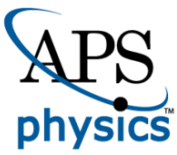

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Ionic Transport Coefficients of Dense Plasmas without Molecular Dynamics

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We present a theoretical model that allows fast and accurate evaluation of ionic transport properties of realistic plasmas spanning from warm and dense to hot and dilute conditions, including mixtures. This is achieved by combining a recent kinetic theory based on effective interaction potentials with a model for the equilibrium radial density distribution based on an average atom model and the integral equations theory of fluids. The model should find broad use in applications where non-ideal plasma conditions are traversed, including inertial confinement fusion, compact astrophysical objects, solar and extrasolar planets, and numerous present-day high energy density laboratory experiments.

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Progress in a wide range of research frontiers depends upon a detailed knowledge of the transport properties of warm and hot dense matter, characterized by densities near those of solids and temperatures from several to hundreds of eV. Important examples include the implosion of inertial confinement fusion (ICF) capsules [1], the structure and evolution of giant planets [2, 3], exoplanets [4, 5], and other compact bodies such as white dwarf stars [6, 7], as well as many new and future high energy density laboratory experiments [8, 9]. Modeling transport processes in dense plasmas presents a considerable challenge because of the combined effects of strong Coulomb coupling of ions and quantum degeneracy of electrons; these systems are too dense for standard plasma theories [10] and too hot for condensed matter theories to apply [11]. Experimental data are sparse and, to date, ab initio computer simulations have been the primary method to calculate transport properties [12–14]. Although these are often accurate, the approach is limited by its computational cost. In practice, modeling the macroscopic evolution of high energy density plasmas involves hydrodynamic simulations that require transport coefficients in the form of formulas or tables. Building tables is often impractical with ab initio simulations because many of the systems of interest evolve through several decades of density and temperature, and contain mixtures of ionic species. An approximate theory that can predict reasonably accurate transport coefficients at low computational cost is highly desirable.

This Letter presents the first theory to obtain fast and accurate predictions of ionic transport coefficients over a wide range of physical conditions, including warm and hot dense matter, as well as mixtures. The model, which treats the electrons fully quantum mechanically and the ions classically, is fast to evaluate (the calculation time is reduced by a few orders of magnitude compared with ab initio simulations), reduces to the traditional theory in the ideal plasma limit, and is accurate for effective Coulomb coupling strengths up to ~ 30, thus covering a wide range of physical conditions. The model results from the merger of two recent developments; namely the effective potential theory (EPT) [15, 16], which successfully extends conventional plasma transport theory into the strongly coupled regime, together with an average atom model (AA-TCP) that can effectively model the ionic structure and electronic properties of warm and hot dense matter [17, 18]. Below, the transport theory obtained by combining these two models is introduced and benchmarked against molecular dynamics (MD) simulations for diffusion and viscosity for a range of physical conditions representative of current research needs. The plasmas studied include hydrogen, aluminum and iron, which are important in ICF research, basic high energy density plasma experiments and planets, respectively, as well as carbon-helium mixtures, which occur in white dwarf stars. We first recall the main characteristics of the EPT and AA-TCP theories and explain how they naturally combine to make an efficient model of transport processes; further details are given in the Supplemental Material [19].

The EPT is based on the Boltzmann kinetic theory of gases (more precisely, on its extension by Enskog to treat dense gases). However, instead of using the bare pair interaction potential to compute the cross section for binary collisions, an effective potential is used in order to account for the effect of the surrounding medium on the mutual interactions between a pair of colliding ions. In a mixture, an effective potential $\phi_{\alpha\beta}(r)$ is defined for each pair (α, β) of ionic species. The effective potential is set equal to the so-called potential of mean force, which corresponds to the interaction between two ions held a distance r apart when the surrounding particles of the plasma are canonically averaged over all configurations; this potential of mean force is simply related to the ion pair distribution function $g_{\alpha\beta}(r)$; by definition $g_{\alpha\beta}(r) = e^{-\phi_{\alpha\beta}(r)/k_B T}$, where T is the temperature [20]. In addition, following Enskog, the Boltzmann collision operator is corrected by a collision probability

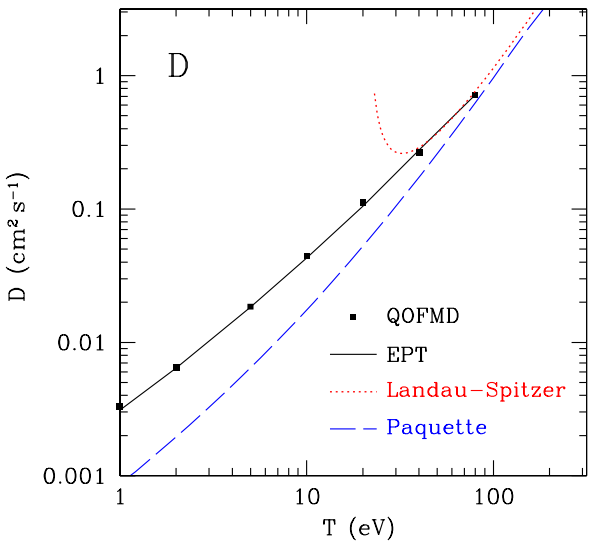

FIG. 1: (color online) Coefficient of self-diffusion of deuterium plasmas at 4.04 g cm^{-3} and temperatures in the range 1 - 80 eV. The EPT results, which were here evaluated using the pair-distribution functions $g(r)$ calculated with the QOFMD simulations, are in excellent agreement with the latter, while the traditional weak-coupling (Landau-Spitzer; red dotted line) plasma theory breaks down at low temperatures. The model of Paquette et al. [21] (blue dashed line) is widely used in stellar astrophysics.

enhancement factor that accounts for the exclusion radius r^* surrounding each ion due to the strong repulsion at short-range. In practice, r^* is chosen to maximizes the accuracy of the theory, leading to the criterion $g_{\alpha\beta}(r^*) = 0.87$ [16]. The transport coefficients are then obtained by substituting the effective potentials in the well-known Chapman-Enskog formulas that result from the solution of the Boltzmann equation with the Enskog correction. Thus, the EPT offers access to the transport properties from the knowledge of only the pair distribution functions, the same static quantities that are typically used to calculate the thermodynamic properties. By comparing with MD simulations of the idealized one-component plasma model, the EPT was previously shown to successfully extend the binary collision approach well into the strongly coupled regime (up to Coulomb coupling parameters $\Gamma \sim 30$).

In fact, and this is an important element of the present work, the applicability of the EPT theory is not limited to simple models like the one-component plasma. This is demonstrated in Fig. 1 by a comparison between accurate quantum molecular dynamic simulations [14, 22] (see below) and the EPT theory for the self-diffusion coefficient of deuterium plasmas at 4.04 g cm^{-3} and temperatures ranging from 1 to 80 eV. Under these conditions, deuterium is fully ionized and the effective ion coupling parameter ranges from $\Gamma_{\rm eff} = 9.5$ to 0.25 (as determined from the first peak of the pair-distribution function [23]). The corresponding electron degeneracy parameter ranges from $k_BT/E_F = 0.024$ to 1.9, where E_F is the Fermi energy. Here the EPT was evaluated using the ion pair-distribution function $g(r)$ extracted from the quantum simulations. Both calculations are in excellent agreement, with the EPT deviating from the latter by at most 6%. On the other hand, the traditional Landau-Spitzer formula completely breaks down at temperatures below 40 eV ($\Gamma_{\rm eff} = 0.6$). The current standard for extending Landau-Spitzer theory to stronger coupling was developed by Paquette et al. [21]. Figure 1 shows that the EPT is much more accurate than this for temperatures below approximately 50 eV. This shows that the EPT gives very good results for realistic systems when an accurate pair distribution function is available. However, each of these data points required a computationally costly calculation of the pair distribution function [24]. In order for the EPT theory to be useful from a practical standpoint, a tool that can promptly generate accurate pair distribution functions for realistic dense plasmas is required.

The recent AA-TCP model [17, 18] provides an effective solution to this problem. This is an all-electron model that has no adjustable parameters; the only inputs are the chemical species (nuclear charges, atomic masses, number fractions), the plasma temperature and the mass density. Ionic mixtures can be handled as well as pure plasmas without additional approximations. Briefly, the model calculates the electronic structure of a (spherically symmetric) central ion and of the surrounding valence (screening) electrons using finite-temperature density functional theory where electrons are subject to both the central nuclear potential and the potential of surrounding ions. When coupled with the theory of fluids, the electron density profile around a nucleus gives ion-ion pair potentials $V_{\alpha\beta}(r)$ and self-consistent ionic pair distribution functions $g_{\alpha\beta}(r)$ (see Supplemental Material). Those $g_{\alpha\beta}(r)$ have been shown to be in excellent agreement with ab initio simulations for a wide range of elements, temperatures, and densities [17, 18, 25, 26], and are obtained at a fraction of the computational cost. These accurate $g_{\alpha\beta}(r)$ can be used as input to the EPT theory to quickly and accurately evaluate the ionic transport coefficients of plasmas simply by specifying the density, temperature and material composition. The $g_{\alpha\beta}(r)$ are calculated up to 15-25 ion sphere radii to ensure the convergence of the EPT calculation. Below, the resulting model, referred to as EPT+AA, is benchmarked against molecular dynamics (MD) simulations for a range of relevant conditions.

Two kinds of MD simulations are performed, namely quantum orbital-free MD (QOFMD) and pseudo-atom MD (PAMD). QOFMD is representative of the state-of-the-art in simulations of dense plasmas, and provides reference values of transport properties, while PAMD is

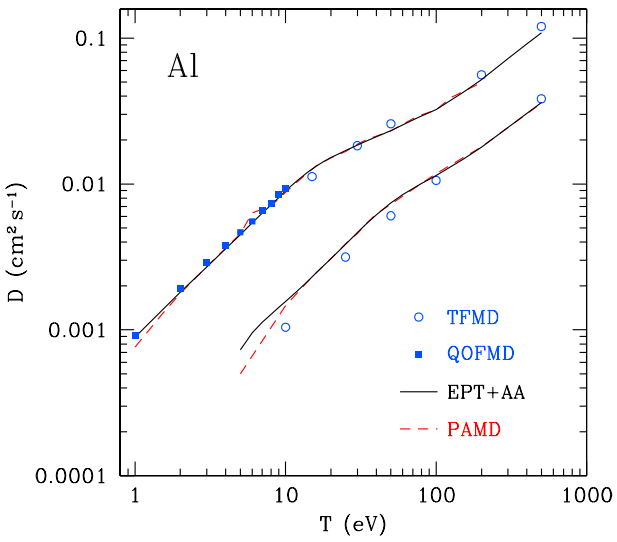

FIG. 2: (color online) Self-diffusion coefficient of aluminum plasmas at solid density (2.7 g cm^{-3} upper curves) and ten times compressed (27 g cm^{-3}; lower curves). The PAMD results agree very well with the ab initio simulations (QOFMD and TFMD at higher temperatures) which shows that the pair potential from the AA-TCP model is accurate. Using this same potential, the EPT reproduces the PAMD very closely except for very strong plasma coupling.

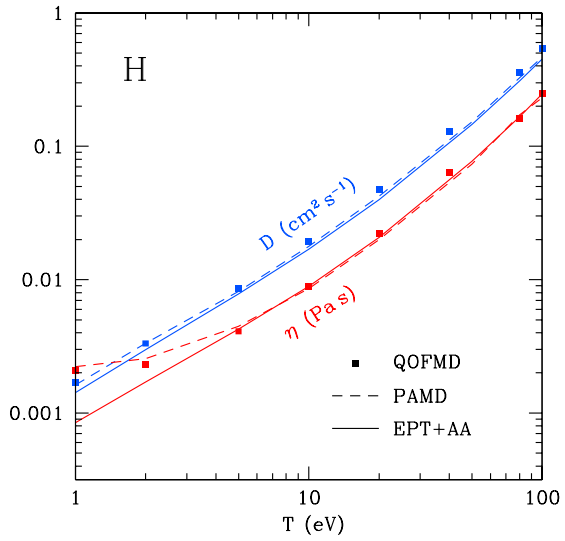

FIG. 3: (color online) Coefficients of self-diffusion D (upper blue curves) and shear viscosity η (lower red curves) of hydrogen plasmas at 8 g cm^{-3}. Both the PAMD and EPT results are in very good agreement with the ab initio QOFMD values. The larger deviations occur at low temperatures where the plasma is very strongly coupled.

used here to demonstrate the internal consistency of the present model. In QOFMD, both electrons and ions are evolved in time within the Born-Oppenheimer approximation. Ions propagate classically according to Newton's equations, while conduction electrons are described with finite-temperature density functional theory using either the accurate orbital-free method of [14, 22] or the high temperature, high density limit, namely the Thomas-Fermi (TF) approximation (we use the latter when QOFMD is numerically impractical). In PAMD [26], the ions alone are propagated in time and interact through pair potentials $V_{\alpha\beta}(r)$ in a classical MD simulation. The $V_{\alpha\beta}(r)$ are outputs of the AA-TCP model and describe the mutual ion interactions screened by the valence electrons. By construction, PAMD simulations reproduce the pair-distribution functions $g_{\alpha\beta}(r)$ of AA-TCP to high accuracy and they give access to the ionic transport properties. Given the pair distribution $g_{\alpha\beta}$ and corresponding potential $V_{\alpha\beta}$ from the AA-TCP model, we compare below the transport coefficients obtained from the EPT+AA model using $g_{\alpha\beta}$ to those extracted from a PAMD simulation using $V_{\alpha\beta}$. These examples further validate the EPT for dense plasmas modeled with realistic potentials. Details of the MD simulations are given in the Supplemental Material.

For practical and fundamental reasons, aluminum is often used in experiments on warm dense matter [27, 28]. While it is usually regarded as a simple metal from the condensed matter point of view, warm dense aluminum is more complex and exhibits temperature- and pressure-ionization, both captured by the AA-TCP model. Figure 2 shows the self-diffusion coefficient of aluminum plasmas at solid density and ten times compressed for temperatures from 1 eV to 500 eV. Under such conditions, the average ionization state varies from $\bar{Z} = 3$ to ~ 12, the effective coupling strength from $\Gamma_{\rm eff} \sim 72$ to ~ 2, and the electron degeneracy parameter from $k_BT/E_F = 0.05$ to 17. The EPT+AA model is in excellent agreement with the PAMD simulations for temperatures corresponding to $\Gamma_{\rm eff} < 30$. The diffusion coefficient calculated with EPT is within 2% of the value from the MD simulations, which reflects the statistical uncertainty of the latter. At lower temperatures where $\Gamma_{\rm eff} > 30$ (e.g. 10 eV at 27 g cm^{-3}), the EPT results rise above the MD value, as expected [16]. The present model gives self-diffusion coefficients that reproduce those from QOFMD and TFMD simulations of Al plasmas to better than 8% for $2 < T < 500$ eV at solid density [26].

The intense experimental campaign to understand the implosion of ICF capsules at the National Ignition Facility has brought increased scrutiny to the transport properties in the warm and hot dense matter created during the compression phase. Hydrogen (more precisely its isotopes deuterium and tritium) is an essential element of these experiments. Figure 3 shows the coefficients of self-diffusion and of shear viscosity of hydrogen plasmas

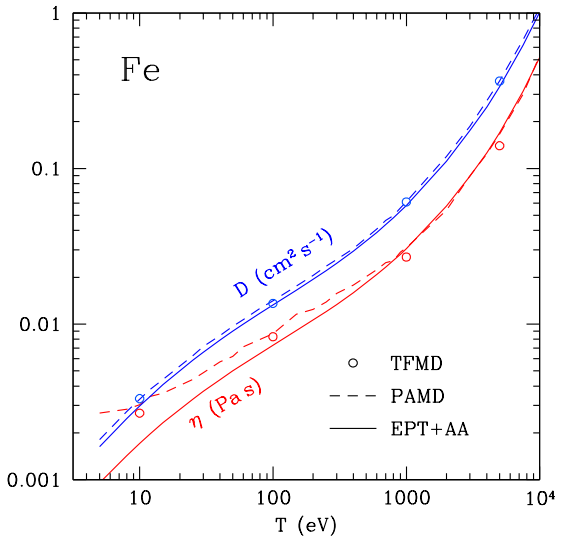

FIG. 4: (color online) Coefficient of self-diffusion D (upper blue curves) and of shear viscosity η (lower red curves) of iron plasmas at 7.87 g cm^{-3}. These calculations are based on the Thomas-Fermi model of the electrons.

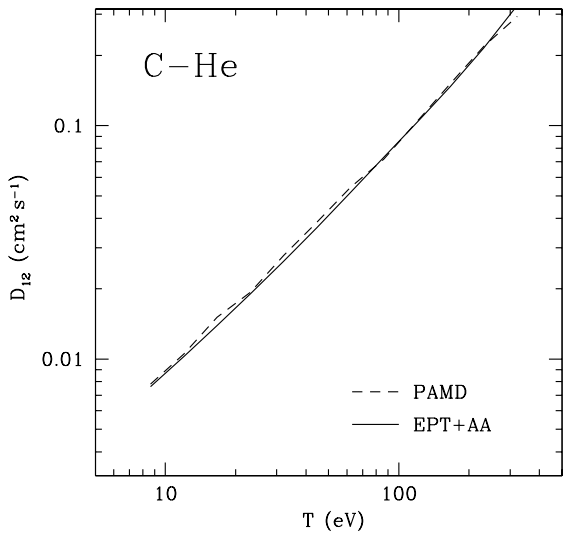

FIG. 5: Inter-diffusion coefficient D_{12} of C/He mixtures (60% C, 40% He by number) at 10 g cm^{-3}. The agreement between EPT+AA and PAMD is excellent, especially considering the level of noise in the latter calculations (±3%).

at 8 g cm^{-3} for T = 1 - 100 eV. Under these conditions, our model indicates that hydrogen is fully ionized, the effective Coulomb coupling parameter ranges from $\Gamma_{\rm eff}$ = 21 to 0.4, and the electron degeneracy parameter from k_BT/E_F = 0.01 to 1. The EPT+AA diffusion coefficients deviate from the PAMD values by $3-12\%$, and from the QOFMD values by $9-16\%$. The coefficient of shear viscosity η agrees even better for $T \gtrsim 5$ eV ($\Gamma_{\rm eff} \lesssim 8$) as the differences between EPT and both the PAMD and QOFMD are everywhere within 6%.

Iron is a key constituent of the core of terrestrial and gaseous giant planets in our Solar System and in other planetary systems. The cores of young, massive gaseous planets can reach temperatures of several eV [5]. Figure 4 shows the self-diffusion coefficient and shear viscosity coefficient of iron plasmas at solid density and T = 5 to 10^4 eV. These calculations are based on the Thomas-Fermi density functional for the kinetic energy of the electrons. Under such conditions, the average ionization state varies from $\bar{Z}$ = 8 to $\sim$ 18.4, the effective coupling strength from $\Gamma_{\rm eff}$ = 39 to 0.6, and the electron degeneracy parameter from k_BT/E_F = 0.4 to 200. Again, for the self-diffusivity, the EPT+AA is in very good agreement with both the PAMD and QOFMD simulations (here within the Thomas-Fermi approximation); for the viscosity, the EPT results depart from the MD results below $T \sim 100$ eV, as expected [16, 29].

Most systems of interest involve mixtures of plasmas rather than the pure elements we have considered so far. As an example of the general applicability of this approach, we consider the coefficient of inter-diffusion [20] in a warm dense plasma mixture of carbon and helium, which occurs at the core-envelope boundary of white dwarf stars. We recall that D_{12} measures how concentration inhomogeneities spread to uniformity [20]. This process is responsible for the carbon "pollution" at the surface of a number of white dwarf stars that would otherwise have a pure helium composition [30]. The EPT theory presented in [15, 16] is readily extended to mixtures using a multicomponent virial expansion [19, 31, 32]; see also the supplemental material and [33] for other benchmarks. Figure 5 shows the inter-diffusion coefficient of 60% C - 40% He mixtures (by number) at 10 g cm^{-3} and for temperatures from 8 to 300 eV. Again, the EPT+AA is in remarkable agreement with the PAMD simulations, with differences that can reach 11% but typically remain below 3%.

In conclusion, we have presented a practical model to efficiently and accurately evaluate the transport coefficients of warm and hot dense plasmas where traditional theory breaks down. The model combines the effective potential transport theory, which simply relates the transport coefficients to the pair distribution functions, and an efficient average atom model, which accurately determines these functions for given plasma constituents and physical conditions. Such a model will be useful to many areas of high energy density physics and astrophysics, where current uncertainties in the transport coefficients hamper understanding of these systems and their reliable modeling. It has broad applicability to plasmas that are weakly to strongly coupled, and to plasma mixtures. Typically, this model gives diffusion

coefficients and shear viscosities within 10% of the ab initio values, which is excellent considering the uncertainties in the latter. In addition to its computational efficiency, it offers the advantages of being free of the statistical noise inherent to evaluating transport coefficients from the Kubo relations applied to MD simulations, it accounts for electronic shell structure through the average atom model, and it covers both the warm and hot dense matter regimes with a single physical model. The combination of the AA-TCP, PAMD and EPT models provides a path to self-consistent calculation of the electronic structure, ionic charge, the equation of state and the ionic transport properties of dense plasmas. Finally, the present work suggests that an experimental measurement of the pair distribution functions or the structure factors could in principle be used to infer ionic transport properties.

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