Mixed quantum-classical fragmentation dynamics of Ar_nHe_{1000} $(n = 3 - 5)$ ionized by electron impact

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At the very low temperature of their formation ($T \approx 0.4\,\mathrm{K}$), doped helium nanodroplets are highly quantum systems. Modeling their dynamics by classical methods would therefore significantly overestimate the cage effect exerted by helium atoms on the dopant. A decade ago, the fragmentation of Ne_nHe_{100} $(n = 4 - 6)$ ionized by electron impact has been treated by coupling a surface hopping method for the nonadiabatic dynamics of the Ne_n^+ dopant with a classical dynamics, on an effective potential-energy surface including zero-point effects, for helium atoms [1,2]. A broad variety of $Ne_p^+He_q$ $(p \leq n,\ q \leq 100)$ fragments has been detected in theoretical mass spectra after as few as 30 picoseconds of dynamics, most of these fragments being attributed to a dopant ejection mechanism. Although this study was a first step in the right direction, these clusters were too small compared with experimental sizes of typically $10^3 - 10^6$ helium atoms [3]. The present work extends the aforementioned theoretical study to larger argon doped helium nanodroplets, namely $Ar_n^+He_{1000}$ clusters $(n = 3 - 5)$ [4]. While the nonadiabatic dynamics of the ionic dopant and the dynamics of the helium environment are modeled the same way as in our earlier works on neon clusters, the larger number of helium atoms enables the trapping of dopant atoms which favors geminate recombinations, a physical process never observed when investigating the fragmentation of small $Ne_n^+He_{100}$ clusters. Trapping and dopant ejections thus compete with each other during the fragmentation of $Ar_n^+He_{1000}$, which deeply alters the theoretical mass spectra and the kinetics of departing helium atoms. This point is discussed together with the influence of the initial dopant ionization on the fragmentation mechanism. Possible technical improvements of the method are eventually presented to generalize our approach to larger doped helium nanodroplets and make our results more representative of experimental mass spectra.

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