Unraveling the Intrinsic Atomic Physics Behind X-Ray Absorption Line Shifts in Warm Dense Silicon Plasmas

V. V. Karasiev and S. X. Hu

Laboratory for Laser Energetics, University of Rochester

Accurate knowledge of radiative properties of matter in a wide range of material densities across different temperature regimes is of growing importance in many areas of research such as planetary science, astrophysics, and inertial confinement fusion (ICF).[1–4] Our current incomplete understanding of atomic physics in dense plasmas has been demonstrated by the measurements (in Refs. 5–7) of the ionization potential depression (IPD) in warm/hot dense aluminum plasmas, which have called into question the traditional continuum-lowering plasma physics models such as Ecker–Kröll[8] and Stewart–Pyatt.[9]

In this work, we develop a novel methodology based on *all-electron* density functional theory (DFT) for calculating the optical properties of warm dense plasmas of mid-Z materials in a broad range of x-ray photon energies (up to $h\nu \sim 10$ keV). To demonstrate its applicability, we have used this novel method to systematically calculate the x-ray absorption of dense silicon plasmas for a wide range of densities and temperatures (ρ = 0.5 to 500 g/cm^3 and $T = 5 \times 10^3$ to 10^7 K). Based on these data, a first-principles opacity table (FPOT) of silicon has been built for ICF and high-energy-density physics applications. These *ab initio* results revealed interesting trends of density/temperature–induced red-to-blue shifts of K-edge and $1s \rightarrow 2p$ absorption lines along both the isotherm and isochore. These absorption-line shifts provide a ubiquitous measure of the competition between screening of deep bound electrons and screening of outer-shell electrons due to the warm-dense-plasma environment. Our data indicate that one can use the absorption ratio of $1s \rightarrow 2p$ to K edge for characterizing thermodynamic conditions of warm dense plasmas through x-ray spectroscopy measurements coupled with DFT calculations.

These shifts for the T = 500-kK isotherm are summarized in Fig. 1(a) for different silicon densities [solid red and dashed curves (circles) labeled "DFT (this work)"], where the solid and dashed horizontal lines refer to the K edge and $1s \rightarrow 2p$ locations for silicon at ambient density 2.33 g/cm^3 and T = 500 kK. It is clearly seen that both absorption lines have the similar trend of red-to-blue shift as ρ increases. This behavior underlines the competition between electron screening and ion–ion interaction effects on the deeply bound electrons and the outer-shell electrons. Figure 1(b) shows results for the ρ = 35-g/cm^3 isochore [solid red and dashed curves (circles) labeled "DFT (this work)"]. Along this thermodynamic path, the deeply bounded $1s$ states move down as temperature increases as a consequence of the decreased screening by upper-bound electrons (due to thermal-induced ionization). As a result, the K edge shifts upward overall, except for an ~40-eV red shift at temperatures between 125 kK and 250 kK, shown in Fig. 1(b); meanwhile, the $1s \rightarrow 2p$ absorption line exhibits a monotonic blue shift. To further understand these interesting features, we examine the widely used continuum-lowering models. These plasma-physics models predict the IPD for a given plasma condition (density, temperature, and an ion charge state $\overline{Z}$) with respect to an isolated ion. Figure 1(a) indicates that the atomic models of Stewart–Payatt,[9] corrected Stewart–Pyatt,[10] modified ion sphere,[11] and Crowley[12] fail to predict the K-edge red shift, providing only a qualitatively correct trend for the blue shift occurring at $\rho > 50$ g/cm^3.

Finally, we plot the ratio of the $1s \rightarrow 2p$ absorption to the K-edge absorption coefficients in Figs. 1(c) and 1(d), as the function of thermodynamic conditions varies. Figure 1(c) shows this ratio peaks at $\rho \sim 5$ to 7 g/cm^3 for T = 500 kK, then decreases to zero as the density increases. For the constant density shown in Fig. 1(d), the ratio monotonically increases as plasma temperature increases. Guided by such DFT calculations, one can measure this absorption ratio to infer the density and temperature in experiments.

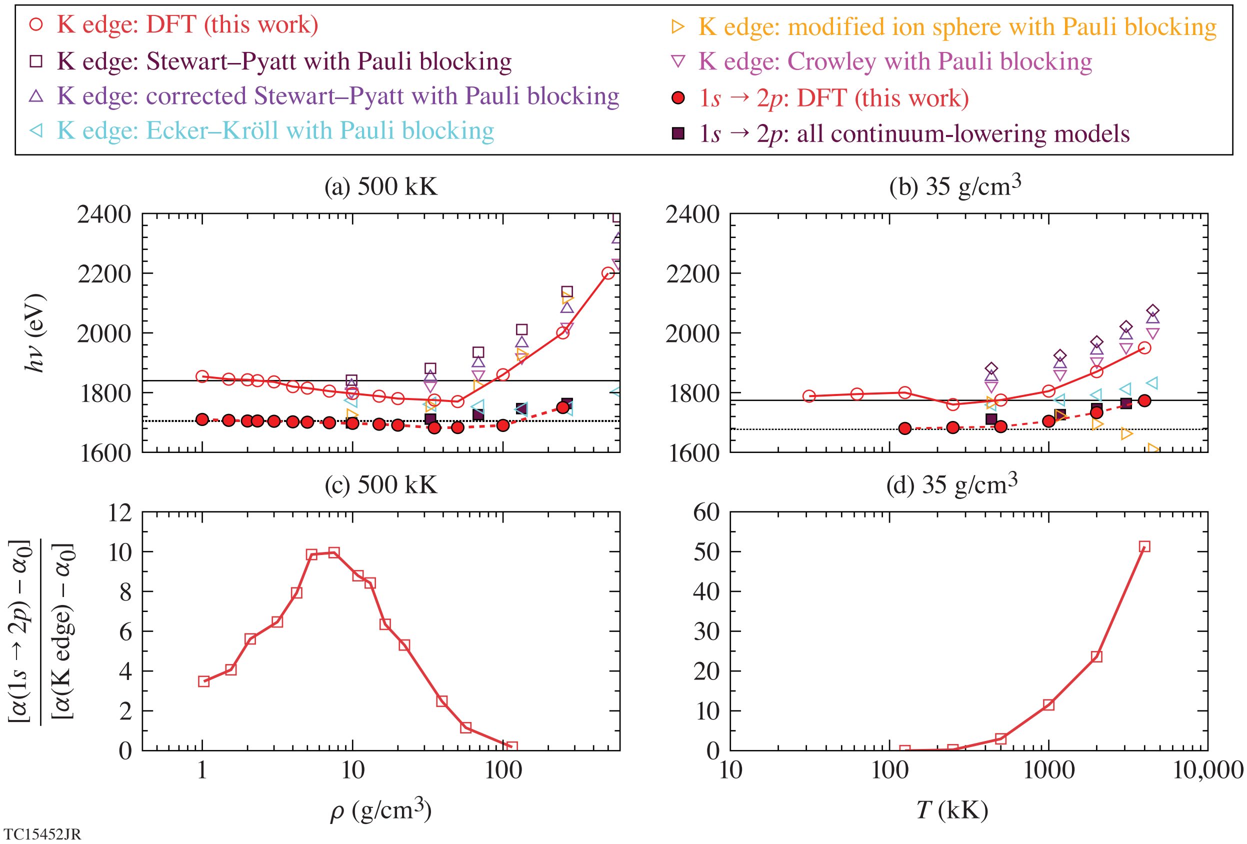

Figure 1
(a) Pressure-induced effect on the K-edge and $1s \rightarrow 2p$ absorption lines in silicon plasmas along the T = 500-kK isotherm; (b) temperature-induced effect on the K-edge and $1s \rightarrow 2p$ absorption line location in silicon plasmas along ρ = 35.0-g/cm^3 isochore; (c) the ratio of absorption coefficients between α (K–L) and α (K-edge) with subtracted L-tail background absorption α_0 along T = 500-kK isotherm; (d) similar to (c) for the ρ = 35.0-g/cm^3 isochore.

The total opacities from our DFT and astrophysical opacity table (AOT) calculations are further compared in Fig. 2. The total/gray Rosseland mean opacity K_R of Si is shown as a function of temperature for three representative cases: ρ = 5, 35, and 250 g/cm^3. Both DFT and AOT total opacities agree well for temperatures above a few tens of eV; however, the AOT model significantly underestimates the total opacity for ρ = 5 at temperatures below 10 eV. This finding is similar to the comparison between DFT and AOT for CH plasmas.[13] For densities ρ = 35 and 250 g/cm^3 at low temperatures (near 10 eV and below), the AOT total opacities are higher than the reference DFT values.

In conclusion, a novel free-energy DFT-based methodology has been developed that enables us to perform first-principles calculations of x-ray absorption in warm dense mid-/high-Z plasmas for a wide range of photon energies and plasma conditions. Applying the developed method to warm dense silicon plasmas, we revealed interesting red-to-blue shifts of K-edge and K–L absorption, which are explained by the competition between the free-electron screening of the K-shell core electrons and the screening of outer L-shell and M-shell electrons. Observing the fact that the relative magnitude of the K–L and K-edge absorption strongly depend on plasma environment, we propose using the ratio of $1s \rightarrow 2p$ absorption to the K-edge absorption to characterize the thermodynamic properties of dense plasmas through the x-ray spectroscopy technique. Novel methodology developed in this work was applied for systematic calculations of absorption and mean grouped opacity of silicon plasmas in a wide range of thermodynamic conditions. The resulting FPOT data were compared with the widely used AOT model. We found significant quantitative and qualitative discrepancies.

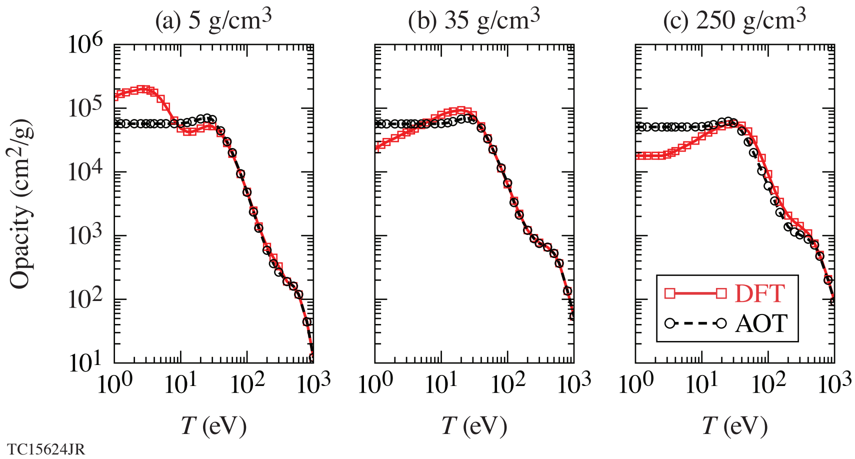

Figure 2
Total Rosseland mean opacity of silicon at $\rho = 5$, 35, and 250 g/cm^3 as a function of temperature as predicted by our first-principles DFT calculations and AOT model.

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