change of the Fermi–Dirac distribution function due to nonthermal electrons for \( T_\epsilon = 300 \) K (solid line) and \( T_\epsilon = 5000 \) K (dashed line). The dotted vertical line indicates the position of energy Fermi \( \epsilon_F \).](image-url)
approximation for transition metals such as Ni. This is demonstrated by the form of the DOS \( N(\varepsilon) \) (illustrated in Fig. 2 based on simulations performed in Ref. [41]). It is evident that DOS for Ni is not constant around \( \varepsilon_F \) and therefore, the correct expression for \( \delta_0 \) should result from the solution of Eqs. (3, 4)

\[
\delta_0(t', x, y, z) = \frac{W(t', x, y, z)\delta t'}{\int_{\varepsilon_F - \hbar}^{\varepsilon_F + \hbar} (f(\varepsilon - \hbar) \{1 - f(\varepsilon)\} - f(\varepsilon) \{1 - f(\varepsilon + \hbar)\})N(\varepsilon)\delta \varepsilon}.
\]

(5)

The complexity of DOS for Ni suggests that Eq. 5 leads to a more complicated expression for the computation of \( \delta_0 \) and eventually \( \Delta f \) should have a different magnitude from the one illustrated in Fig. 1. Due to the lack of a simple analytical expression for \( N(\varepsilon) \) for energies in that range, the computation of \( \Delta f \) is derived through a numerical solution of Eqs. (1–5) in which \( N(\varepsilon) \) is derived through fitting of the data in Fig. 2a (see Fig. 2b, which shows \( \Delta f \) for Nickel at \( T_e = 300 \) and 5000 K). The magnitude of the energy independent \( \delta_0 \) yields a nonthermal change \( \Delta f \) which has a smaller depth at smaller electron temperatures compared to that for a constant \( N(\varepsilon) \) (see Figs. 1, 2b). Furthermore, compared to Fig. 1, \( \Delta f \) appears to decrease. This indicates that both the range and the magnitude of the values of the DOS of the material play an important role in the evaluation of the nonthermal electron distribution function \( \Delta f \) that should not be overlooked. On the other hand, the initial nonthermal electron distribution relaxes at a rate which is determined both from an electron–electron and electron–phonon interaction provided by the following expression [29]

\[
\Delta f_{NT}(\varepsilon, t - t', x, y, z) = \exp \left( -\frac{t - t'}{\tau_{ee}} - \frac{t - t'}{\tau_{f}} \right) \Delta f(\varepsilon, t', x, y, z),
\]

(6)

where \( \tau_{ee} = \frac{128}{\sqrt{3\pi^2}a_p} \left( \frac{e_F}{\varepsilon_F} \right)^2 \equiv \tau_0 \left( \frac{e_F}{\varepsilon_F} \right)^2 \) and \( \omega_p \) is the plasma frequency (\( \hbar \omega_p = 15.92 \) eV for Ni [42] which leads to \( \tau_0 \approx 2 \) fs and \( \omega_p = 2.41 \times 10^{16} \) rad/s) and \( e_F \approx 9.7 \) eV for Ni [43]. By contrast, the electron–phonon collision is provided by the expression \( \tau_{ep} = \frac{\hbar}{\epsilon_F} \theta_D \), where \( \tau_f \approx 2.25 \) fs for Ni [43] stands for the time between two subsequent collisions with the lattice of nonthermal quasiparticles of energy \( \varepsilon - e_F = \hbar \nu \) while \( \theta_D (\theta_D = 450 \) K, for Ni [44]) is the Debye temperature.

The rate of the energy density \( n \) that is transferred from the nonthermal electron distribution to both the thermal electrons and the lattice is described by the following expression [29]

\[
\frac{dn(t, t')}{dt} = \int_{\varepsilon_F - \hbar}^{\varepsilon_F + \hbar} \Delta f_{NT}(\varepsilon, t - t', x, y, z)N(\varepsilon)\delta \varepsilon
\]

\[
= \int_{\varepsilon_F - \hbar}^{\varepsilon_F + \hbar} \left( \frac{1}{\tau_0} \left( \frac{\varepsilon - e_F}{\varepsilon_F} \right)^2 + \frac{1}{\tau_{ep}} \right) \Delta f(\varepsilon, t', x, y, z)
\]

\[
\times \exp \left( -\frac{t - t'}{\tau_0} \left( \frac{\varepsilon - e_F}{\varepsilon_F} \right)^2 - \frac{t - t'}{\tau_{ep}} \right) N(\varepsilon)\delta \varepsilon
\]

\[
\equiv \frac{dn_{ee}(t, t')}{dt} + \frac{dn_{ph}(t, t')}{dt}
\]

(7)

where

Fig. 2  a The electron DOS of Ni (data calculated in Ref. [41]), b change of the Fermi–Dirac distribution function for Ni due to nonthermal electrons for \( T_e = 300 \) K (solid line) and \( T_e = 5000 \) K (dashed line). The dotted vertical line indicates the position of energy Fermi \( e_F \)
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\[ \frac{\partial u_{ce}(t, t')}{\partial t} = -\frac{1}{\tau_0} \int_{\varepsilon_{F}+h\nu}^{\varepsilon_\text{F}} \left( \frac{\varepsilon - \varepsilon_\text{F}}{\varepsilon_\text{F}} \right)^2 \Delta f(\varepsilon, t, x, y, z) \times \exp \left( \frac{-t - t'}{\tau_0} \left( \frac{\varepsilon - \varepsilon_\text{F}}{\varepsilon_\text{F}} \right)^2 \right) N(\varepsilon) \varepsilon \, d\varepsilon \]

and

\[ \frac{\partial u_{cl}(t, t')}{\partial t} = -\left( \frac{1}{\varepsilon_{ep}} \right) \int_{\varepsilon_{F}-h\nu}^{\varepsilon_{F}+h\nu} \Delta f(\varepsilon, t, x, y, z) \times \exp \left( \frac{-t - t'}{\tau_0} \left( \frac{\varepsilon - \varepsilon_\text{F}}{\varepsilon_\text{F}} \right)^2 \right) N(\varepsilon) \varepsilon \, d\varepsilon \]

The quantities \( u_{ce} \) and \( u_{cl} \) correspond to the rate of energy density exchanged between the NTH electrons and (1) the TH electrons and (2) the lattice through electron–electron and electron–phonon scattering, respectively. Time \( t \) corresponds to the time point at which the NTH distribution relaxes to \( \Delta f \) at \( t = t' \) as seen in Eq. 8. The corresponding total energy relaxation (i.e., at time \( t \)) is derived by adding the infinitesimal contributions of excitation for time intervals \( dt' \) at all times \( t' < t \). As a result, the following expressions are obtained by integrating over \( t' \),

\[ \frac{\partial U_{ce}(t)}{\partial t} = \int_0^t \frac{\partial u_{ce}(t, t')}{\partial t} \, dt' \]

\[ \frac{\partial U_{cl}(t)}{\partial t} = \int_0^t \frac{\partial u_{cl}(t, t')}{\partial t} \, dt' \]

Regarding a more precise estimation of the effect of the pulse length (or fluence) to the DOS, it is also important to consider a detailed band structure with DOS, especially in the medium and high-fluence regimes. Such fluences lead to laser-induced surface modifications, since the transfer of electrons from high-density \( d \)-bands to low-density \( s(p) \)-bands results in the fluence-dependent saturation of the predominant interband absorption (i.e., it also influences the intraband absorption term). This effect is characteristic for semiconductors [45], but also occurs for \( d \)-metals [46]. However, the predominant role of the present work aims to reveal the differences in the thermomechanical effects at low fluences, and therefore, corrections due to band filling and induced saturation effects are not taken into account. Similarly, high fluences are expected to influence the characteristics of the nonthermal electron population away from the range \( [\varepsilon_\text{F} - h\nu, \varepsilon_\text{F} + h\nu] \). It turns out that the effect of the tail in the \( \Delta f \) function for relatively small attained values of \( T_\text{c} \) (Figs. 1, 2, 7) is not significantly large to produce remarkably enhanced thermomechanical response.

### 2.2 Optical properties

It is well known that the transient variation of the dielectric constant through the temperature dependence of the electron relaxation time potentially leads to a change of the optical properties of the material during the irradiation time that needs to be evaluated [47, 48]. More specifically, this behavior has been noted in noble metals (such as Au and Cu) where a static consideration of the optical properties led to an incorrect estimation of the energy deposition which was reflected from an underestimation of the thermal response of the material and the damage threshold [47, 48]. Therefore, a more complete approach should be based on a rigorous approach of considering a dynamic change of the optical properties. Furthermore, the nonthermal electron presence is also expected to influence the variation of the dielectric constant [33], and therefore, relevant corrections to the dielectric constant values are necessary. On the other hand, pump–probe experiments demonstrate small variations for the dielectric constant of Ni [40]. Similarly, small variations for Ni have also been observed in a more recent work [49]. The noteworthy differences between the optical property behavior between noble metals and Ni may be due to the remarkably smaller electron heat capacity of copper with respect to the value of \( C_\text{c} \) for Ni. Therefore, in this work, the dynamic character of the optical parameters (i.e., absorption coefficient \( \alpha \), and reflectivity \( R \)) is ignored (see Table 1).

| Table 1 Parameters for Ni |
|---------------------------|
| Parameter | Value |
| \( C_\text{c} \) [J/m K] | Fitting [41] |
| \( C_\text{L} \) [J Kg\(^{-1}\) K\(^{-1}\)] | Fitting [50] |
| \( G_{cl} \) [Wm\(^{-1}\)K\(^{-1}\)] | Fitting [41] |
| \( A_\nu \) [10\(^7\) s\(^{-1}\) K\(^{-2}\)] | 0.59 [51, 52] |
| \( B_\nu \) [10\(^{13}\) s\(^{-1}\) K\(^{-1}\) cm\(^{-1}\)] | 1.4 [51, 52] |
| \( k_\text{so} \) [Jm\(^{-1}\)s\(^{-1}\)K\(^{-1}\)] | 90 [52] |
| \( T_\text{mel} \) [K] | 1728 [53] |
| \( T_\text{0} \) [K] | 300 |
| \( a' \) [10\(^{-6}\)K\(^{-1}\)] | 13.4 [53] |
| \( E \) [GPa] | 200 [53] |
| \( \nu \) | 0.31 [53] |
| \( \rho_\text{L} \) [Kg/m\(^3\)] | 8908 [53] |
| \( \varepsilon_\text{F} \) [eV] | 9.7 [53] |
| \( R \) | 0.74128 [54] |
| \( \sigma \) [10\(^7\) cm\(^{-1}\)] | 7.6851 [54] |
2.3 Elasticity equations

The mechanical response of the material is described by the differential equations of dynamic elasticity which correlate the stress and strain generation and the induced displacement as a result of the thermal expansion and the lattice temperature ($T_L$) variation.

\[
\rho_L \frac{\partial^2 V_i}{\partial t^2} = \sum_{j=1}^{3} \frac{\partial \sigma_{ij}}{\partial x^j}
\]

\[
\sigma_{ij} = 2\mu \varepsilon_{ij} + \lambda \sum_{k=1}^{3} \varepsilon_{kk} \delta_{ij} - \delta_{ij}(3\lambda + 2\mu)\alpha'(T_L - T_0) \tag{11}
\]

\[
\varepsilon_{ij} = \frac{1}{2} \left( \frac{\partial V_i}{\partial x^j} + \frac{\partial V_j}{\partial x^i} \right)
\]

where $V_i$ corresponds to the displacements along the $x$ ($i=1$), $y$ ($i=2$), $z$ ($i=3$) direction, $\sigma_{ij}$ are components of the stress tensor which is a $3 \times 3$ matrix; if $i=j$, $\sigma_{ij}$ is the orthogonal normal stress in the $i$-direction, otherwise $\sigma_{ij}$ is the orthogonal shear stress [55]; similarly, $\varepsilon_{ij}$ are components of the strain tensor which is a $3 \times 3$ matrix; if $i=j$, $\varepsilon_{ij}$ is the orthogonal normal strain in the $i$-direction, otherwise $\varepsilon_{ij}$ is the orthogonal shear strain; finally, $\delta_{ij}$ is the Kronecker delta function [56]. On the other hand, the Lame’s coefficients $\lambda$ and $\mu$ (for Ni, $\lambda = 1.25 \times 10^3$ and $\mu = 7.63 \times 10^3$), respectively, $\alpha'$ stands for the thermal expansion of Ni and $\rho_L$ is the density of the material. The Lame’s coefficients $\lambda$ and $\mu$ are related to the Poisson’s ratio ($\nu$) and Young’s modulus ($E$) through the relations $\nu = \lambda/(2(\lambda + \mu))$ and $E = \mu (2\mu + 3\lambda)/(\lambda + \mu)$.

2.4 Generalised energy balance equations

To describe the influence of the ultrafast electron dynamics on the relaxation procedure, and the thermomechanical response of the material, a revised version of the TTM is used that includes the early and transient interaction of the nonthermal electron distribution with the electron and lattice baths [29, 30]. Hence, the following set of equations is employed to investigate the spatio-temporal distribution of the produced thermalized electron ($T_e$) and lattice ($T_L$) temperatures of the assembly.

\[
C_e \frac{\partial T_e}{\partial t} = \nabla \left( k_e \nabla T_e \right) - G_{el}(T_e - T_L) + \frac{\partial U_{ee}}{\partial t}
\]

\[
C_L \frac{\partial T_L}{\partial t} = G_{el}(T_e - T_L) - (3\lambda + 2\mu)\alpha' T_L \sum_{j=1}^{3} \varepsilon_{jj} + \frac{\partial U_{el}}{\partial t} \tag{12}
\]

where the subscripts $e$ and $L$ are associated with electrons and lattice, respectively, $k_e (= k_{\text{el}} BT_e/(A_e(T_e)^2 + B_e T_e))$ is the thermal conductivity of the electrons, $C_e$ and $C_L$ are the heat capacity of electrons and lattice, respectively, and $G_{el}$ is the electron–phonon coupling factor. The parameters $C_e$ and $G_{el}$ are taken from Ref. [41]. (i.e. by using a fitting procedure).

The last additive source terms in Eqs. (12) account for the energy density transfer from the TH distribution to the NTH electrons ($\partial U_{el}/\partial t$) and lattice ($\partial U_{el}/\partial t$), respectively. Therefore, thermalisation dynamics of the NTH electrons is incorporated into the model in the “source terms” to describe: (1) the generation of the thermalized electron subsystem and (2) the interaction of NTH electrons with lattice. The two source terms include the features of the excitation pulse but they also incorporate the relaxation of the NTH electron population through Eq. 8 [29].

3 Numerical solution

Due to the inherent complexity of Eqs. (1–12), an analytical solution is not feasible, and therefore, a numerical approach is pursued. Numerical simulations have been performed using the finite difference method while the discretization of time and space has been chosen to satisfy the von Neumann stability criterion. Furthermore, on the boundaries, Neumann boundary conditions are applied while heat losses at the front and back surfaces of the material are assumed to be negligible. The initial conditions are $T_e(t=0) = T_L(t=0) = 300 \text{ K}$, while stresses, strains, and displacements are set to zero at $t=0$. Furthermore, the vertical stress $\sigma_{ee}$ of the upper surface is taken to be zero at all times (i.e., stress free). The parameters for Ni used in the simulation are summarised in Table 1. The values of the laser beam features used in the simulation are: the (peak) fluence is $E_p = I_0/(2\sqrt{\ln 2})$, where $I_0$ stands for the peak value of the intensity, spot radius $R_0$ (where the intensity falls to $I/e^2$) is equal to $15 \mu \text{m}$, and pulse duration values lie in the range [10 fs, 2.3 ps]. The wavelength of the beam is $\lambda = 800$ nm. We note that, for the sake of simplicity, in the first part of this work, the laser beam conditions are selected so that they: (1) emphasise the role of the nonthermal electrons and (2) are insufficient to induce material melting or plastic deformation. Hence, only elastic displacements are assumed. Thus, three different values of the fluence, $E_p = 20$, 40, and 60 mJ/cm$^2$, were chosen to simulate the mechanisms that follow the above requirements.

In principle, a common approach followed to solve problems that involve elastic displacements [55] or hydrodynamics [10] is the employment of a staggered grid finite difference method which is found to be effective in suppressing numerical oscillations. Unlike the conventional finite difference method, temperatures ($T_e$ and $T_L$) and normal stresses $\sigma_{ii}$ are computed at the centre of each element.
while time derivatives of the displacements and first-order spatial derivative terms are evaluated at locations midway between consecutive grid points. Furthermore, shear stresses $\sigma_{ij}$ are evaluated at the grid points. Numerical integration is allowed to move to the next time step provided that all variables at every element satisfy a predefined convergence tolerance of $\pm 0.1\%$.

Furthermore, in the second part of this work, to explore conditions that lead to material damage (i.e., $T_L > T_{melt}$), a thermal criterion is applied to determine the damage threshold. In that case, Eqs. 12 are solved for $E_p = 80$ and $120$ mJ/cm$^2$ by ignoring strain development. We note, that in a previous work, melting and disintegration of Nickel films were explored after irradiation with ultrashort pulses by using a combined atomistic-continuum modelling [38]. Certainly, a revised model that incorporates rTTM and atomistic simulations could allow a more precise damage threshold estimation, however, it is outside the scope of the present study to focus on this aspect.

### 4 Results and discussion

To determine the role of the nonthermal electrons in the heat exchange and relaxation process, it is important to provide a computed estimate of the energy density per unit time stored in the nonthermal electronic distribution. Figure 3 illustrates the absorbed power density (the only source term that appears in the traditional TTM) and the source terms $\partial U_{ee}/\partial t$ and $\partial U_{el}/\partial t$. It is evident that the traditional TTM overestimates the amount of internal energy for thermal electrons due to the fact that the formation of a nonthermal electron distribution is not included in the description (Fig. 3). Simulation results illustrated in Fig. 3 indicate that compared to the TTM, the revised model predicts a delay of the heating process of both the TH electrons and the lattice. More specifically, the maximum power density transferred to the thermal electrons and lattice systems (dashed and solid lines, respectively) is shifted compared to whether it is assumed that instantaneous electron thermalisation occurs (dashed–dotted line). It is evident from the temporal profile of the heat sources that the electronic dynamics is dominated by electron–electron scattering in the initial stages. On the other hand, compared to noble metals such as Au and Cu [29, 30, 35], there exists a gradually significant nonthermal electron–lattice interaction for Ni which is attributed to the large electron–phonon coupling constant.

In contrast to pulse duration variation that is expected to influence the aforementioned power densities, variation of fluence is not expected to yield different values for the ratios between these quantities, and therefore, the shapes of the predicted temporal evolution are the same as those in Fig. 3. This is attributed to the fact that the fluence enters the power densities as a multiplicative coefficient and thereby it is cancelled out when relevant ratios are computed.

The solution of Eqs. 12 allows the investigation of the thermal response of the system through the analysis of thermalisation process and the evolution of $T_e$ and $T_L$. A comparison of the maximum surface electron and lattice
temperatures as a function of time (for four different pulse durations, $t_p = 10$, 50, 170, and 1.5 ps) for $E_p = 40$ mJ/cm$^2$ simulated with the traditional TTM and rTTM is presented in Fig. 4. By comparing the two models, it is evident that the electron temperature peak is reached with a short delay with respect to the value attained if the TTM model is employed. This consequence is ascribed to the erroneous assumption of an instantaneous creation of thermal electrons and heating process of the electron and lattice baths predicted from the TTM model; in practice, there is a finite time required for the creation of the nonthermal electrons which leads to a delayed heating of both the thermal electrons and lattice (Fig. 5). This argument is also supported by the temporal evolution of the power densities (Fig. 3) and it is evident

Fig. 4 Electron and lattice evolution for $t_p = 10$ fs (a), 50 fs (b), 170 fs (c), and 1.5 ps (d) derived from rTTM and TTM. ($E_p = 40$ mJ/cm$^2$, 800 nm laser wavelength, $R_0 = 15$ µm)

Fig. 5 Maximum electron temperature evolution computed using rTTM (solid line) or TTM (dashed line) for $t_p = 10$ fs (a), 50 fs (b), 170 fs (c), 1.5 ps (d). Vertical dotted line indicates the heating delay of the electron system. ($E_p = 40$ mJ/cm$^2$, 800 nm laser wavelength, $R_0 = 15$ µm)
that the delay is more pronounced for small pulse durations rather than longer excitation periods where the delay disappears ($\tau_p = 1.5 \text{ ps}$). A similar behavior is exhibited at different values of the fluence value ($E_p = 20$ and $60 \text{ mJ/cm}^2$) (see Supplementary Material). The employment of the revised model indicates that the incorporation of the contribution of the nonthermal electrons lowers the electron temperature. The decrease of the electron temperature is a physical consequence of the elastic energy loss due mainly to the scattering of the nonthermal electrons from the electronic bath. By contrast, a comparison of the $T_e$ and $T_L$ evolution curves demonstrates that despite the delayed heating of the electron and lattice systems, respectively, electron–phonon relaxation is expected earlier if the rTTM is used (Fig. 4). On the other hand, the electron–phonon interaction accounts for the increase of the lattice temperature, which is further enhanced by the term that describes the interaction of the nonthermal electrons with the lattice (i.e., $\partial U_{el}/\partial t$). The induced smaller $T_e$ predicted by rTTM implies a stronger electron–phonon interaction (i.e., Nickel is characterised by an increasing smaller electron–phonon coupling constant at smaller $T_e$ [41]) which results in a faster relaxation process compared to predictions from the traditional TTM (Fig. 4).

In Fig. 6, the TH electron population internal energy density $U_{TH}$ is also computed by means of the expression $C_e = n_e \partial (U_{TH}) / \partial T_e$, ($n_e$ is the free electron density, while $C_e$ is computed from Ref. [41]) if TTM or rTTM are used (solid line or dashed line) as a function of the pulse duration [57]. It is evident that the energy stored in the NTH electron population is larger if the classical TTM is employed which underlines the overestimation due to the traditional approach. While TTM predicts an instantaneous thermalisation of the electrons, rTTM shows that the energy of the TH electrons (and therefore their population) is smaller due to the formation of a NTH population that exchanges energies with both the TH electrons and the lattice. Therefore, at later times (Figs. 4, 5, 6) the TH electron population relaxes to the one predicted by TTM.

Thermal response of the system has been, also, quantified for various pulse duration values in the range [10 fs, 2.3 ps] for $E_p = 20$, 40, and $60 \text{ mJ/cm}^2$, and the predicted values of the maximum electron and lattice temperatures are illustrated in Fig. 7. The decrease of the maximum electron temperature with increasing pulse duration is illustrated in the graph, while a small increase of the lattice temperature is produced. It is noted that the maximum lattice temperature discrepancy between the values predicted by TTM and rTTM increases for larger fluences. It is evident, though, that the computed maximum lattice temperature is below $T_{\text{melt}}$ that shows surface damage does not occur for the laser beam conditions of the simulations.

Furthermore, variation of the lattice temperature induced by the absorption of the ultrashort optical pulse leads to mechanical effects due to the generation of thermal stresses. On the other hand, thermal stress produces strain generation and propagation. The spatio-temporal strain/stress pulse shape is determined by the solution of Eqs. 11, 12. To emphasise on the differences of the magnitude and spatial distribution of the strains and stresses predicted by the rTTM and TTM, the components of these fields along the direction of energy propagation ($z$-axis) are calculated and illustrated in Figs. 8 and 9, respectively.

**Fig. 6** Internal energy density stored in the TH electron population computed from TTM (solid line) and rTTM (dashed line) as a function of pulse duration ($E_p = 40 \text{ mJ/cm}^2$, 800 nm laser wavelength, $R_0 = 15 \text{ µm}$).
The strain $\varepsilon_{zz}$ is always positive at $z = 0$ due to the fact the stress-free boundary condition does not imply any stretching perpendicular to the free surface. Furthermore, the solution of the second-order (with respect to time) differential equation for the displacements (first equation of Eq. 11) yields a two-term propagating part, one showing a positive strain and a second with symmetric negative strain (result of the reflection on the surface). The two terms exhibit an exponential decay with length equal to $l/\alpha$. The exponential decay is reflected also on the stress fields which offer a more accurate calculation of the exponential decay length (Fig. 9). Similar results for the strain pulse propagation have been presented in the previous works for picosecond light pulses [58, 59]. Furthermore, reflectivity changes due to strain generation after ultrashort-pulsed laser irradiation of thin films on silicon surfaces have been also recently investigated [37, 60]. Comparing the strain values at $z = 0$ with the results predicted in previous works [58, 59], the nonconstant $\varepsilon_{zz}(z = 0, t)$ at all times is attributed to the fact that in the current simulations, lattice temperature rise after irradiation neither occurs instantaneously nor remains constant.

It is evident by estimating the spatial position of the lowest strain or stress values at different times (Figs. 8, 9) that the strain and stress pulses propagate at a speed equal to approximately, 5578 m/s which corresponds to the longitudinal sound velocity in Ni ($= \sqrt{(2 \mu + \lambda)/\rho_l}$ [59]). The pulses are illustrated at four different timepoints, $t = 8, 15, 20$, and 30 ps, after the arrival of the laser pulse on the surface of the material. In addition to $E_p = 40$ mJ/cm$^2$, simulations have been performed for $E_p = 20$ and 60 mJ/cm$^2$ where similar results (from a qualitative point of view) are deduced.

The comparison of the $\varepsilon_{zz}$ and $\sigma_{zz}$ pulses using rTTM and TTM demonstrates first the temporal shift of the waves that result from the delay of the lattice heating predicted by the revised model. Furthermore, the shapes of the strain and stress pulses derived from the two models do not appear to be significantly different at large timepoints. By contrast, there exists a larger than 20% increase in both the strain and stress size in a region near the surface at small times after laser irradiation (see Figs. 8, 9). The deviations are attributed to the enhanced lattice temperature values produced by the nonthermal electron interaction with lattice which is very pronounced at small timepoints while relaxation to values comparable to the ones predicted by the TTM is expected at bigger times (Fig. 4).

An experimental validation of the proposed mechanism is required to test the adequacy of the theoretical model, however, the scope of this work is primarily related to the introduction of a consistent theoretical framework that will take into account: (1) the energy balance between the thermal electron and the lattice baths enriched with the contribution of nonthermal electrons, (2) the influence of the DOS around the Fermi energy (Ni is characterised by a nonconstant DOS around $\epsilon_F$), and (3) the thermomechanical response of the material due to material heating. Nevertheless, theoretical investigation of the thermomechanical response of the material and comparison of the simulation results with experimental observables in previous works where a simpler version of the model was used [37, 60] confirm the primary importance and validation of the proposed underlying physical mechanism of laser matter interaction and associated processes. On the other hand, by ignoring the mechanical response of the system, previous works that assume a revised TTM based on the inclusion of nonthermal electron interaction with both thermalized electrons and lattice and the introduction of correction terms in the source
term (as presented also in Ref. [29]) show a satisfactory agreement with results in pump–probe experiments [32–36] in noble metals with small \(G_{el}\).

The above description indicates that single-shot laser irradiation at low fluence does not influence the strain/stress fields shape (at larger times) if the role of the nonthermal electrons is taken into account. Nevertheless, the picture is expected to alter drastically in different laser beam conditions. For example, in multiple shot experiments of small temporal delays between the subsequent pulses (i.e., train-pulse technology), the approximately 20% variation of the strain wave amplitude or the substantially smaller electron temperature produced due to excitation could influence (through accumulation effects) surface micromachining.
techniques and applications. Furthermore, the significant deviation of the magnitude of the strain fields at small time-points (i.e., ~ 8 ps) could also influence mechanical properties of bilayered materials (for example, thin films on silicon surfaces [37, 60]) where strong acoustic waves are expected to be reflected on the interface and interfere strongly with the propagating strain leading to a more complex total strain.

The aforementioned simulations aimed, first, to underline the differences between theoretical predictions TTM and rTTM in conditions that do not induce phase transition. It is important to emphasise on the significant impact of the discrepancy between the predicted maximum lattice temperatures derived from the two models. This prospect is expected to have a very important impact on material properties and industrial applicability in terms of capability to modulate laser parameters as it will provide a more accurate and precise range of fluences to avoid surface damage. Regarding the employment of the model to explore mechanisms related to the onset of damage, simulations have also been performed for fluences that lead to material melting. More specifically, the application of the rTTM model indicates that for $E_p = 80$ or 120 mJ/cm$^2$, the maximum lattice temperature exceeds $T_{melt}$ which suggests that material melts and surface modification occurs (Figs. 10, 11). Maximum electron and lattice temperature dependence on pulse duration is the same as at lower fluences (Fig. 7), however, the temperatures reached are higher and the discrepancy predicted by TTM and rTTM increases at larger laser energies (Fig. 10). A comparison of the computed maximum lattice temperatures for all values of $E_p$ demonstrates that the maximum lattice temperature predicted using the revised model (rTTM) is always higher than that computed by means of TTM (Fig. 11). It is evident that the deviation of the maximum $T_L$ predicted from the revised model and the classical TTM increases as the fluence increases. This outcome is expected at higher energies, as a larger population of nonthermal electrons is produced, and therefore, their influence is larger. An estimation of the fluence that leads to temperatures larger than $T_{melt}$ derived from rTTM and TTM are 67 and 99 mJ/cm$^2$, respectively. The 33% difference in the damage threshold determination is significant and it reveals the important thermal effects to the material due to the consideration of direct interaction of the nonthermal (hot) electrons with the lattice.

A more conclusive exploration of the role of the pulse duration (and fluence) in the determination of the damage thresholds (i.e., characteristic for medium and high fluences) requires a thorough investigation of the underlying physical processes. More specifically, in photoexcitation by infrared (IR) and visible range for fs/ps laser pulses, relaxation phenomena, heat conduction and (possible) two-photon interband infrared-absorption are expected to influence the damage thresholds [46, 61, 62]. Furthermore, a transient evolution of the DOS should be also taken into account while a more rigorous estimation of the optical properties of the irradiated material during the pulse duration should be evaluated.

Certainly, an experimental confirmation of the theoretical findings of the present work is necessary to validate the damage thresholds predicted by the model; nevertheless, the predicted substantial decrease of the damage threshold with respect to the estimation by means of the conventional TTM emphasises on the significant role of the physical processes in the early stages of irradiation.
which should not be neglected. The theoretical framework can also be revised to describe more efficiently the underlying physics for medium and high fluences (where effects related to band filling, considerable interband transitions, possible two-photon interband IR-absorption and induced changes in the relaxation times should be taken into account) towards estimating more accurately the damage thresholds. Nevertheless, the proposed framework suggests that the rTTM could be incorporated as a complementary module in a multiscale framework that takes also into account electron thermalisation mechanisms as well as processes at longer time-scales (i.e., phase transitions, fluid transport, solidification, ablation, etc).

5 Conclusions

A detailed theoretical framework was presented that describes both the ultrafast dynamics of electrons and the induced strains and stresses in metals with strong electron–phonon coupling constant and complex DOS after irradiation with ultrashort pulsed lasers. The revised TTM incorporates the interaction of the nonthermal electrons with both the thermal electrons and the lattice while an additional component is included to describe the thermomechanical effects. A parametric analysis was performed for a range of pulse duration and fluence values that shows the differences from the predictions of the classical TTM. It is demonstrated that the employment of the revised TTM yields remarkably large values (~20%) for the induced strains especially at small timepoints after irradiation which is expected to be of paramount importance depending on the laser processing techniques (i.e., laser-pulse train processing). On the other hand, simulation results indicate also that the proposed underlying physical mechanism leads to a substantially (~33%) lower damage threshold for the irradiated material. This a very useful aspect from an industrial point of view towards estimating a more accurate damage threshold that is very significant for laser manufacturing approaches.

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