Quantum-mechanical calculation of ionization potential lowering in dense plasmas

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Abstract

The charged environment within a dense plasma leads to the phenomenon of **ionization potential depression (IPD)** for ions embedded in the plasma. Accurate predictions of the IPD effect are of crucial importance for modeling atomic processes occurring within dense plasmas. Several theoretical models have been developed to describe the IPD effect, with frequently discrepant predictions. Only recently, first experiments on IPD in Al plasma have been performed with an x-ray free-electron laser (XFEL), where their results were found to be in disagreement with the widely-used IPD model by **Stewart and Pyatt**. Another experiment on Al, at the Orion laser, showed disagreement with the model by **Ecker and Kröll**. This controversy shows a strong need for a rigorous and consistent theoretical approach to calculate the IPD effect. Here we propose such an approach: a **two-step Hartree-Fock-Slater model**. With this parameter-free model we can accurately describe the experimental Al data and validate the accuracy of standard IPD models. Our model can be a useful tool for calculating atomic properties within dense plasmas with wide-ranging applications to studies on warm dense matter, shock experiments, planetary science, inertial confinement fusion and studies of non-equilibrium plasmas created with XFELs.

Introduction

Dense plasmas

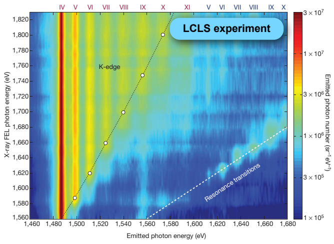
> The dense plasma state is a common phase of matter in the universe and can be found in all types of stars.

> The dense plasmas are created during experiments involving high-power light sources such as National Ignition Facility (NIF) and recently developed x-ray free-electron lasers (XFELs).

> IPD (Ionization Potential Depression): The screening by the dense free-electron environment shifts the atomic energy levels, leading to a reduction of the ionization potentials.

> Quantitative predictions of IPD are of crucial importance for a correct understanding and accurate modeling of any atomic processes occurring within a dense plasma environment.

> Only recent experiments provide quantitative data to verify theoretical models whose predictions sometimes differed extensively.



Vinko *et al.*, *Nature* **483**, 59 (2012).

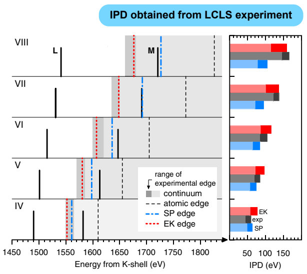
Recent experiments for Al dense plasmas

> The LCLS experiment measured *K*-edge thresholds and *K*α emission from solid-density Al plasma (*T*=0~80 eV).

- *K*α fluorescence detected and spectrally resolved as a function of the incoming photon energy
- the onset of the incident photon energy corresponds to the *K*-edge; IPD for *K*-shell obtained for each charge state

> The Orion experiment investigated *K*-shell emissions from hot dense Al plasma (*T*=700 eV).

- Lyβ and Heβ transition lines measured as the density increases
- The 3*p* state becomes unbound due to the IPD effect when the density is larger than 8~10 g/cm³.



Ciricosta *et al.*, *Phys. Rev. Lett.* **109**, 065002 (2012).

Controversy in theoretical IPD models: EK vs. SP

> EK model: Ecker & Kröll, *Phys. Fluids* **6**, 62 (1963)

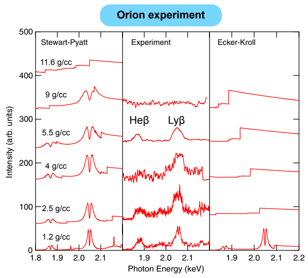
> SP model: Stewart & Pyatt, *Astrophys. J.* **144**, 1203 (1966).

- extended for weakly coupled plasmas

> The LCLS results agrees with the modified EK model, but disagrees with the extensively used SP model.

> The Orion results could only be described with the SP model, whereas the EK model showed a clear disagreement with their data.

> There is a strong need for a rigorous and consistent theoretical approach able to calculate the IPD effect for plasmas in different coupling regimes.



Hoarty *et al.*, *Phys. Rev. Lett.* **110**, 265003 (2013).

Two-step Hartree-Fock-Slater model

Step 1

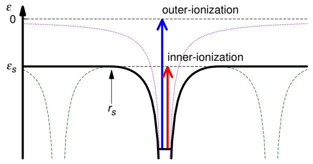
> obtain the electronic structure of an ion embedded in the electron plasma from the finite-temperature approach, assuming thermalization of bound electrons within the free-electron plasma

Step 2

> treat individual electronic configurations of plasma ions to provide a description of discrete transitions

> solve the Schrödinger equation with XATOM

- muffin-tin approximation for the atomic potential
- Slater exchange potential
- numerical grid method (generalized pseudospectral method)
- bound and continuum states obtained by diagonalizing the discretized Hamiltonian with the same atomic potential
- no boundary condition at the Wigner-Seitz radius; use a sufficiently large maximum radius



Schematic diagram of an atomic model in a plasma

First step: average-atom calculation

> one of the finite-temperature approaches

> quantum mechanical approach with the muffin-tin approximation

> grand-canonical ensemble at a given temperature

> electron density with fractional occupation numbers

$$\rho(\mathbf{r},T) = \sum_p |\psi_p(\mathbf{r})|^2\, \tilde{n}_p(\mu,T)$$

> fractional occupation numbers according to the Fermi-Dirac distribution with a chemical potential

$$\tilde{n}_p(\mu,T) = \frac{1}{1+e^{(\varepsilon_p-\mu)/T}}$$

> charge neutrality of the plasma on average, assuming that electrons do not escape from the plasma

$$N_{\text{elec}} = \int_{r\le r_s} d^3r\ \rho(\mathbf{r},T)$$

> chemical potential determined from the charge neutrality condition

$$N_{\text{elec}} - \sum_p \left(\int_{r\le r_s} d^3r\ |\psi_p(\mathbf{r})|^2 \right) \tilde{n}_p(\mu,T) = 0$$

> self-consistent field calculation: the chemical potential and the muffin-tin flat potential as well as orbitals and orbital energies are obtained self-consistently

Second step: fixed-config. calculation

> fixed configuration calculation using a microcanonical ensemble in the presence of the free-electron (plasma electron) density

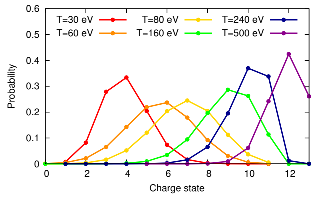
> probability of finding one bound-state configuration within the grand-canonical ensemble from the first step

$$P_{[n_b]} = \prod_b^{\text{bound}} \frac{e^{-(\varepsilon_b-\mu)n_b/T}}{1+e^{-(\varepsilon_b-\mu)/T}},$$

> free-electron density obtained from the first step

$$\rho_f(\mathbf{r}) = \sum_p^{\text{continuum}} |\psi_p(\mathbf{r})|^2\, \tilde{n}_p(\mu,T)$$

> bound-electron density is self-consistently updated, whereas free-electron density is fixed during the SCF procedure.



Probability distribution of charge states for different temperatures

XATOM toolkit

Theory

We implement an integrated toolkit, XATOM, to treat x-ray-induced processes based on nonrelativistic quantum electrodynamics and perturbation theory within the Hartree–Fock–Slater model. It has been extended to treat the electronic structure of atoms in a solid or a plasma with the muffin-tin approximation.

Physical processes

> Photoionization

> Auger (Coster–Kronig) decay

> Fluorescence

> Shake-off

> Elastic x-ray scattering

> Resonant elastic x-ray scattering (dispersion correction)

Using the plasma extension, these processes can be treated with screening effect in a plasma environment.

Damage dynamics

To simulate electronic damage dynamics in intense x-ray pulses, we use the rate equation approach with photoionization cross sections, Auger rates, and fluorescence rates, for all possible *n*-hole electronic configurations for all possible +*n* charge states.

Applications

> Ionization, relaxation, and scattering dynamics at high intensity

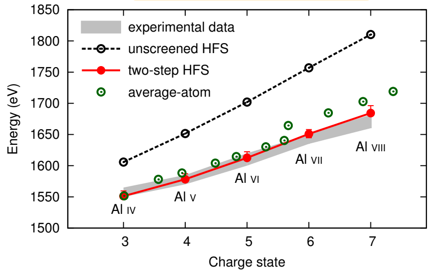
> Charge distribution analysis of noble gases in XFELs

> Photoelectron / Auger / fluorescence spectra

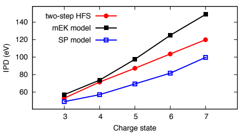
> Multi-wavelength anomalous diffraction at high intensity

Comparison with experiments: strongly and weakly coupled Al plasmas

Comparison with LCLS experiment:
***T*=10–80 eV and solid density (2.7 g/cm³)**



IPD comparison



> We choose the most probable bound-electron configuration (not always ground-state configuration) of the most probable charge state.

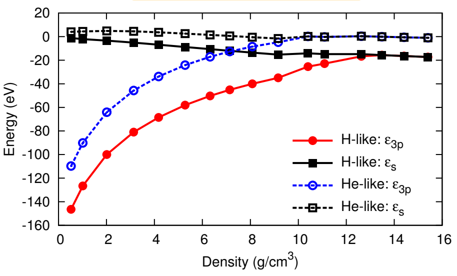
> Our two-step HFS calculations of the *K*-shell threshold energies agree with the LCLS experiment, while the average-atom model alone (first step only) fails in reproducing experiment.

> Our model computes the energy shifts of all individual orbitals with and without plasma screening, providing IPDs in a consistent manner.

> For the IPD model, neither the mEK model nor the SP model are close to our two-step HFS approach.

> Our two-step HFS model can calculate individual *K*α transition lines, whereas the average-atom model alone cannot.

Comparison with Orion experiment:
***T*=700 eV and 0.5~4×solid density**



> The H-like Al is calculated with the exact one-electron potential and the He-like Al is calculated with the exact Hartree-Fock potential.

> For this high temperature, the plasma electron density contributes to only classical Coulomb interaction.

> The 3p state of H-like Al (He-like Al) is no longer bound after the density is larger than ~12 g/cm³ (~10 g/cm³), so the Lyβ (Heβ) transition line would disappear.

> Our results are consistent with the experimental data and the results of the SP model.

Conclusions

> We extend the standard HFS approach for calculating atomic energy levels for ions embedded in a plasma.

> Our two-step HFS model includes (i) average-atom calculation at a given temperature and (ii) fixed-configuration calculation taking into account the free-electron density.

> For the LCLS experiment, our results on the *K*-shell threshold energies of different charge states within Al plasma shows good agreement with experimental data. Our calculated IPDs lie between the SP and mEK models.

> For the Orion experiment, our prediction on the 3p state is in good agreement with the SP model and experimental data.

> Our model provides a reliable calculation in both weakly and strongly coupled plasma regimes.

> The two-step HFS model can be a useful tool for calculating atomic properties within plasmas with wide-ranging applications.

References

> Present work: Son, Thiele, Jurek, Ziaja & Santra, (submitted).

> LCLS experiment: Vinko *et al.*, *Nature* **482**, 59 (2012); Ciricosta *et al.*, *Phys. Rev. Lett.* **109**, 065002 (2012)

> Orion experiment: Hoarty *et al.*, *Phys. Rev. Lett.* **110**, 265003 (2013); Hoarty *et al.*, *High Energy Density Phys.* **9**, 661 (2013).

> Finite-temperature approach: Mermin, *Ann. Phys.* **21**, 99 (1963).

> Average-atom model: Rozsnyai, *Phys. Rev. A* **5**, 1137 (1972); Blenski & Ishikawa, *Phys. Rev. E* **51**, 4869 (1995); Johnson *et al.*, *J. Quant. Spectrosc. Radiat. Transfer* **99**, 327 (2006); Sahoo *et al.*, *Phys. Rev. E* **77**, 046402 (2008); Johnson *et al.*, *Phys. Rev. E* **86**, 036410 (2012).

> XATOM: Son, Young & Santra, *Phys. Rev. A* **83**, 033402 (2011).

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