Interpretation of thermodynamic quantities in ab-initio non-equilibrium molecular dynamics

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Irradiation of solids with ultrashort laser pulses opened an exciting field of research. New emerging physics is connected with formation of warm dense matter (WDM) at the initial transient state of material evolution after energy deposition into electron subsystem. WDM in such ultrafast phenomena is a non-equilibrium state that makes it very challenging for theory, modelling and simulation, especially in two-temperature (2T) systems when electron and ion subsystems can be considered in quasi-equilibrium at $T_e > T_i$. Laser ablation is one of the major phenomena where WDM properties are crucially (e.g. [1,2]).

Both in continuum and in atomistic models of ablation it is assumed that the quasi-equilibrium 2T-WDM can be described using thermodynamic concepts. The free energy is represented as $F(\rho;T_i;T_e) = F_e+F_i$, where $F_e = E_b+F_{fe}$, where E_b is the binding energy, F_{fe} is the free energy of free (ionized, delocalized), F_i is the free energy of the ion subsystem. The corresponding representation of pressure is

$$P = P_e + P_i = P_b + P_{fe} + P_i \,. \qquad (1)$$

Usually the first term is called the cold or binding pressure and depends on density $P_b = P_b(\rho)$, the second term is the kinetic electronic pressure $P_{fe} = P_{fe}(n_{fe};T_e)$ and the third term is the thermal ionic pressure $P_i = P_i(\rho;T_i)$. It is the total pressure P that defines the mass transfer in continuum 2T-WDM models. In the atomistic 2T-WDM models sometimes the so-called blast force is introduced in order to describe the influence of kinetic pressure of electrons on the ion subsystem. The kinetic pressure of electrons is assumed to be a manifestation of free electrons. The difficulty connected with P_{fe} calculations stems from the fact that for this purpose one needs the number density of free electrons n_{fe} (i.e. the degree of ionization).

In this work we discuss two connected questions [3,4]. On examples of aluminum and gold using the finite temperature Kohn-Sham density functional theory we are making an attempt to analyze the electronic contribution to the total pressure in 2T-WDM metals and to clarify the representation (1). Another question is the separation of electrons into bound and free that is a general problem for non-ideal plasma physics.

The comparison of this approach with other techniques [5,6] will be discussed. As well as possible alleviation of these problems within the wave packets models for WDM [7,8].

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