Theoretical Approach of Laser Excited Solids

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Abstract: This paper introduce the basic concepts of the theoretical description of laser- induced structural changes in solids. The approaches used are mainly based on the calculation of the time-dependent potential energy surface and an explicit treatment of electronic degrees of freedom.Key words: Workforce diversity, disability, old age, human resource management, equal opportunity.

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INTRODUCTIO

The physics and the resulting phenomena of laser excitation of materials can be summarized as follow: In most of the cases, intense femtosecond laser pulses are able to produce in solids a situation of extreme non equilibrium state in which electrons acquire very high temperatures while the lattice remains cold. This is due to the fact that on the time scale on which the laser interacts with solids, the laser energy is initially cou- pled with the carriers yielding to the creation of electron-hole pairs (the excited electrons and holes thermalize on a very short timescale [STvdL04]). The exchange energy between electrons and ions resulting from the relaxation process, takes place usually in some picoseconds after the laser excitation peak. This relaxation time is usually referred to as electron-phonon coupling time and causes delayed heating of the lattice. Thus, a significant increase of the lattice temperature is observed only within several picoseconds. Therefore the immediate change of the lattice structure after the laser excitation is entirely due to the presence of a hot electron-hole plasma and no thermal motions are involved. Furthermore, depending on the density of excited carriers and the laser characteristics, this extremely rapid excitation can lead to several structural transformations such as solid-liquid [SAPF96a, JHL+03, LLSTeA05], solid-solid [JGB99a, CTS+ 01, KEH+07, JGB01, CBRZ08, RTD+07, KTM03] phase transitions, and also generation of coherent phonons [PKKS92, SPK93, NHM+ 01, MMHK04, IKU06, ZTG06a].

In fact, the types of phenomenon which occurs after the laser excited solids can be di- vided into three main classes depending upon the laser characteristics. In the case of low laser energy, the interaction produces only an increase in the electronic pressure resulting to a volume expansion of the heated region while the cold region (unexcited part) is compressed (see below for the case of cerium). However depending on the symmetry of the studied system, the generation of coherent phonons is possible at low fluences (for example diamond structure of germanium [PKKS92, IKU06], rhombohedral struc- ture of bismuth [MMHK04, ZTG06b]). At high laser energies when a certain fluence threshold is exceeded, the interaction may give rise to ultrafast non-thermal melting [STBB+ 03, BBSS06, ZTG06b, ZWG+08] and ablation [RSTvdLA04].

The knowledge of time dependent electronically excited potential energy surfaces is essential for describing properly these different processes.

During the irradiation of an absorbing crystalline material, the incident optical energy is coupled to the carriers, which are excited from bonding (valence states) to anti-bonding states (conduction band states). Consequently, the inter-atomic forces are therefore mod-laser pulse

This scheme shows the effect of a generation of dense electron-hole plasma on the potential energy surface which governs the atomic motions. Situation before the action of the laser pulse: the solid is in thermodynamically equilibrium. For low temperatures, the electrons fill the states up to the Fermi level. The atoms are at the equilibrium positions of the ground state potential energy surface. The laser pulse changes the electronic occupations. This leads to rapid changes in the potential energy landscape. As a consequence, the lattice becomes unstable and forces appear on the atoms, driving a structural change.

In the ground state (electron temperature T0) electrons show a Fermi-like distribution. During the absorption of laser energy we assist to a non-equilibrium distribution followed by a rapid thermalization of carriers at temperatures much higher than the ground state temperature.

From the ideas mentioned above, one can conclude that a realistic description of laser heating of solids, must take into account explicitly the electrons as well as the atomic degrees of freedom. Thus, the link between electrons and

atomic structure for a given material has to be established in an explicit manner in order to consider properly the laser excitation effects.

From the ground state (electron temperature T0) to the excited states (electron temperature T), a non-equilib- rium state is created followed by a rapid thermalization process at a higher temperature T >> T0 .

Difficult problems to face when constructing theoretical models to study laser matter interaction. This is because many physical processes can be expected depending on the nature of the materials which are treated. Up to now there is no single model which can be used to describe all the physical phenomena resulting from femtosecond laser excitation of solids.

Recently, a theory using the two-temperature molecular dynamics model, which takes into account the electron dynamics, has been developed for metals [IZ03]. Although this improved method works well for metallic systems, it is not suitable for all the materials. First principle methods have been also used by Parrinello and coworkers [GMCP89, SAPF96b, VGCC96]. These methods are mainly based on the treatment of ions and electrons with density functional theory in the local density approximation. Within these models, an instant creation of electron hole plasma is assumed at time t = 0 so that the duration of laser pulse is set to zero ($\tau = 0$). The first principle methods do not allow an explicit time-dependent treatment of the laser parameters. These methods would not permit a study of materials in a range of laser intensities and durations. Moreover, mod- els based on first principle molecular dynamics simulation, are limited by the fact that they are computationally expensive and therefore do not permit to study systems with large number of atoms.

Thus, there is a need to develop methods which allow to explicitly include the different laser parameters (duration, intensities, fluences) to study the interaction of ultra-short laser with solids. An appropriate theory would also permit to handle the electronic non equilibrium states.

The basics approach used in the present study, is inspired from the pioneer works of Stampfli and Bennemann [SB90, SB92, SB94] and the recent works of Jeschke and Gar- cia [JG, GJ03, RGV+05]. The method proposed by Stampfli and Bennemann was very successful on describing the laser induce lattice destabilization of covalent systems silicon, carbon and germanium. The approach consist of analyzing the instabilities caused by the presence of hot electron plasma in terms of one or two phonons degrees of freedom by assuming that the entropy of the excited electrons remains constant during and after the laser excitation. Later on Jeschke and Garcia proposed an improvment of the model. The proposed model is based on tight binding molecular dynamics, thus the lattice dy- namics is described in real space. They used Born Oppenheimer approximation for the simultaneous integration of the equations of motion of electrons and ions. An explicit time-dependent treatment of the electronic occupation changes due to the laser excitation has been introduced. It allowed to explicitly handle the time- dependent laser parameters (pulse duration, intensity, absorbed energy), and study the effect of the variation of those parameters on the dynamics of the lattice.

Although the basic approach remains the same for the description of the electronic non- equilibrium states, the way to compute the potential energy surface which governs the dynamics is different from a particular system to another (depending on the type of elec- tronic structure, we have to deal with). Thus, the previous theoretical works need to be extended for the study of particular systems. We propose here different approaches to study the dynamics of cerium, samarium sulfide, bismuth and germanium upon femtosecond laser irradiation.

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