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Modeling saturable absorption for ultra short X-ray pulses

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ABSTRACT

Saturable absorption was recently observed in transmission measurements above the $L_{II,III}$ edge of pure Al thin films using ultra short X-ray pulses at a free-electron-laser (FEL) facility. The high fluence reachable by FEL pulses, the shortness of the pulse duration, and the typical lifetime of the excited state are all important factors enabling observation of the phenomenon. We devised a simplified theoretical approach describing the saturation phenomenon using a three-channel model containing ground, excited and relaxed states. This phenomenological model explicitly includes the interaction between the solid and photon field in a semi-classical way, and the resulting non-linear coupled equation is solved numerically. We successfully applied this model to recent experimental results obtained using FEL radiation.

1. Introduction

Saturable absorption is the effect of increasing transmittance of photons for increasing intensity, and is a well-known phenomenon for the visible and near-visible region of the electromagnetic spectrum, exploited also in laser technology [1,2]. In fact, the Lambert–Beer law is based on the fact that each photon absorption event is independent and involves excitations of electrons from the same ground state. In particular, this implies that photons should not cause optical saturation or optical pumping, since such effects deplete the lower level and possibly give also rise to stimulated emission. A serious depletion of the lower photon-absorbing level can be obtained using photon field intensities sufficiently high to overcome relaxation from the upper level.

Saturation in the occupation of the available states can be obtained using ultrashort pulses of photons of enough intensity which induce changes in the observed photon absorption cross-section. However, the description of photon-matter interaction at high intensities requires specific models to be devised, accounting for the various effects contributing to a modification of the cross-section (relaxation of final state, ultrafast electron heating, and so on).

The importance of developing proper models for such experiments is epitomized by the results of Nagler et al. [3], obtained using the free-electron laser in Hamburg (FLASH), generating subpicosecond soft X-ray pulses with intensities up to and in excess of 10^{16} W cm^{-2} ($\sim$ 200 J/cm^{-2} for each pulse). In that work [3], saturable soft X-ray absorption of an ultrathin aluminum foil was observed, reaching deposited energies which allowed creation of highly uniform warm dense matter conditions, a regime exceedingly difficult to reach in laboratory studies, but of great interest in various fields including high-pressure and planetary science, astrophysics, and plasma production.

As mentioned above, warm dense matter (WDM) at electron temperatures in the 1–10 eV range can be generated by using the FEL radiation and various ultrafast techniques can be used to probe WDM properties in that regime, exploiting the time structure of the pump and probes. The unique intensity, energy domain and time structure of the FELs can be used to probe metastable and/or excited matter under extreme conditions. The availability of a tunable FEL radiation in the ultra-violet (UV) and soft X-ray ranges like Flash (Hamburg) [4] and Fermi@Elettra (Trieste) [5] and in the hard X-ray range like LCLS (Stanford) [6], SACLA (Spring-8) [7] and the future XFEL (presently under construction, Hamburg) gives us an extraordinary experimental opportunity [8] for probing microscopic properties of dense matter under extreme conditions. The new experimental possibilities offered by FELs open the way to forefront research in theoretical and computational simulations oriented to develop and understand the results of spectroscopy techniques used at these sources. An open and fundamental problem is certainly the development of suitable models for saturable absorption, which also give information on how the photon energy is deposited in the solid.

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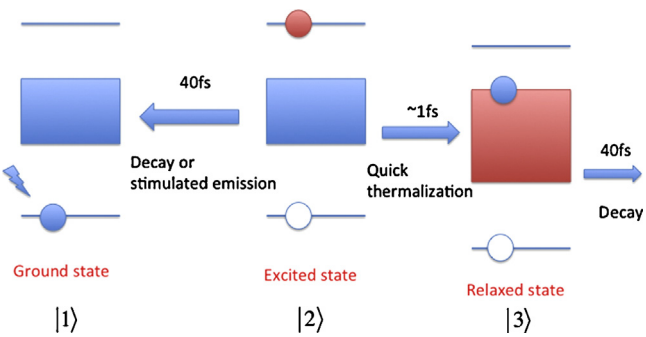

Fig. 1. Three channel model as described by Eqs. (1)–(3). In the present specific application the excited and relaxed states decay to the ground state with tyical lifetimes $\tau\sim$ 40 fs [21], while the decay to the relaxed transient state is much faster ($\tau_{23}\sim$ 1 fs). In fact, thermalization of electrons occurs by intrinsic and extrinsic losses due to multiple-scattering of photoelectrons, with a typical mean free path of about 5–10 Å(corresponding to relaxation times of 1 fs). We assumed that the lifetime of the 2*p* hole in transient state is the same in the excited and relaxed transient states.

A reliable theoretical model is needed for describing the dynamical processes occurring during the X-ray pulse absorption, and to estimate the temperature of the system. There have been several theoretical works on Al VUV/X-FEL intense pulse absorption. Examples range from a "dynamical" model (small cluster) [9,10] with incoherent approximation [11] which has the interaction and feedback with classical photon field, to a "steady" model [12,13] which neglects the feedback. In this communication we briefly describe a simplified approach describing the saturation phenomenon using a three-channel model containing ground, excited and relaxed states. This phenomenological model is conceived to provide a reliable calculation scheme to study the transmittance as a function of the fluence and the interaction dynamics considering explicitly the size effects of the pulse width and thickness of the absorbing medium within the nanometric scale.

2. Theory

The optical absorption process can be described starting from the quantum Liouville equation which provides a standard and rigorous way of treating nonlinear problems of laser optics. In VUV or X-ray energy region, due to their high frequency, we can introduce the conventional Markov approximation [9,14] which neglects the coupling between diagonal and off-diagonal components of the transitions of the equation, thus simplifying the treatment. By introducing this approximation, we end up in the conventional rate equation coupled with all possible excited states [9]. In our application we further simplify this model using a minimal set of channels, still describing the saturation effect in an effective way for the VUV-XFEL regime. We will consider only three states: ground $|1\rangle$, excited $|2\rangle$ and an intermediate relaxed state $|3\rangle$. Each state is a many-body one (not just a single one-electron orbital). In our model, the relaxed state $|3\rangle$ is thought to represent the ensemble of all possible relaxed states. This three channel model is schematically shown in Fig. 1. In the following discussion we employ a set of semiclassical phenomenological equations, which is commonly used for the study of non-linear optics (see ex. Section 5 of Ref. [1] or Section 4 of Ref. [2]).

The set of rate equations for the variation of the occupation numbers N_1, N_2 and N_3 (for the three states) are then reduced to:

$$\frac{dN_1(z,t)}{dt} = \frac{a(z,t)I(z,t)}{\hbar\omega} + \frac{N_2(z,t)}{\tau_{21}} + \frac{N_3(z,t)}{\tau_{31}} \tag{1}$$

$$\frac{dN_2(z,t)}{dt} = -\frac{a(z,t)I(z,t)}{\hbar\omega} - \frac{N_2(z,t)}{\tau_{21}} - \frac{N_2(z,t)}{\tau_{23}} \tag{2}$$

$$\frac{dN_3(z,t)}{dt} = \frac{N_2(z,t)}{\tau_{23}} - \frac{N_3(z,t)}{\tau_{31}} \tag{3}$$

where

$$a(z,t) = \sigma\left(N_2(z,t) - \frac{d_2}{d_1}N_1(z,t)\right) \tag{4}$$

$$N = N_1(z,t) + N_2(z,t) + N_3(z,t) = const. \tag{5}$$

$a(z, t)$ is a generalized form of the absorption coefficient, for linear absorption process as Lambert–Beer, it must be a constant. The occupation numbers depend thus on the photon field intensity $I(z, t)$ at time t and position z, the photon absorption cross-section σ at given photon energy $\hbar\omega$, and on the relaxation times τ between the various states. d_2/d_1 is the degeneracy ratio of the states. These equations are coupled with the transport equation of the incoming laser pulse, within the classical electrodynamics limit:

$$\frac{dI(z,t)}{dz} + \frac{1}{c}\frac{dI(z,t)}{dt} = a(z,t)I(z,t). \tag{6}$$

Within this model, the absorption and stimulated emission by laser radiation is related to the transitions between the ground $|1\rangle$ and excited $|2\rangle$ states. While the relaxed state $|3\rangle$ does not participate to these processes, it can be reached by the decay from state $|2\rangle$, and it can decay to state $|1\rangle$ by emitting a photon or through other processes. Eq. (4) describes the change in the absorption coefficient due to the variation in the occupation number of ground, N_1, and excited, N_2, states, missing in the so-called Lambert–Beer law. The change in occupation numbers described in Eqs. (1)–(3) is assumed to follow a simple sum rule for which the number of total states is preserved during the excitation process (see Eq. (5)).

This picture can be reasonable for ultra-short pulses (less than 1 ps), since for longer pulsewidths, one should consider more complicated physical processes, such as transmission of temperature from hot electrons to phonons following a natural thermalization process.

The simple dynamical absorption model described above has been implemented in a computer code, which is able to calculate various interesting measurable quantities as a function of photon energy, fluence, thickness and composition of a thin absorbing film (pictorial view sketched in the left side of Fig. 2). In this simple model, the laser pulse travels and interacts with a target. Inside the target absorption, stimulated emission and decay occur, and this changes the shape of the pulse along its way through the film. The program can treat any material composition, including the possibility of including oxidized layers in both sides of target which is often realistic. In the right panel of Fig. 2, it is presented an example of the computed result for Al thin film which will be discussed later. For low pulse fluences, linear transmission (Lambert–Beer) is obtained as shown by black line decaying exponentially with the thickness, while for high intensity there is a non-linear transmission and the decay tends to be linear with the thickness of the target as shown by blue line. The transmission can vary with the position of the pulse, namely the forefront part decays as Lambert–Beer, since interacting electrons in target are all in their ground states ("cold"), while the tail part (depending on the number of photons, pulse width and relaxation times) find electrons filling the excited states ("hot"), so that photons can be only absorbed through different mechanisms.

An important quantity related to the photon absorption phenomenon is the deposited energy, that in the high fluence limit is able to induce the so-called warm dense matter conditions. In our model, the energy is transferred to the valence electrons that are considered to be nearly free electrons within this regime.

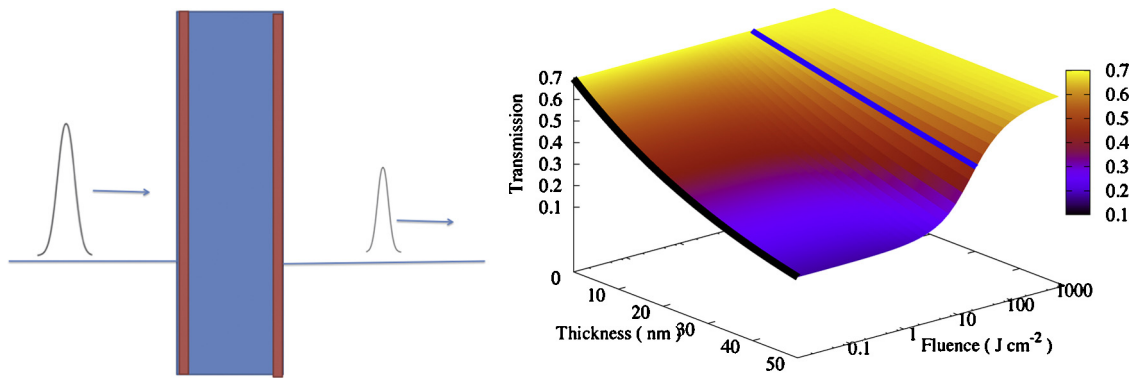

Fig. 2. The left panel is a sketch of the computational model. The incident Gaussian shaped FEL pulse comes from the left, then interact with macroscopic target and go to the detector to the right. The phenomenon is treated in one dimension. During traveling inside target, absorption, stimulated emission and decay take place at any point of the target dynamically with time. Oxidized layers are set in both sides of target. The right panel shows an example of computed results for Al thin film discussed later. For lower limit of fluence the transmission decays exponentially as Lambert–Beer law (black line), while for the high fluence after the saturation threshold it decays linearly (blue line). (For interpretation of the references to color in this figure legend, the reader is referred to the web version of the article.)

The total absorbed energy per volume can be simply estimated by:

$$E_{abs} = (\hbar\omega - I)(\rho_{ph}^{in} - \rho_{ph}^{out}) = \frac{1}{L}\left(1 - \frac{I}{\hbar\omega}\right)\phi(0)(1-T) \quad (7)$$

where I is the ionization energy and $\hbar\omega$ is the energy of a photon. ρ_{ph}^{in} and ρ_{ph}^{out} are the density of number of photon per volume for incident and outgoing pulses, respectively, L is the thickness of the sample, $\phi(z)$ is the incident fluence at position z and the transmittance of the film is defined as, $T \equiv \phi(L)/\phi(0) = \rho_{ph}^{out}/\rho_{ph}^{in}$. We here suppose that the absorbed energy was distributed only to valence electrons while all the other electrons are frozen. In this way, the averaged energy per electron must be E_{abs} divided by the number of valence electrons n_e.

The effective electron temperature is estimated by using the Maxwell–Boltzmann distribution [23], where the average electron energy is related to the electron temperature T_e (measured in energy units):

$$\langle E \rangle = \frac{3}{2}\langle T_e \rangle. \quad (8)$$

A useful expression for evaluating the electron temperature T_e using transmission data of thin films can be thus retrieved using the above equations:

$$T_e = \frac{2}{3n_eL}\left(1 - \frac{I}{\hbar\omega}\right)\phi(0)(\alpha - T) \quad (9)$$

where α is a reduction factor taking into account all the residual photon absorption related to different phenomena not taken into account in the present model ($\alpha = 1$ being the ideal case) limiting the maximal fluence measured at the sample. The factor is useful for precise comparison with experimental data, which can contain spurious absorption sources or counting problems.

3. Results and discussion

As mentioned above, saturable absorption has been first observed for soft X-rays using FEL pulses by Nagler et al. [3], and some other preliminary studies have been recently published by other groups [15–18]. In the pioneering experiment of Nagler et al. [3], single-shot transmission data of a 53 nm Al foil were collected using 92 eV FEL ultra-short (15 fs) photon pulses up to fluences in the 200 J/cm^2 range. The energies of Al $L_{II,III}$ edge were 73.1 and 72.7 eV, respectively. The main contribution to the absorption comes from these core level excitations, since the absorption cross section of the core excitation is 100 times bigger than free-free absorption.[13] The kinetic energy of the excited electrons is thus about 20 eV, which is a continuum state.

We have applied the three-channel model presented in the previous section to describe the absorption phenomena as reported in Fig. 1, defining suitable ground $|1\rangle$, excited $|2\rangle$ and transient relaxed $|3\rangle$ states. Thermalization of the excited photoelectrons in continuum states occurs by intrinsic or extrinsic loss associated with multiple scattering [19] processes within the solid. While the intrinsic loss occurs within the typical excitation times, the extrinsic loss takes place within typical timescales related to the mean free path of the excited electrons. Since the kinetic energy of the excited photoelectrons is about 20 eV, the mean free path is in the range 5–10 Å[20], which corresponds to relaxation times of about 1 fs. The 2p hole can be filled through several processes involving the decay of the excited photoelectron (Auger process, fluorescence etc.). We neglect any appreciable change of the lifetime of the 2p hole in both excited and relaxed transient states, so the same relaxation time (40 fs) is used (corresponding to the 2p lifetime of the excited state from the ground state at 0 K [21]).

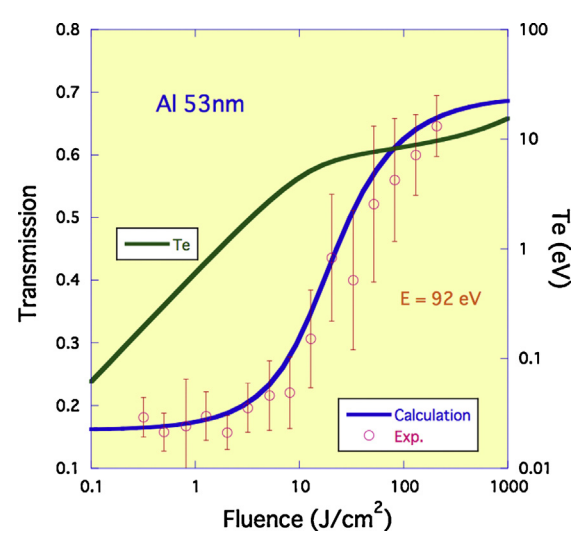

Fig. 3. Transmission of Al ultra thin film as a function of the incident fluence of a FEL pulse. The result of transmission measurements at the FLASH facility (see Ref. [3]) is compared with our calculation. The electronic temperature calculated with Eq. (9) is also shown with the axis in right side.

In Fig. 3 we compare the results of our calculations with the experimental data by Nagler et al. [3]. In the calculation, we have considered two Al_2O_3 oxidation layers for both sides of the Al film as shown in Fig. 2 (thickness 4 nm [22] each). The reduction factor α for incident fluence was set to fit the transmission in linear region in low fluence. The agreement between calculations and experimental data is very good. Possible temperature effects are not considered but it is reasonable to assume that they do not affect appreciably the observed trend. The non-linear effect can be thus explained as purely the result of optical non-linear saturation phenomena.

In the same figure (Fig. 3) we have reported the electron temperature of valence electrons, immediately after the irradiation. We neglected any electron–phonon energy exchange mechanism, since the duration of pulse width is much shorter than the time of thermalization via phonons. The calculation of T_e was performed using the simple relationship of Eq. (9). Temperature is shown to increase almost linearly until reaching the fluence saturation threshold. The temperature moderately increases upon saturation reaching values around 10 eV, almost the same as reported in the original paper [3].

4. Summary

We devised a simplified theoretical model which includes thermalization effects describing saturation phenomena for ultra-short pulse XFEL with high fluence. The rate equations for the electron occupation of states are coupled with the classical field dynamically with time at each position of a macroscopic sample, and they are solved numerically using a newly developed code. We applied the model to the transmission of a Gaussian shaped pulse with energy of 92 eV to ultrathin Al foil. We obtained a very good agreement between theory and experimental results by Nagler et al. [3]. We concluded that the non-linear saturation phenomena observed for transmission data of an Al film can be interpreted at the present level of accuracy by using the dynamics of the optical process without explicit inclusion of temperature effects.

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