Quantum Modeling of Electronic Charge Density in Warm Dense Matter

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***Abstract*— Warm dense matter plasmas can be produced by high-energy optical or free-electron X-ray lasers, Z-pinch X-ray sources, and nuclear detonations. These plasmas are the basis of inertial confinement fusion efforts at National Ignition Facility. The *ab initio* quantum modeling of warm dense aluminum (Al) plasmas is performed. We present several images of electronic charge density taken from investigations of structural optimization of Al atoms in warm dense plasma.**

***Index Terms*— Density functional theory, plasma properties, quantum computing.**

The regime of warm dense matter (WDM) defines the state between plasmas and solids [1]. The density of WDM is ranging from 0.1 to 10-fold solid density and temperature from 1 to 100 eV, where thermal, Coulomb, and Fermi energies are nearly equal. A complex interplay between collective ion couplings and quantum effects is occurred. Therefore, it is very challenging to model the physics of WDM due to the lack of a dominating energy scale and physical behavior [2]. Accounting for all atomic and electronic interactions at a truly *ab initio* quantum level is required to model the crucial WDM physics. In this paper, we present images of electronic charge density in warm dense Al plasmas taken from our *ab initio* quantum calculations.

The Quantum-Espresso (QE) package [3] that represents an open-source package of computer codes for electronic-structure calculations and materials modeling are used to perform *ab initio* quantum calculations. It is based on density-functional theory (DFT), plane waves, and pseudopotentials. A core component of the QE package [3] is the plane-wave self-consistent field model for self-consistent calculations of electronic properties (ground-state energies, one-electron orbitals, atomic forces, stresses, and so on) within the DFT framework using a plane-wave basis set and pseudopotentials. The generalized gradient approximation is used for the exchange-correlation functional. The atomic potential is described by *ab initio* ultrasoft pseudopotential. The plane-wave energy cutoff is 30 Ry and an $8 \times 8 \times 8$ Monkhorst-Pack k-point mesh is used for Brillouin zone integration.

The results on the optimization of eight Al atoms and distribution of their electronic charge density are shown in Fig. 1. The setup of simulation system is shown in Fig. 1(a) for Al at solid density. A cubic cell is filled with 8 Al atoms. Computational requirements scale as $O(N^3)$ with the number of atoms N in a cell. It is then replicated in all directions to produce a periodic array. A single layer of cell images around a central cell is shown in Fig. 1(a). The central cell is surrounded by 26 nearest neighbors. The Hamiltonian accounts for the interaction of all 216 atoms in the central and 26 replicated cells. An external compensation field is introduced in the vacuum region around the replicated cells in order to prevent the artificial field due to edge effects. If an Al atom leaves a central cell during quantum simulations, it is replaced by an image Al atom that enters from the opposite side. Thus, the number of Al atoms in a cell is not changed. In such a way, an infinite dense matter is represented with small number of atoms that experience forces as if they were in bulk environment. The eight Al atoms were initially placed in a central cell at random locations. The structural optimization of nuclear positions is then performed to reach equilibrium. The structure was relaxed until forces on all atoms are lower than 0.001 Ry/bohr. During optimization, formation and breaking of chemical bonds between Al atoms were observed. However, the bonding of Al was not stable.

The 3-D isosurface (in green) of the electronic charge density in equilibrium is shown in Fig. 1(b). It is plotted for an isovalue of 0.01 electrons/bohr^3. The Al atoms are also displayed as pink balls. The isosurface serves as a canvas indicating the distribution of the electronic charge density. It is seen that the Al atoms are in equilibrium, since the isosurface envelops them uniformly. The 2-D isolines (contour plots) of the electronic charge density are shown in Fig. 1(c) for different positions (panels 1–4) within a central cell. The contour lines in Fig. 1(c) in panel 1 are displayed near the front of a cell from the side of an observer. Two Al atoms are also explicitly shown. The other Al atoms are located behind [shown as shadow spheres in Fig. 1(c) in panel 1]. The plane of contour lines is then moved into the cell, and the 2-D isolines are shown in Fig. 1(c) in panels 2–4 for some locations. Those in Fig. 1(c) in panel 4 correspond to the location near the backside of the cell. The Al atoms that explicitly appear as the isoline plane moves toward the backside of the cell are illustrated [all eight atoms are shown in Fig. 1(c) in panel 4]. In Fig. 1(c), the blue color corresponds to a largest value of electronic charge density, and the red color indicates the smallest charge density. It is seen that the electrons are localized around the Al nuclei (blue-green hills). However, there are also valleys of the low charge density between the Al atoms (red wells). In valleys, the electronic charge density

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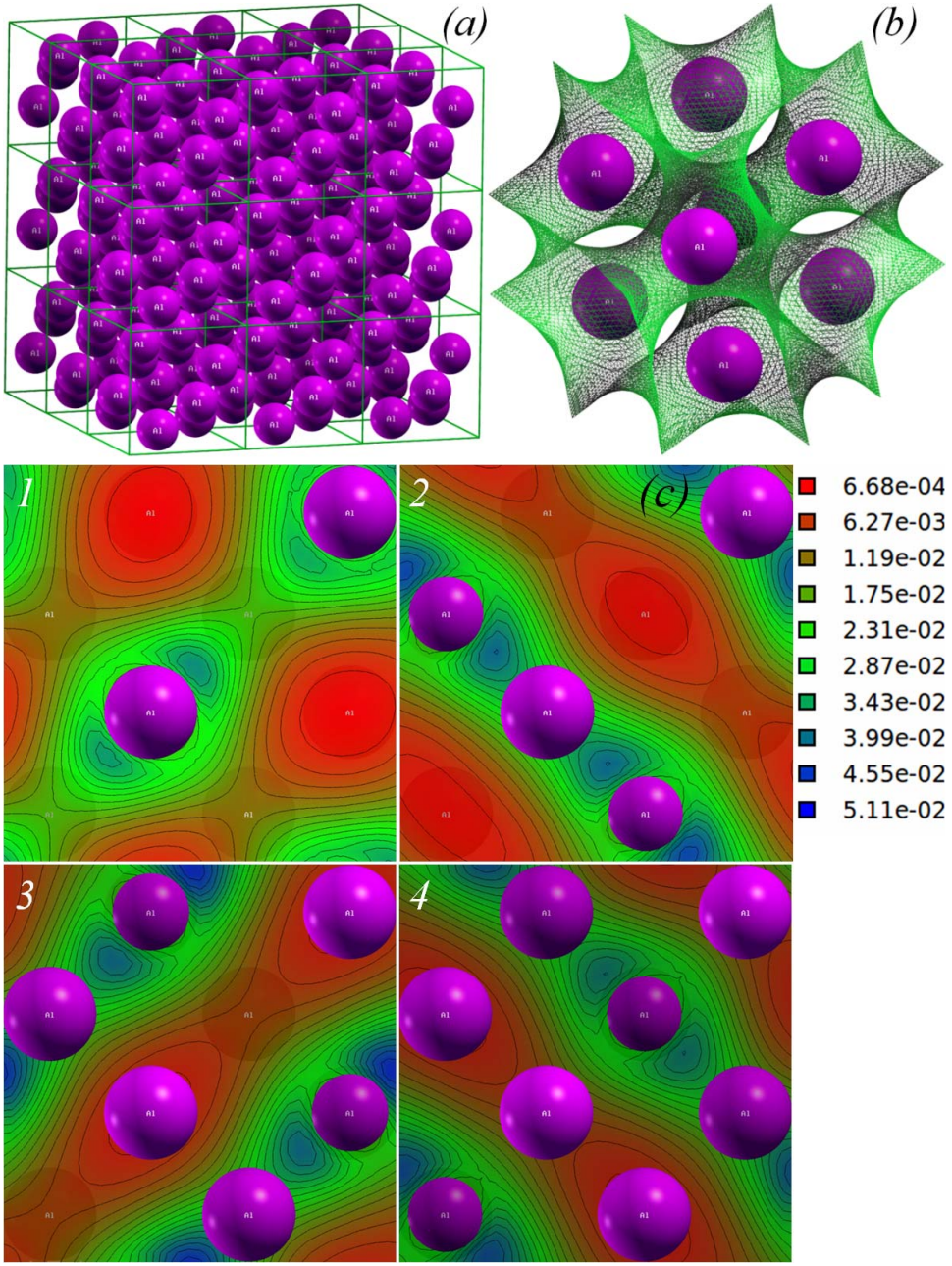

Fig. 1. (a) Setup of simulation system for quantum calculations of electronic charge density in warm dense Al plasma. (b) 3-D isosurface for an isovalue of 0.01 electrons/bohr^3. (c) 2-D isolines of electronic density in a computational cell at different positions shown in panels 1–4.

is about two orders of magnitude lower compared with that near the nuclei.

REFERENCES

[1] M. Koenig *et al.*, "Progress in the study of warm dense matter," *Plasma Phys. Controlled Fusion*, vol. 47, no. 12B, pp. B441–B449, 2005.

[2] J. J. Barnard *et al.*, "Theory and simulation of warm dense matter targets," *Nucl. Instrum. Methods Phys. Res. A*, vol. 577, nos. 1–2, pp. 275–283, 2007.

[3] P. Giannozzi *et al.*, "QUANTUM ESPRESSO: A modular and open-source software project for quantum simulations of materials," *J. Phys., Condens. Matter*, vol. 21, no. 39, p. 395502, 2009.