

Time-resolved and XUV spectroscopy of helium nanodroplets

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Synopsis The ultrafast dynamics of pure and doped helium nanodroplets is studied using VIS and XUV femto-second pump-probe spectroscopy in combination with ion and electron imaging detection.

Helium (He) nanodroplets currently attract considerable interest mainly for two reasons. 1) Due to their quantum fluid nature He nanodroplets feature extraordinary properties such as microscopic superfluidity. 2) He nanodroplets can be considered as a nearly ideal matrix for spectroscopy of embedded molecules and aggregates due to their transparency for light up to the extreme ultraviolet (XUV) spectral range, and their ability to efficiently aggregate and cool embedded species.

However, the short-time dynamics of He nanodroplets is still largely unexplored. Recently, strong perturbations of the rotational motion of embedded molecules induced by the He droplet environment was observed for the case of impulsive excitation [1]. In this contribution we study the ultrafast response to laser excitation of a prototype system consisting of an alkali metal atom residing at the surface of He nanodroplets. Using femtosecond pump-probe spectroscopy in combination with ion and electron imaging detection we follow in real-time the desorption of the excited atom off the He droplet surface (see Fig. 1) as well as the formation of alkali-He exciplex molecules.

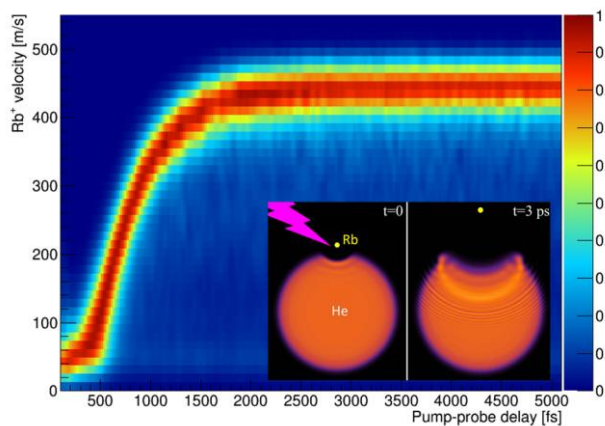


Figure 1. Time-evolution of the velocity of an excited Rb atom ejected off the surface of a He nanodroplet. Inset: TDDFT simulation [2].

Upon irradiation of He droplets with XUV light at $h\nu \geq 21$ eV, where He droplets are strongly absorbing, a complex photo dynamics is initiated by the excitation or ionization of He atoms inside the droplets [3]. The dynamic of relaxation, fragmentation, as well as indirect ionization processes are studied using synchrotron and free-electron-laser (Fermi@Elettra) radiation. In particular, the real-time dynamics of pure He nanodroplets excited by tunable femtosecond XUV-pulses is studied in a pump-probe experiment. The time-resolved photoelectron spectra reveal ultrafast intra- as well as interband relaxation dynamics, see Fig. 2.

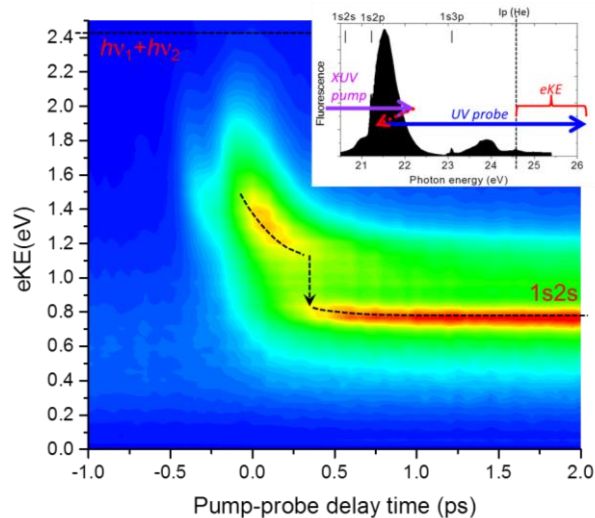


Figure 2. Pump-probe delay dependent photoelectron spectrum of pure He nanodroplets measured by XUV (22.2eV)/UV (4.8eV) fs pump-probe R2PI.

References

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