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A Unified Concept of an Effective One Component Plasma for Hot Dense Plasmas

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Orbital-free molecular dynamics simulations are used to benchmark two popular models for hot dense plasmas: the one component plasma (OCP) and the Yukawa model. A unified concept emerges where an effective OCP (eOCP) is constructed from the short-range structure of the plasma. An unambiguous ionization and the screening length can be defined and used for a Yukawa system, which reproduces the long range structure with finite compressibility. Similarly, the dispersion relation of longitudinal waves is consistent with the screened model at vanishing wavenumber but merges with the OCP at high wavenumber. Additionally, the eOCP reproduces the overall relaxation timescales of the correlation functions associated with ionic motion. In the hot dense regime, this unified concept of eOCP can be fruitfully applied to deduce properties such as the equation of state, ionic transport coefficients, and the ion feature in x-ray Thomson scattering experiments.

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Matter in the universe is very often found in extreme states, at high pressure (> 1 Mbar) and high temperature (> 1 eV). Such conditions, relevant to planetary interiors [1], dwarf stars, and neutron star crusts [2], can now be reproduced in experiments using high-energy [3] and x-ray free-electron lasers [4] and are routinely met in inertial confinement fusion studies [5]. This hot dense plasmas (HDP) regime is an extension to high temperatures (\simeq keV) of the warm dense matter (WDM) concept [6], more focused on the transition between normal matter and plasmas. In both WDM and HDP regimes, atoms are partially ionized, electrons partially degenerate, and the Coulomb coupling is strong, leading to a liquidlike structure. There is no small parameter enabling a theoretical treatment in perturbation, and the physical description is usually provided by very demanding state-of-the-art quantum *ab initio* simulations. The theoretical description of HDP is a formidable challenge, since these methods reach their limits of applicability. Fortunately, the orbital-free method within a Thomas-Fermi formulation [7] extends to high temperatures the capability of quantum simulations. It is also desirable to rely on simple models in the first design and interpretation of experiments to setup large scale simulations. Such models have to be benchmarked against representative HDP simulations. Here we propose a unified concept of an effective one component plasma that fully describes the complicated nature of strongly correlated plasma without any free parameters. This model offers insights of fundamental focus in plasma physics and is relevant to research areas like astrophysics and fusion science.

The one component plasma (OCP) [8, 9] is a popular model which consists of a single species of ions immersed in a neutralizing background of electrons. Its static and dynamical properties depend on only one dimensionless parameter, the Coulomb coupling parameter $\Gamma = Q^2 e^2 / a k_B T$ where a is the Wigner-Seitz (ws) radius $a = (3/4\pi n)^{1/3}$, n is the ionic density, Q the ionization, e the fundamental charge, and T the temperature. Since the OCP model provides a formulation in which all its properties are either analytical or tabulated, it is used as a practical representation of Coulomb coupling in many situations encountered in hot dense plasmas although it represents a limiting situation in which the electrons are fully degenerate. Attempts to go beyond this simple model belong to the family of screened systems in which the bare coulomb interaction is replaced by a Yukawa potential [10], for instance. In the Yukawa model, a screening length is obtained within linear response theory in the small wavenumber k (long distance) limit for given values of ionization, temperature, and density [11]. In practice, the Yukawa model is deeply modified in the interpretation of x-ray Thomson scattering experiments by the introduction of short-range hard-core corrections that extends further than the first neighbors range [12, 13]. All these simplifying assumptions can obscure the diagnostic of the phenomena at play as is revealed by more realistic models [14–16] and recent experiments [17].

These approaches are not satisfactory for actual plasmas because ionization is not a well-defined quantity and the screening length definition is somewhat arbitrary. To provide a more realistic modeling of hot and dense plasmas, we have developed a simple finite temperature Thomas-Fermi orbital-free formulation coupled with molecular dynamics (OFMD) [7]. With the same inputs as the quantum molecular dynamics simulations

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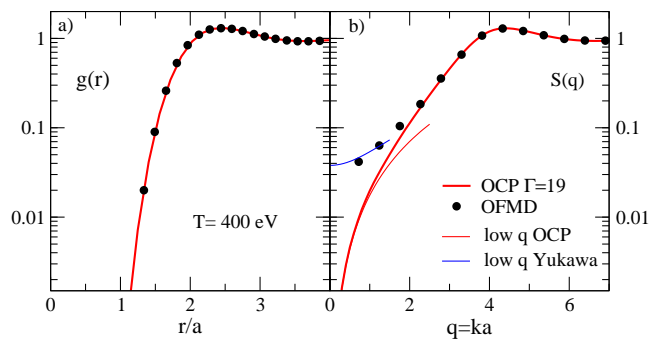


FIG. 1: a) OFMD pair distribution function for tungsten at 40 g/cm^3 and 400 eV (black points) compared with the OCP result (red solid line) at $\Gamma = 19$. b) Corresponding static structure factor. The blue line is the low q expansion of Yukawa structure factor using the effective ionization of the OCP fit. The thin red line is the OCP low- q expansion. Note the logarithmic scale to emphasize the differences at low wavenumber q .

with orbitals, *i.e.*, atomic number, density, and temperature, the OFMD simulations extend the range of accessible thermodynamic states without limits on temperature [7, 18–22]. A particularly interesting feature is the possibility to perform direct simulations of mixtures [23–29] to check the validity of mixing rules for thermodynamical [24] and transport properties [30].

In this Letter, we examine the relationship between the OFMD simulations of plasmas and the simple OCP or Yukawa formulations. We present a unifying concept combining the merits of both formulations. We give arguments supporting the use of the OCP model for the properties involving short-range correlations, including the equation of state [31] and the transport coefficients. We show that the quantities related to long-range correlations, forming the collective modes, such as the compressibility and the sound speed, are better reproduced by the Yukawa model once the ionization has been consistently defined. We have investigated two cases of very different atomic numbers relevant to the HDP regime: tungsten twice compressed between 100 and 5000 eV, and germanium at normal density between 100 and 800 eV. We used OFMD in the simplest formalism (Thomas-Fermi) for simulations. Relying on the well-known Thomas-Fermi scaling laws [31], we anticipate that our conclusions apply equally to any element in the HDP regime.

An interesting feature, evidenced in [19], is that the structure of the plasma, revealed by the pair distribution function (pdf) generated from OFMD simulations, can be precisely fitted by the OCP (see also [32]). This procedure defines the effective OCP (eOCP) with the effective coupling parameter $\Gamma_e = Q_e^2 e^2 / ak_B T$ and ionization Q_e . A similar procedure has been also invoked by Ott *et al.* [33] to characterize the coupling for Yukawa systems, which gives results very close to an adjustment

Elt	T	Γ_e	Q_e	Q_{TF}	θ	κ	T_{ω_p}
	eV						a.u.
W	100	19	12.7	14.2	1.9	2.1	580
	200	19	18.0	19.5	3.0	1.8	409
	400	19	25.4	27.9	4.7	1.5	290
	800	19	35.9	39.4	7.5	1.3	205
	1200	17	41.6	46.9	10.	1.1	177
	5000	10	65.1	67.3	31.	0.7	113
Ge	100	8	10.0	10.7	4.7	1.6	822
	200	8	14.1	15.4	7.5	1.3	583
	400	8	20.0	20.9	12.	1.1	411
	800	7	26.4	26.0	20.	0.9	312

TABLE I: Ionizations and plasma parameters for a tungsten plasma at 40 g/cm^3 and a germanium plasma at 5.3 g/cm^3 . $\theta = k_B T / E_F$ where E_F is the Fermi energy. κ is the inverse screening length at finite temperature in units of the ws radius a . Plasma periods $T_{\omega_p} = 2\pi / \omega_p$ are given in atomic units.

by hand. Ott’s method provides a quantitative basis to the effective OCP concept [34]. We show in Fig. 1a such an adjustment extracted from a series of simulations on tungsten at 40 g/cm^3 and between 100 and 5000 eV. We chose a temperature of 400 eV which is just in the region of the Γ -plateau where the structure is independent of the temperature [31, 35]. This peculiar evolution is due to the increase of ionization that compensates for the increase of temperature. It is worth noting that the structure is exactly the same with exchange (TFD) and gradient-corrected functionals [36], leading to the same effective coupling. Values of plasma parameters deduced from the eOCP analysis are given in Table I for tungsten at 40 g/cm^3 between 100 and 5000 eV, and germanium at 5 g/cm^3 between 100 and 800 eV. The details of the OFMD simulations and various formulas can be found in Supplemental Material [34].

We see in Fig. 1a for tungsten at 400 eV that the eOCP pdf at $\Gamma_e = 19$ perfectly matches the pdf obtained from OFMD. From the value of the coupling parameter Γ_e , we can deduce an effective ionization $Q_e = 25.4$ which appears to be 10% lower than an estimate within the average atom (AA) framework using the same Thomas-Fermi functional, Q_{TF} (see Table I). This suggests that the piling up of electrons around each ion is different in the OFMD and AA approaches, leading to different ionization and screening at short distance [37]. In any case, both approaches here account for the *nonlinear* contributions to screening close to the ions, contrary to the Yukawa model where screening is always considered within linear response. Within the eOCP model, the nonlinear screening *at short distance* is embodied in the effective charge Q_e . The good agreement between the eOCP and the OFMD results at short distance deteriorates at long distance (small $q = ka$) as revealed by the calculation of the static structure factor $S(q)$ shown in

Elt	T	Γ_e	P_{OFMD}	P_{eff}	P_{SESA}
	eV		Mbar	Mbar	Mbar
	100	8	85	80	80
Ge	200	8	229	217	221
	400	8	608	596	596
	800	7	1509	1551	1494

TABLE II: Equation of state of germanium at 5.3 g/cm^3 . P_{OFMD} is the pressure obtained by simulations, P_{eff} is the sum of the eOCP contribution and the electronic component as given by Nikiforov [42] (see Supplemental Material [34]) and P_{SESA} is the corresponding SESAME equation of state [43].

Fig. 1b. At vanishing q , $S_{\text{OCP}}(q)$ goes to zero as $q^2/3\Gamma$ [38] due to the long range of the Coulomb potential, whereas $S_{\text{OFMD}}(q)$ goes to a finite value proportional to the isothermal compressibility. Actually, screening effects must be introduced at long distance. Assuming a Yukawa pair potential with an inverse finite temperature screening length $\kappa = k_{\text{FT}}a$ [11, 39, 40], the resulting $S_{\text{Y}}(q)$ tends to a finite value as $(q^2 + \kappa^2)/(q^2 + \kappa^2 + 3\Gamma)$ at vanishing q . Using the effective charge Q_e as a definition of the ionization to compute the screening constant κ , the low q expansion of $S_{\text{Y}}(q)$ connects seamlessly with the OFMD results. For tungsten at 400 eV, the low q expansion of $S_{\text{Y}}(q)$ is given as a blue line on Fig. 1b. An extensive comparison with OFMD results will be presented in a forthcoming paper. This connection between OCP and Yukawa models through the definition of an effective charge is absent in traditional modeling where the ionization used to compute the screening length is left as a free parameter. The ionization is often assumed to be complete or deduced from an average atom calculation. Here we extract the effective charge from the static structure of the pdf. It can also be parameterized from a limited set of simulations using the Thomas-Fermi scaling laws. We left the presentation of this parameterization to a future paper.

A straightforward application of the eOCP concept concerns the equation of state. Very often, the ion thermal part is difficult to evaluate and is simplified or taken as an interpolation between the solid and the perfect gas. In the OFMD simulations this contribution is explicitly computed. In the eOCP approach the ion thermal contribution is constructed from analytical OCP fits [41] taken at Γ_e and the electron contribution from the corresponding finite temperature Fermi gas, as fitted by Nikiforov *et al.* [42], at the electronic density corresponding to Q_e . We show in Table II for the case of germanium that the sum of these two contributions P_{eff} agree to better than 10% with the SESAME equations of state [31, 43] or the present direct simulations with OFMD.

We turn now to the dynamical properties. It is well-known that the long-wavelength excitations of the charged versus neutral systems are notably different.

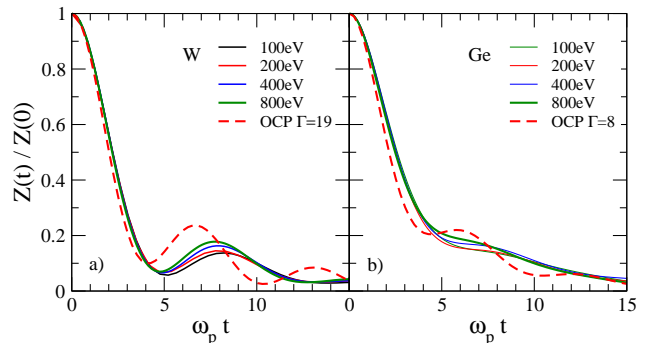


FIG. 2: a) OFMD velocity autocorrelation functions of tungsten at 40 g/cm^3 as a function of time in units of the inverse plasma frequency for each temperature, given in Table I, compared with the effective OCP one (red dashed line). b) Same as a) except for germanium at 5.3 g/cm^3 .

Baus [44] described these differences using a kinetic theory of the fluctuation spectra. The long range behavior of the Coulomb potential, with its singularity at $q \rightarrow 0$, is responsible for the various differences, here addressed using OFMD simulations. First, we consider the velocity autocorrelation function $Z(t)$ (VACF), which characterizes the individual motion and coupling with the collective modes. As such, it depends on correlation at both short and long ranges. Fig. 2a shows the VACF of tungsten between 100 and 800 eV in units of the inverse effective plasma frequency $\omega_p^2 = 4\pi n Q_e^2 e^2 / M$ of each case, which is ionization dependent (M is the ion mass). We observe that all VACFs are almost synchronized over a wide range of temperature, which reflects the Γ -plateau behavior. Notice that the short time behavior stays close to the eOCP. This indicates that the corresponding Einstein frequencies ω_E are close to the OCP values of $\omega_p/\sqrt{3}$. The relaxation timescales of the VACFs of eOCP and OFMD are comparable although the frequencies of oscillations around the average are different. The same behavior is also observed in Fig. 2b for germanium with weaker oscillations corresponding to a lower effective coupling parameter Γ_e . It is possible to get better agreement with the eOCP VACFs by a renormalization of the eOCP mass that depends on screening. This is beyond the scope of this paper and will be treated in a forthcoming paper.

The preceding analysis suggests that the eOCP concept can be used to predict transport coefficients by using standard OCP fits (see [30] and references therein) with the effective coupling parameter Γ_e . Both diffusion coefficients and viscosities are obtained from OFMD simulations by the Green-Kubo relations (see [45–47] for diffusion and [48] for viscosity). Good agreement for viscosity and diffusion for both tungsten and germanium is found with the eOCP formulation as shown in Fig. 3. Comparisons for plasmas of other species for such an approach

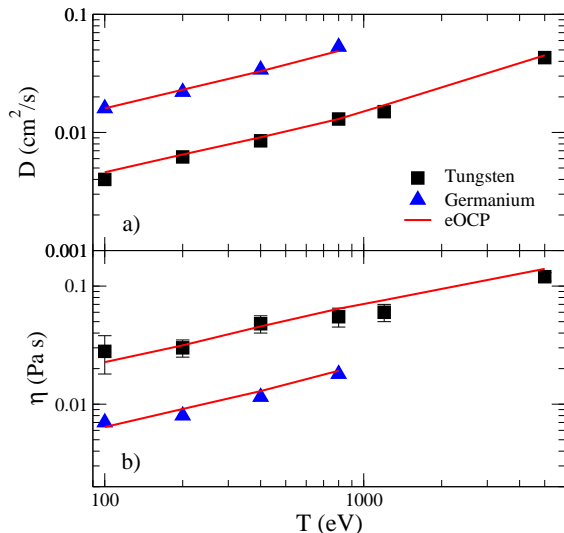


FIG. 3: Comparison between OFMD simulations (black squares for tungsten at 40 g/cm^3 and blue triangles for germanium at 5.3 g/cm^3) and effective OCP (red solid lines) for (a) diffusion and (b) viscosity.

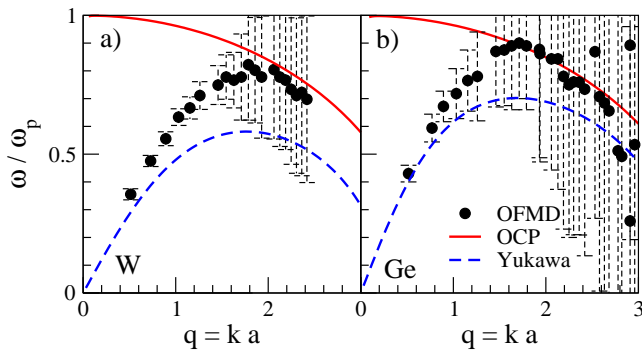


FIG. 4: Dispersion relations of longitudinal waves for a) tungsten at 40 g/cm^3 and 400 eV and b) germanium at 5.3 g/cm^3 and 400 eV . Black points: OFMD peak value with FWHM. The dashed blue line is the low q dispersion relation for Yukawa system in the quasi-localized charge approximation proposed by Rosenberg and Kalman [49], using the equation of state of Ref. [10]. The red line is the OCP dispersion relation at $\Gamma = 19$ for tungsten, and $\Gamma = 8$ for germanium, respectively.

using the TF ionization can be found in Ref. [30].

Finally, collective modes are evidenced by the dynamical structure factor $S(q, \omega)$ [50], which is of particular importance for x-ray scattering experiments. Here we focus on ion collective properties. The calculation of this quantity is well documented, and we follow White *et al.* [51] and Rüter *et al.* [52]. By collecting peak frequencies of the OFMD simulations of $S(q, \omega)$ versus q , and full widths at half maximum (FWHMs) of these features, we produce the dispersion relations shown in Fig. 4a for tungsten and Fig. 4b for germanium, which can be fitted by $\omega = c_s q/a$ at low q , yielding the sound speed c_s . As in the case of the static structure, we observe good agreement with the Yukawa dispersion relation at vanishing q . We used the relation proposed by Rosenberg and Kalman [49] within the quasi-localized charge approximation, which is particularly well adapted to the wave dispersion in strong-coupling situations [53]. For finite wavenumber (typically $q > 0.5$), the frequencies of the OFMD modes are slowly drifting out of the Yukawa curve and join smoothly with the eOCP values for $q > 1.5$.

To summarize, a unified concept for hot dense plasmas combining the OCP and Yukawa models is proposed. Its merits have been assessed using orbital-free molecular dynamics simulations in the hot and dense regime. The OCP and Yukawa models give complementary information about the simulated plasmas, providing a comprehensive description of their static and dynamical properties. The concept of an effective OCP connects these models through an effective ionization that is unambiguously defined. The eOCP facet is well adapted for short-range correlations and for a straightforward evaluation of the equation of state and transport coefficients. The properties related to the correlations at large distance, like the sound speed and the compressibility, need an explicit account of the electron screening. Here the Yukawa facet of this unified concept, based on an eOCP ionization, is a sensible approximation in this range where linear response theory applies.

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