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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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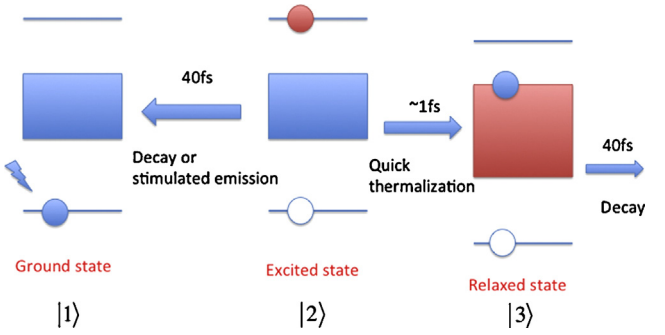

Fig. 1. Three channel model as described by Eqs. (1)–(3). In the present specific application the excited and relaxed states decay to the ground state with tyical lifetimes $\tau\sim 40$ fs [21], while the decay to the relaxed transient state is much faster ($\tau_{23}\sim 1$ fs). In fact, thermalization 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:

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

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

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

where

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

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

$a(z,t)$ 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:

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

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.

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