Direct inner-shell photoionization of Xe atoms embedded in helium nanodroplets

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Abstract

We present the first measurements of photoelectron spectra of atomic clusters embedded in superfluid helium (He) nanodroplets. Owing to the large absorption cross section of xenon (Xe) around 100 eV photon energy (4d inner-shell ionization), direct dopant photoionization exceeds charge transfer ionization via the ionized He droplets. Despite the predominant creation of Xe\(^{2+}\) and Xe\(^{3+}\) by subsequent Auger decay of free Xe atoms, for Xe embedded in He droplets only singly charged Xe\(^{+}\), \(k = 1, 2, 3\) fragments are observed. Broad Xe\(^{+}\) ion kinetic-energy distributions indicate Coulomb explosion of the ions due to electron transfer to the primary Auger ions from surrounding neutral atoms. The electron spectra correlated with Xe ions emitted from the He nanodroplets contain a low-energy feature and nearly unshifted Xe photolines. These results pave the way to extreme ultraviolet and x-ray photoelectron spectroscopy of clusters and molecular complexes embedded in He nanodroplets.

Keywords: inner-shell photoionization, doped He nanodroplets, Auger decay, charge transfer, Coulomb explosion, electron-ion coincidence spectroscopy, synchrotron radiation

(Some figures may appear in colour only in the online journal)

1. Introduction

Helium (He) nanodroplets are widely used as an ultracold matrix for spectroscopy of embedded molecules and nanostuctures [1, 2]. The main benefits of He nanodroplets are the high resolution of absorption spectra in the infrared and visible spectral regions and the property of He droplets to efficiently form molecular aggregates that thermalize to the droplet temperature of 0.37 K. Performing spectroscopy at higher photon energies where the dopants or even the He droplets are directly ionized is not straightforward, though; the strong interaction of photoions and electrons with the He droplet tends to massively shift and broaden the electron spectral lines as well as to alter the fragmentation dynamics compared to the gas phase [3]. Therefore, only few photoelectron spectroscopic studies of dopants have been reported, all of which employed resonant multi-photon ionization schemes [4–10].

However, single-photon photoionization of doped He nanodroplets has recently turned out to be a rewarding approach for studying various types of fundamental correlated electronic decay processes such as interatomic Coulombic decay (ICD) [11–15] and electron-transfer mediated decay (ETMD) [16–18]. Although the photon energy exceeded the dopant's ionization energy \(E_i\) in those studies, dopants were always ionized indirectly through the excited or ionized He. This is due to the large total absorption cross section of He nanodroplets containing thousands of He atoms (\(\sim 25\) Mbarn per He atom for the dominant 1s\(^2\)p\(^1\)P absorption resonance at the photon energy \(h\nu = 21.6\) eV) that usually largely exceeds the absorption cross section of one or a few dopant atoms or molecules. Excitations, He\(^+\), and positive charges, He\(^{2+}\), efficiently migrate through the He droplet to the dopant which...
is then ionized by transfer of energy or charge, respectively [15, 19–22]. Large differences in the Penning ionization efficiency and the structure of the Penning electron spectra were found for dopants (alkali metals) attached to the surface of He nanodroplets compared to those immersed in the droplet interior. This was rationalized by the tendency of He$^+$ to migrate towards the droplet surface [23, 24], whereas He$^+$ remains in the bulk of the droplets [23, 25]. Using photoelectron–photoion coincidence (PEPICO) detection, we have previously measured high Penning ionization yields for alkali metals, whereas the efficiency of Penning ionization for heavier rare gas atoms was lower than that for charge transfer ionization [21]. The Penning ionization electron spectra were found to feature either sharp lines reflecting the He energy levels and the dopants’ $E_i$ [15, 21], a broad distribution peaked at low energies [22], or a combination of both [20, 26].

Here, we present the first experimental study where dopant atoms attached to He nanodroplets are directly photoionized and electron and ion spectra are recorded. This is achieved using Xe as a dopant and setting $h\nu \sim 100$ eV where Xe features a pronounced maximum of the 4d-shell ionization cross section, whereas the absorption cross section of He is down by a factor $\sim 1/20$ compared to the value near the ionization energy of He, $E_i^{He}$. Photoions from free atoms in the gas phase are mostly produced in doubly and triply charged states as a result of normal or cascaded Auger decay, respectively. In contrast, from doped He droplets mostly singly charged Xe$^+$ as well as small Xe$^{n+}$ clusters are emitted. These points at efficient partial neutralization of multiply charged cations in He nanodroplets by electron transfer to the dopant photoion from neutral dopant atoms surrounding it. Electron spectra exhibit sharp unshifted photolines from the embedded Xe clusters as well as a pronounced low-energy distribution indicative for electron–He scattering and electron localization.

### 2. Experimental

The experiments are performed using a He nanodroplet apparatus combined with a velocity-map imaging-photoelectron–photoion coincidence (VMI-PEPICO) spectrometer installed at the GasPhase beamline of Elektra-Sincrotrone Trieste, Italy. The apparatus has been described in detail elsewhere [21, 27]. Briefly, a beam of He nanodroplets is produced by continuously expanding pressurized He (50 bar) of high purity out of a cold nozzle (14 K) with a diameter of 5 μm into vacuum, resulting in a mean droplet size of $N_{He} = 2.3 \times 10^4$ He atoms per droplet. The He droplets were doped with Xe atoms by leaking Xe gas into a doping gas cell of length 30 mm. The measurements presented in this paper were done at a Xe pressure in the doping cell of 4.3 $\times 10^{-4}$ mbar. This corresponds to a mean number of 24 Xe dopants per He droplet. A mechanical beam chopper at the entrance of the doping chamber is used for discriminating droplet-beam correlated signals from the background. In the detector chamber, the He droplet beam crosses the synchrotron beam at the center of the VMI-PEPICO detector at right angles. By detecting either electrons or ions with the VMI detector in coincidence with the corresponding particles of opposite charge on the TOF detector, we obtain either ion mass-correlated electron images or mass-selected ion images. Kinetic-energy distributions of electrons and ions are obtained by Abel inversion of the images [28]. The energy resolution of the electron spectra obtained in this way is $\Delta E/E = 6\%$.

### 3. Results and discussion

In our previous PEPICO study of Ar-doped He nanodroplets it appeared that heavier rare gas atoms solvated in the droplet interior are inefficiently Penning ionized through excited He [21]. In contrast, Wang et al. had previously reported well-resolved Penning electron spectra of Kr and Xe indicating that Penning ionization of Kr and Xe embedded in He nanodroplets is quite efficient [20].

To resolve these conflicting findings, we record the total electron yield of Xe-doped He nanodroplets in the wide photon energy range $h\nu = 20–160$ eV. Those electrons emitted from Xe dopants embedded in He droplets are extracted from the data by first subtracting from the total electron signal (chopper open) those electrons emitted by ionization of the background gas (chopper closed). Then, we subtract from the measurement done with Xe doping a reference measurement where the Xe doping was turned off. The resulting electron yield spectrum is shown in figure 1.

In the range $h\nu < E_i^{He}$, the electron yield closely follows the absorption spectrum of pure He nanodroplets, which is dominated by the 1s2p $^1P$ absorption resonance of He nanodroplets at $h\nu = 21.6$ eV [29]. We conclude that Xe is indeed efficiently Penning ionized by excited He nanodroplets, similar to our previous finding for alkali and alkaline-earth metals [3, 17, 21]. The reason why we previously measured much lower Penning ionization signals from rare-gas dopants than from the metals was that rare-gas Penning ions formed inside the He droplets tend to remain bound to the droplets even if the Penning electrons are emitted. It is well known that atomic ions as well as molecular ions in their vibronic ground state form stable complexes with He atoms (so-called ‘Atkins snowballs’) which are deeply bound inside He nanodroplets [30–33]. Even alkali ions created in close vicinity of the He droplet surface tend to sink into the droplet and become solvated [34–36]. As in our previous experiments we always detected electrons and ions in coincidence, both escaped detection. Nevertheless, our conclusion that the surface location of alkali atoms facilitates He droplet Penning ionization remains true. The proportion of Penning ionization signals measured at $h\nu = 21.6$ eV versus dopant ionizations by charge transfer at $h\nu > 24.6$ eV is about 5 times higher for alkali metals than for Xe [21]. Besides, it has been shown by electron impact ionization that small alkali clusters residing at the droplet surface are more efficiently Penning ionized than large alkali clusters which sink into the droplet interior [37, 38].

At higher extreme ultraviolet (XUV) photon energies $h\nu > 70$ eV, the yield of electrons is lower than that measured at the He 1s2p $^1P$ resonance but clearly shows a broad maximum centered around $h\nu \sim 100$ eV. In this range of $h\nu$ the electron yield closely follows the absorption cross section of Xe atoms which is dominated by a maximum of the 4d-subshell
photoionization cross section, also called ‘giant resonance’ [39, 40]. This is a clear indication that now the detected electrons are mostly emitted by the Xe dopants. Photoionization of the He droplets followed by charge transfer ionization of the Xe dopants, which is the dominant dopant-ionization mechanism near \( E_{\text{He}} \), contributes to a lesser extent. This is due to the large difference in absorption cross sections of Xe (23.6 Mbarn) and He (0.52 Mbarn) at \( h\nu = 90 \) eV [41]. Given the droplet size of about \( 2.3 \times 10^6 \) He atoms and the estimated Xe dopant clusters size of 24 Xe atoms, we obtain a ratio of the efficiencies of direct photoionization of embedded Xe vs indirect charge transfer ionization of about 7 assuming a charge transfer ionization probability of the Xe cluster of 1% [42, 43].

This value is in good agreement with the signal contrast from on-resonant (\( h\nu \sim 100 \) eV) Xe photoionization with respect to the off-resonant (\( h\nu \sim 150 \) eV) background measured here (figure 1). Thus, we have demonstrated for the first time that direct one-photon ionization of dopants embedded in He nanodroplets is possible, at least at high XUV photon energies \( h\nu \gg E_{\text{He}} \) where He nanodroplets are nearly transparent.

When Xe is 4d-subshell ionized, a cascaded Auger decay takes place resulting in multiply charged Xe ions [40]. Figure 2(c) shows typical mass spectra recorded for Xe atoms in the background gas (black line) and for Xe embedded in He nanodroplets (red line) at \( h\nu = 90 \) eV. Clearly, the dominant charge states from Auger decay are Xe\(^{2+}\) and Xe\(^{3+}\), whereas Xe\(^+\) is hardly visible when normalizing the ion signal scale to the Xe\(^{2+}\) peak. Note that the abundances of the charged Xe\(^{2+}\) and Xe\(^{3+}\) ions are likely enhanced compared to Xe\(^+\) and in particular to Xe\(^{2+}\) and Xe\(^{3+}\) due to a higher detection sensitivity. In contrast, just below and above \( E_{\text{He}} \) (figures 2(a) and (b)), Xe\(^+\) is by far the most abundant product. The small contribution of Xe\(^{2+}\) in the mass spectrum at \( h\nu = 25 \) eV is likely due to one-photon double ionization of Xe by second-order synchrotron radiation which is quite abundant at that photon energy.

The He droplet-correlated Xe\(^+\) signal at \( h\nu = 19 \) eV < \( E_{\text{He}} \) (red line in figure 2(a)) is nearly absent. The small Xe\(^+\) peak likely stems from imperfect discrimination of the He droplet-correlated signal from the background. As the He droplets are neither excited nor ionized at that photon energy, indirect ionization of Xe through the He is suppressed and direct photoionization of Xe dopants does not significantly contribute to the ion signal. At \( h\nu = 25 \) eV > \( E_{\text{He}} \) (red line in figure 2(b)), however, He droplet-correlated Xe ions are present mostly as small Xe\(^{k+}\) clusters, \( k = 1,2,3 \). The fragmentation of these small Xe dopant clusters is due to charge transfer ionization through the ionized He nanodroplets, as it has been discussed in reference [43].

One main result of this work is that at \( h\nu = 90 \) eV, the He droplet-correlated Xe\(^+\) mass spectrum (red line in figure 2(c)) again consists of small singly charged Xe\(^{+}\) clusters, although Xe ionization is mainly due to direct Xe 4d-ionization which predominantly creates Xe\(^{2+}\) and Xe\(^{3+}\). Thus, multiply charged ions in He nanodroplets are very efficiently partly neutralized. Electron transfer from neighboring neutral atoms to the multiply charged ions can occur on a subfemtosecond time scale, i.e. faster than the Auger process [44]. Partial neutralization of doubly charged metals in He nanodroplets created by electron-transfer mediated decay was observed previously.

**Figure 1.** Electron yield spectrum due to Xe atoms embedded in He nanodroplets as a function of the photon energy. The signal below the ionization energy of He (to the left of the break in the \( h\nu \)-axis) is due to indirect ionization of Xe through excited and autoionized He, the part at \( h\nu > 70 \) eV is mostly due to direct photoionization of the Xe dopants.

**Figure 2.** Mass spectra of Xe-doped He nanodroplets recorded at different photon energies. The ion signal correlated to the doped He nanodroplets (red line) is discriminated from the background (black line) through a mechanical chopper that interrupts the He nanodroplet beam.
Even pure Xe clusters irradiated by soft and hard x-rays have recently been found to efficiently quench high charge states created by Auger ionization \cite{14, 17}. This was interpreted by electron–ion recombination which is a common process in expanding nanoplasmas \cite{45}. Here, we show that electron transfer to multiply charged ions is highly efficient even in the absence of a nanoplasma. Likely, in \cite{45} also electron transfer from neutral Xe contributed to the measured highly abundant Xe$^+$ signals \cite{45}. In our experiment (figure 2(c)), where the He nanodroplets contain small Xe clusters, likely electron transfer between Xe atoms is the main mechanism. We note that even when we reduced the number of doped Xe atoms down to the detection limit of Xe ions from He droplets in an attempt to dope the He droplets by single Xe atoms, no Xe$^{2+}$ were present although the latter cannot be neutralized by He as $E_{\text{v}}^{\text{He}^+} < E_{\text{v}}^{\text{Xe}^+}$. The likely reason is that a multiply charged cation is strongly bound to a He droplet by forming a snowball complex \cite{30} and thus evades its detection. Likewise, no experimental evidence for charge transfer to the multiply charged Xe$^{3+}$ from expanding nanoplasmas \cite{45}. Here, we show that we note that the Xe$^+$ ionization in conjunction with electron transfer (figure 2(c)), where the Xe$^+$ peak is higher and broader in the latter case. This is likely due to Coulomb explosion of the two or three Xe$^+$ ions formed from Xe$^{2+}$ or Xe$^{3+}$ by electron transfer, respectively. When two ions are formed with substantial kinetic energy, they are less prone to be trapped by the He droplet and are therefore detected with higher probability \cite{14, 17}.

Further evidence for the formation of Xe$^+$ by Coulomb explosion is obtained from directly measuring the ion kinetic energy by velocity-map imaging the Xe$^+$ on a position sensitive detector in the ion-imaging mode. The Xe$^+$ and Xe$^{2+}$ ion kinetic energy distributions inferred from ion images are displayed in figure 3 in comparison with the ion kinetic energy distributions for Xe$^{2+}$ and Xe$^{3+}$ from free Xe atoms. The Xe$^+$ ion spectrum consists of a broad feature that peaks around 1.5 eV and extends up to 6.5 eV, whereas the Xe$^{2+}$ and Xe$^{3+}$ spectra both exhibit only a very narrow peak at 0 eV. The width of these peaks reflects the experimental resolution.

For Xe dimers, the bond length is $R = 4.36$ Å \cite{47}. Assuming instantaneous formation of an ion pair Xe$^+$ + Xe$^+$ by Auger decay and electron transfer, the kinetic energy release (KER) due to Coulomb explosion according to the repulsive Coulomb potential $\epsilon^2/(4\pi\varepsilon_0 R)$ is estimated to 3.3 eV. Thus, each Xe$^+$ ion acquires a kinetic energy of 1.7 eV, which is in good agreement with the maximum of the measured kinetic energy distribution. For larger Xe clusters, the KER is expected to be higher since the interatomic distance between two nearest-neighbor atoms is slightly shorter as it approaches the bond length in bulk Xe, 4.26 Å \cite{48}.

The tail in the kinetic energy distribution extending up to 6.5 eV in the Xe$^+$ spectra is likely due to Coulomb explosion of three Xe$^+$ ions after creation of one Xe$^{3+}$+ by Auger decay followed by electron charge transfer from two neighboring Xe atoms. The kinetic energy of the Xe$^+$ for the Coulomb explosion of a Xe trimer system are expected to range between 3.3 eV and 4.5 eV depending on the initial configuration. Furthermore, when Coulomb explosion occurs in a larger Xe cluster where one charge is localized on one Xe atom and the other is localized on the remaining cluster Xe$_{n-1}$, the Xe$^+$ acquires a kinetic energy up to the full KER in the limit of a very large Xe$_{n-1}$. This kinematic effect adds to the asymmetric broadening of the Xe$^+$ kinetic energy distribution towards higher energies.

The Xe$^{2+}$ ion spectrum shows a bimodal distribution with a trailing edge (0.5–4 eV) that resembles the one of the Xe$^+$ ion spectrum (1–6 eV) but scaled down to lower energy. Again, this may be due to the kinematic effect, from which we expect a factor of 2 lower energy of Xe$^{2+}$ than Xe$^+$ for the case that Coulomb explosion occurs from the Xe$^{2+}$ system. The peak at < 0.1 eV seen in the Xe$^{2+}$ spectrum might be related to a non-thermal ejection process that occurs for vibrationally excited molecular ions \cite{49}, assuming that part of the Coulomb exploding Xe$^{2+}$ are fully decelerated by collisions with surrounding Xe and He atoms in the droplets prior to ejection.

Figure 4 shows electron spectra for Xe atoms measured in coincidence with atomic Xe$^{2+}$ and Xe$^{3+}$ ions (panel (a)) as well as those measured in coincidence with Xe$^+$, Xe$^{2+}$ and Xe$^{3+}$ emitted from He droplets (panels (b) and (c)). The two energy-resolved 4d$^{-1}_{5/2}$ and 4d$^{-1}_{3/2}$ lines seen in the atomic PES spectra for Xe$^{2+}$ and Xe$^{3+}$ are also present in the He...
Electron spectra recorded for Xe atoms (a) and for He droplets doped with Xe atoms (b) and (c) at photon energy $h\nu = 90\,\text{eV}$. Black and red spectrum in (a) are recorded in coincidence with Xe$^{2+}$ and Xe$^{3+}$, respectively. The spectra denoted in black, red and blue in (b) recorded in coincidence with Xe$^{+}$, Xe$^{2+}$ and Xe$^{3+}$, respectively. The black, red and blue curves in (c) show electron spectra measured in double coincidence with Xe$^{+}$/Xe$^{2+}$, Xe$^{2+}$/Xe$^{3+}$ and Xe$^{+}$/Xe$^{3+}$, respectively. The vertical dashed lines show the energy positions of the atomic 4d$^{5/2}$ and 4d$^{3/2}$ lines according to [50].

Figure 5. Close-ups of the 4d$^{5/2}$ and 4d$^{3/2}$ photolines in case of free Xe atoms and Xe clusters formed inside He nanodroplets and irradiated with an XUV photons of energy $h\nu = 90\,\text{eV}$. The solid curves depicted in red are Lorentzian fit functions to determine the energy positions of the 4d$^{5/2}$ and 4d$^{3/2}$ photolines in each electron spectrum.
clusters. The fact that the photoline spectra are nearly unshifted (figure 4(b)) may indicate that Xe\textsuperscript{+} atomic fragments tend to be emitted from 4d-ionized Xe\textsubscript{2} or small Xe clusters whose electron spectra are only weakly perturbed, whereas larger Xe clusters, which feature more strongly perturbed electron spectra, fragment more likely into Xe\textsubscript{2}\textsuperscript{+} and Xe\textsubscript{3}\textsuperscript{+}.

### 4. Conclusion

We have reported the first experimental evidence of direct single-photon ionization of dopants embedded in He nanodroplets. We exploit the large absorption cross section for 4d inner-shell ionization of Xe at a photon energy around \( h\nu = 100 \text{ eV} \) where He has a low absorption cross section. For Xe clusters formed inside He nanodroplets, multiply charged Xe atoms created by Auger decay are efficiently partially neutralized into singly charged Xe\textsuperscript{k+}, \( k = 1, 2, 3 \), clusters by electron transfer from surrounding neutral Xe and He atoms. Subsequent Coulomb explosion generates Xe\textsuperscript{k+} ions with up to 6.5 eV of kinetic energy. The electron spectra of droplet-bound Xe clusters feature both an enhanced low-energy component indicative for electron–He scattering, and nearly unshifted 4d photoline.

These results demonstrate that photoelectron spectroscopy of clusters embedded in He nanodroplets in the XUV range is possible. Very likely, the same holds for x-rays, provided a sufficiently sensitive detection scheme is used that copes with the low target density. This paves the way to x-ray photoelectron spectroscopy of unconventional atomic clusters and molecular complexes, which can form in He nanodroplets owing to their unique quantum fluid properties [59–62]. Furthermore, the selective multiple ionization of dopants in He nanodroplets is an efficient mechanism for igniting a nanoplasma [63]. Probing the dynamics of nanoplasmas by ultrashort XUV and x-ray pulses has attracted considerable attention in the free-electron laser science community [46, 64–66].

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