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LA-UR--89-3020

DE90 000607

TITLE *Many-Electron Effects on Dynamic Processes in Dense Matter*AUTHOR(S) *Stephen M. Younger*SUBMITTED TO *Yamada Conference on Strongly Coupled Plasma Physics
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MANY-ELECTRON EFFECTS ON DYNAMIC PROCESSES IN DENSE MATTER

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Large scale calculations quantum mechanical calculations of dynamic processes in dense helium suggest that quasi-molecular effects may play an important role in determining the transport properties of dense plasma.

1. INTRODUCTION

Recent experiments suggest that many-electron processes may be important in determining both the transport and radiative properties of strongly interacting plasma. In x-ray measurements of the high density implosion of an argon filled laser fusion capsule Hooper et al.¹ observed a prominent high energy feature associated with the heliumlike 1s-2p transition. This feature occurred only near peak compression when the plasma was *both* hot and dense. Although it is typical to observe a rich satellite structure at lower photon energies, structures at high energy are unusual, and are difficult to interpret in terms of a single-atom model of the emitter. We have suggested that such features may be associated with the formation of transient *molecular* complexes resulting from stochastic thermal motion within the plasma, i.e., encounters close enough to allow significant hybridization of the atomic orbitals. If an electron collision occurs while the ions are in such a configuration, the resulting photon emission will occur at the instantaneous *molecular* transition energy, which can be shifted to higher energy as the ion pair approaches the united atom limit.

At lower temperatures, two stage gas gun measurements of the shock Hugoniot of helium at densities up to 0.6 g/cc and inferred temperatures of 2 eV have been performed by Nellis et al.² Modeling of these experiments suggests that the effective interatomic interaction at high density is substantially softer than one would expect from accurate theoretical calculations or measurements of the binary He-He interaction.

These experiments suggest that the dynamics of matter at high density may be more complex than previously anticipated. Indeed, both of the above results indicate that many-center interactions are important in determining the properties of dense matter. Theories which include only a single atom in an "effective" medium may omit important dynamic correlations wherein the forces within and between atoms are determined by the instantaneous configuration of its neighbors. We have recently performed a series of calculations aimed at addressing this point.

2. SELF-CONSISTENT FIELD MOLECULAR DYNAMICS

In order to explicitly study the effects of many-atom interactions at high densities we have developed a new theory of dynamic matter based on a synthesis of techniques from quantum chemistry and molecular dynamics. Our method consists of the repetition of three steps. First, we use a molecular orbital Hartree-Fock approximation to compute the electronic structure of a large

cluster of atoms. Using this wavefunction to derive a charge density, we calculate the net electrostatic forces acting on each of the nuclei. Finally, we allow the nuclei to move slightly in response to the forces. By repeating these three steps we are able to generate time histories of the atomic motion, allowing later postprocessing of quantities such as correlation functions and transport coefficients. This is an adiabatic approximation propagating a ground state configuration in the Born-Oppenheimer approximation.³

3. DYNAMIC SCREENING

3.1. Static Calculations.

Before describing the results of the full SCFMD calculations, it is worthwhile to examine the effect of many-atom screening on a small configuration of atoms. Figure 1 shows the effect of neighbor atoms on the binary interaction force. If the total force on atom B were simply a superposition of pair potentials the ratio in Figure 1 would always be one, since the neighbor atoms are symmetric about the test atom. The reduction in the force when neighbors are present is the result of the self-consistent redistribution of electronic charge density throughout the cluster. The addition of the nuclear forces of atoms B, C, and D results in a net potential which is deepest at the location of atom B. This deep potential draws in electronic charge density, which serves to screen atom B from the effect of atom A.

FIGURE 1

Effect of near neighbors on interatomic force. Solid: electrostatic forces. Dashed: energy gradient forces.

FIGURE 2

Four regimes in dense plasma. d is the internuclear distance, λ is a characteristic electron wavelength.

In a related study⁴, calculations were performed on a body-center-cubic cluster of nine atoms using a finite temperature Hartree-Fock-Slater approximation. As the lattice spacing was decreased, i.e. the density increased, the effective number of bound electrons at first decreased reflecting the onset of pressure ionization. At higher densities, however, the bound population *increased* as the electrons were attracted to the deep and extended potential well created by the

superposition of the nine nuclear potentials.

These calculations led us to propose that dense plasma can be divided into four regimes, classified by the relationship of the interionic spacing to the characteristic wavelength of the electrons. As shown in Figure 2, at low densities the electrons occupy separate potential wells and the kinetics can be modeled as sums of binary encounters. As the density increases the tails of the atomic wavefunctions begin to overlap, resulting in screening of the nuclear potentials and a rise in atomic eigenvalues. This reduction in binding is associated with pressure ionization. As the density increases still further, however, it is possible for clusters of nuclei to *collectively* bind electrons. As in a covalent bond, the electrons are shared among several centers. Since the effective potential in the center of the cluster is deeper than that associated with a single atom, the electrons are more tightly bound and the bound atomic population increases. At very high densities the superposition of atomic potentials results in a dense packing of narrow wells which are singly and collectively unable to bind electrons. This is the limit of a homogeneous electron gas.

Regular crystalline lattices do not exist in a dense plasma. It is possible, however, to form transient clusters of ions due to the stochastic motion of atoms at high temperature. Such clusters are extremely important for the dynamics of the ensemble, since it is in close collisions that the largest momentum transfer occurs. Hence for energy transport in dense matter it is important to self-consistently treat the electronic and ionic motion. This was the purpose of our self-consistent field molecular dynamics calculations.

3.2. Dynamic Calculations

Self-consistent field molecular dynamics calculations on helium at densities between 0.1 and 3.0 g/cc and temperatures of 1 and 5 eV were performed on samples of 23 and 30 atoms. Periodic boundary conditions were imposed to mimic the effects of extended media and to minimize surface effects. The wavefunction was expanded in a Gaussian basis set of two or three functions attached to each of the nuclei.

Figure 3 illustrates the normalized velocity autocorrelation function $Z(t) = \langle \mathbf{v}(t) \cdot \mathbf{v}(0) \rangle / (\mathbf{v}(0) \cdot \mathbf{v}(0))$ where $\mathbf{v}$ is the velocity and the brackets indicate averages over the atoms as well as the initial time. In addition to the SCFMD results, shown by solid lines, we also give the results of classical molecular dynamics calculations performed with several different pair interactions potentials. At low density all of the calculations agree save the one which used the high density potential of Young et al. This is not surprising since this potential *assumes* the presence of nearest neighbors which produce screening. At low density and high temperature it is possible for atoms to approach one another without any neighbors nearby. As the density increases the SCFMD velocity autocorrelation function assumes a progressively longer decay time than those computed from binary pair potentials. This is due to the formation of transient quasi-molecular clusters which dynamically screen the interatomic interactions.

FIGURE 3

Velocity autocorrelation functions for dense helium. — SCFMD; Aziz et al., Hartree-Fock short range potential⁵; — — Aziz et al. quantum Monte Carlo short range potential⁶; — — — Young et al.⁷ derived from high density linear muffin tin calculations which assume the constant presence of several near neighbors; — — — pair potential derived from the same atomic basis set used in our SCFMD calculations.

4. SUMMARY

Self-consistent field molecular dynamics calculations suggest that transient quasi-molecular configurations may have an important effect on dynamic processes in dense matter. Although not explicitly discussed in this paper, we expect that such effects may be especially pronounced for electron transport.

ACKNOWLEDGEMENT

It is a pleasure to acknowledge helpful interactions with Gayle Sugiyama and Alan Harrison of LLNL. This work performed under the auspices of the U.S. DOE by the Lawrence Livermore National Laboratory under Contract No. W-7405-ENG-48.

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