Enormous enhancement of molecular ionization at high x-ray intensity

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Synopsis The ultra-intense and ultra-short x-ray pulses provided by X-ray Free-Electron Lasers (XFELs) sequentially ionize molecular samples many times. In this context, we have recently discovered that under intense x-ray radiation the total charge of a molecule is enhanced compared to the sum of charges of its constituent isolated atoms[2, 3]. We report here on new theoretical results[5] for iodobenzene (C_6H_5I) that show an even stronger ionization enhancement than previously observed for iodomethane (CH_3I)[3].

Molecules exposed to intense x-ray radiation undergo multiple ionization due to repeated core ionization and subsequent Auger decay cascades. In such a scenario the molecule can populate a huge number (trillions for CH_3I) of different highly excited electronic configurations. We employ a kinetic Monte Carlo technique[4] to solve rate equations for the electronic configurations. Within this scheme all the electronic transition rates are calculated on the fly[1, 2].

We have modeled the x-ray radiation damage in CH_3I and C_6H_5I when exposed to very intense x-ray light with photon energy of 8.3 keV and pulse duration of 30 fs, as produced in recent experiments at XFEL[3]. Because the photoabsorption cross section is much lower on carbon and hydrogen atoms, the photoabsorption happens almost exclusively on the iodine atom. In Figure 1 we show the average charge state obtained after such an x-ray pulse as a function of fluence. We observe that the average total charge in a CH_3I molecule is significantly enhanced compared to what one obtains for a simulation with isolated atoms. This effect is explained by recurrent charge rearrangement in the molecule upon multiple x-ray ionization. Electrons on the methyl part are pulled towards the strongly ionized iodine atom and thereby more electrons become available for further ionization that would not have been available in independent atoms.

We now show that this charge-rearrangement-enhanced x-ray ionization of molecules (CREXIM) is much more pronounced for C_6H_5I, where the iodine atom is attached to a more electron rich chemical environment[5]. The final charges on the carbon atoms depend on their relative position to the iodine. Strikingly, we obtain an enormous enhancement for this molecule of $\simeq +20$ charges (see Fig. 1) demonstrating the huge impact of intra molecular charge rearrangement processes for the radiation damage. The large magnitude of the enhancement manifests that intra-molecular electron rearrangment is of huge relevance for prospective XFEL applications at high fluence.

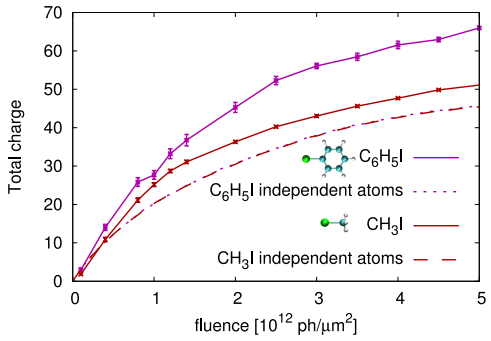

Figure 1. Average charge of CH_3I and C_6H_5I after exposure to the x-ray pulse. The solid line show results from the molecular calculation, the dotted/dashed line from the independent atom model.

References

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