# Coulomb explosion of alkali dimers on a superfluid $^4He_N$ droplet: a $^4$He-TDDFT simulation


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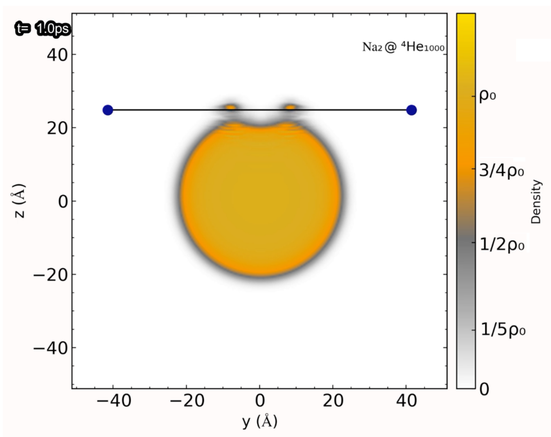
## Abstract

We simulate the Coulomb explosion upon double ionisation of alkali dimers ($Ak_2$, Ak= Li,Na, Rb, Cs) bound to the surface of a superfluid droplet of 1000 to 50000 $^4$He atoms, using $^4$He-TDDFT [1] This work was motivated by the experiment of Kristensen et al. [2],[3], who used this process to deduce the abundance ratio of neutral dimers formed on the surface of a helium nanodroplet in either the singlet X $^1\Sigma_g^+$ or the triplet a $^3\Sigma_u^+$ electronic state. $^4$He-TDDFT is a semi-empirical method describing the helium density at equilibrium (static version) or during real time dynamics, which has proven to be the best compromise between accuracy and the ability to simulate superfluid helium droplets of realistic size. We first obtain the equilibrium configuration of $Ak_2$ singlet (X $^1\Sigma_g^+$) or triplet (a $^3\Sigma_u^+$) on the droplet surface.

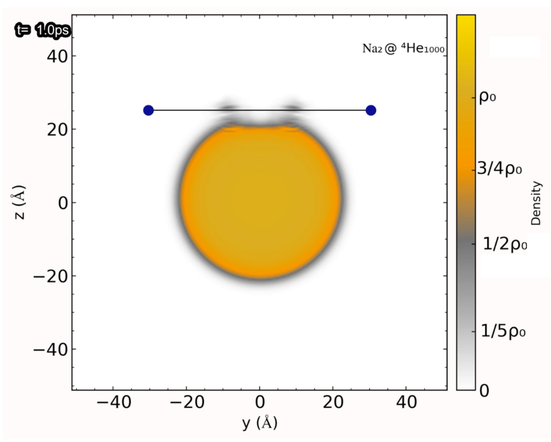

## Coulomb explosion

**$Na_2$@$^4He_{1000}$ after $\Delta t$ = 1 ps**

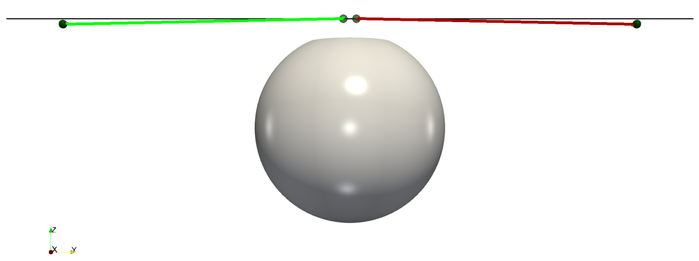
**Singlet state ($^1\Sigma_g^+$)**




**Triplet state ($^3\Sigma_u^+$)**




**Bent Trajectories**



## Conclusions and Perspectives

We have been able to simulate the Coulomb explosion of all $Ak_2$ on $He_{1000}$ and $He_{5000}$

- good agreement with experiment about the kinetic energy peaks position
- several contributions to their width identified: $Ak_2$ vibration, droplet size distribution, $Ak_2$ orientation / droplet surface. But simulated width still too narrow
- **new feature** identified: **bending** of the ions trajectories

## References


[1] M. Barranco, F. Coppens, N. Halberstadt, A. Hernando, A. Leal, D. Mateo, R. Mayol, and M. Pi, "Zero temperature DFT and TDDFT for $^4$He: A short guide for practitioners", Unpublished, 2017.

[2] H. H. Kristensen, Laser-induced rotation of alkali dimers on the surface of helium nanodroplets, Ph. D. Progress Report, June 2022.

[3] H. H. Kristensen, L. Kranabetter, C. A. Schouder, J. Arlt, F. Jensen, and H. Stapelfeldt, *Physical Review B* **107**, 023104 (2023).

[4] H. H. Kristensen, L. Kranabetter, C. A. Shouder, C. Stapper, J. Arlt, M. Mudrich, and H. Stapelfeldt, *arXiv* **2111-1254**, 1 (2022).


## $^4$He TD-DFT : Framework

The equilibrium configuration is determined by using DFT applied to $^4$He atoms at zero temperature. The total energy is expressed as a function of the $^4$He density

$$E[\rho] = T[\rho] + E_c[\rho] = \frac{\hbar^2}{2m}\int d\mathbf{r}\,|\nabla\Psi(\mathbf{r})|^2 + \int d\mathbf{r}\,\mathcal{E}_c[\rho]$$

The functional $\mathcal{E}_c[\rho]$ has been designed to reproduce the experimental dispersion curve for elementary excitations in liquid $^4$He. The ground state structure of the system is obtained solving the Euler-Lagrange (EL) equation resulting from the functional variation of the total energy $E[\rho]$ with respect to $\rho$ or $\Psi(\mathbf{r})$ under the condition of a fixed number of $^4$He atoms.

Dopants are treated as classical particles , therefore He-Dopant interaction is simply added as an external potential.

$$E[\rho] \to E[\rho] + \sum dr\; v(\mathbf{r})\rho(\mathbf{r}) \quad \to \quad \left\{-\frac{\hbar^2}{2m_4}\nabla^2 + \frac{\delta\mathcal{E}_c}{\delta\rho} + \sum_{i=1}^{2} V_{He-Ak}(|\mathbf{r}-\mathbf{r}_i|)\right\}\Psi(\mathbf{r}) = \mu\Psi(\mathbf{r})$$

## $^4$He TD-DFT

"sudden" double ionization:

$$\int \upsilon(\mathbf{r})\rho(\mathbf{r})d\mathbf{r} \quad \text{where} \quad \upsilon(\mathbf{r}) = \sum_{i=1}^{2} V_{He-Ak^+}(|\mathbf{r}-\mathbf{r}_i|)$$

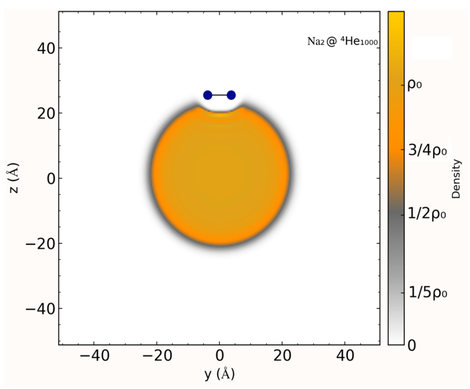
Coupled equations of motion

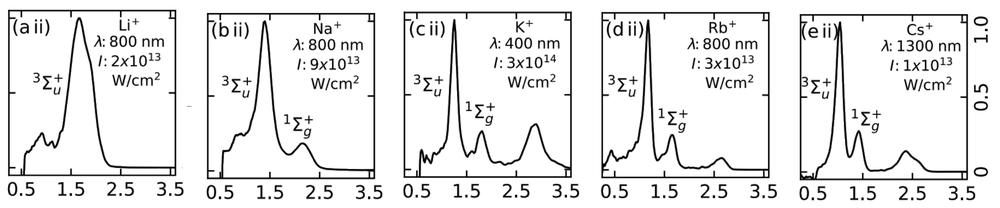
$$i\hbar\frac{\partial}{\partial t}\Psi(\mathbf{r}) = \left[-\frac{\hbar^2}{2m_{He}}\nabla^2 + \frac{\partial\mathcal{E}_c}{\partial\rho(\mathbf{r})} + \sum_{i=1}^{2} V_{He-Ak^+}(|\mathbf{r}-\mathbf{r}_i|)\right]\Psi(\mathbf{r})$$

$$m_{Ak^+}\ddot{\mathbf{r}}_j = -\left\{\int d\mathbf{r}\; V_{He-Ak^+}|\mathbf{r}-\mathbf{r}_j|\;\nabla\rho(\mathbf{r}) + (-1)^{-i}\frac{\mathbf{R}}{R}\frac{d}{dR}[V_{Ak^+-Ak^+}(R)]|_{R=|\mathbf{r}_2-\mathbf{r}_1|}\right\}$$

## Results

Once the statics has been done, we switch the potential from Ak-Ak to $Ak^+$-$Ak^+$ in order to ignite the Coulomb explosion.



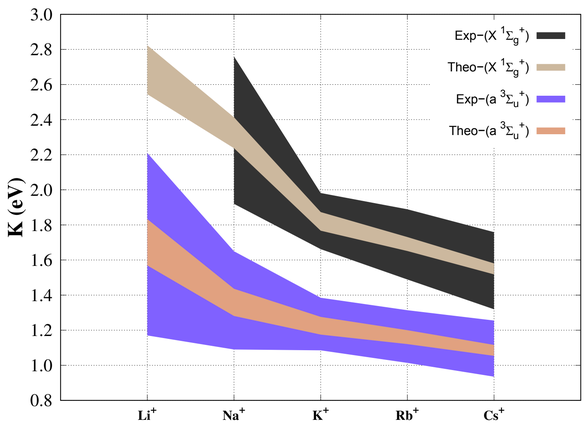




**Experimental Kinetic energy (eV): $Li^+$,$Na^+$,$K^+$,$Rb^+$,$Cs^+$ [3]**

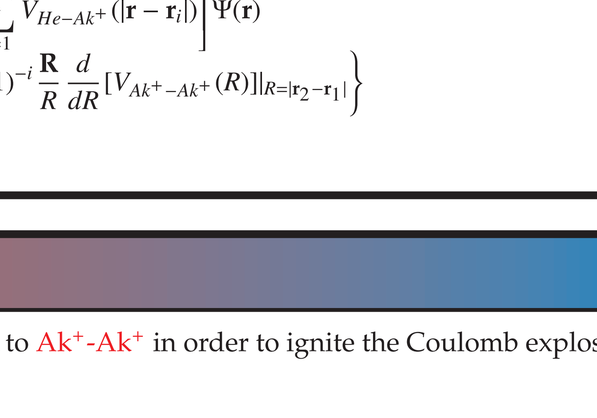
**Kinetic energy per atoms (eV) at req**

| $Ak_2$ | Our work | | Experimental Ref [4] | |
|---|---|---|---|---|
| | X $^1\Sigma_g^+$ | a $^3\Sigma_u^+$ | X $^1\Sigma_g^+$ | a $^3\Sigma_u^+$ |
| Li | 2.690 | 1.743 | - | 1.69 |
| Na | 2.327 | 1.370 | 2.34 | 1.39 |
| K | 1.822 | 1.235 | 1.83 | 1.26 |
| Rb | 1.689 | 1.164 | 1.71 | 1.19 |
| Cs | 1.539 | 1.095 | 1.42 | 1.06 |

**Width of the kinetic energy peaks due to $Ak_2$ vibration**




**Droplet size contribution**




**Final angle deviation**