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# Multipronged Mapping to the Dynamics of a Barium Atom deposited on Argon Clusters - Supplementary Material

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The dynamics of an electronically excited barium atom deposited at the surface of an  $Ar_{\approx 500}$  cluster was explored in a multipronged approach which associates information from frequency-resolved nanosecond experiments and information from femtosecond time-resolved experiments. In both types of experiments, the dynamics is monitored by photoelectron and photoion spectroscopy.

PACS numbers:

## PHOTOIONS IN THE FREQUENCY-RESOLVED EXPERIMENT

We have seen in Sec. 3.2 of the main paper that when reversing the acceleration voltages of the VMI, photoions can be imaged on the detector instead of photoelectrons. By gating the polarization voltage of the VMI,  $Ba^+$  ions are detected selectively. This was performed in the frequency resolved experiment. The resulting photoion images were analyzed in the same way as described in the main paper for the photoelectron images. The corresponding results are shown in Fig. S1 as a 3D-plot, where the  $Ba^+$  signal intensity is plotted as a function of both the velocity of the  $Ba^+$  ions and the wavenumber of the pump laser. The dip observed in the signal intensity across Fig. S1 at  $180\text{ m.s}^{-1}$  is due to a dark region of the detector.

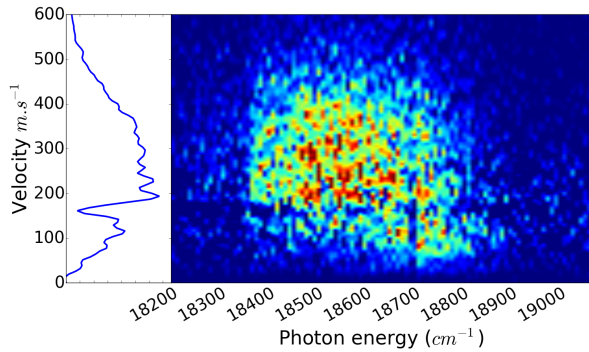


FIG. S1:  $Ba^+$  photoion signal as a function of both the photoion velocity (vertical axis) and wavenumber of the pump laser (horizontal axis). The signal intensity is represented using the same colormap as in the main paper for photoelectrons.

## EJECTED BARIUM FOR CLUSTERS

We have discussed in section 4.1 of the main paper that all the barium atoms which are present in the beam are bound to the argon clusters.

As a results, the signal which is reported in Fig. S1 find their origin in barium atoms that are carried by the argon cluster. Since this signal corresponds to  $Ba^+$  ions, either it is due to barium atoms that have been ejected off the cluster prior ionization by the probe laser (i.e. to barium atoms that have been ejected after they have been excited by the pump laser), or to  $Ba^+$  ion that have desorbed after ionization. Such situation was encountered in the case of Ba atom deposited on helium droplets [1].

## ACTION SPECTRA

The important point to observe in Fig. S1 is that the action spectrum is not the same whether low or high and middle velocities are considered. The optimum laser wavenumber is indeed observed between  $18\,600$  and  $18\,800\text{ cm}^{-1}$  for velocities smaller than  $100\text{ m.s}^{-1}$ , whereas for higher velocities the action spectrum is broad and covers the  $18\,400$ - $18\,750\text{ cm}^{-1}$  range. For this reason, the velocity distribution of the  $Ba^+$  ions was fitted as the sum of two Gaussian distributions, one centered at  $98\text{ m.s}^{-1}$  and the other at  $286\text{ m.s}^{-1}$  plotted.

The top panel of Fig. S2 shows the measured velocity distribution (full blue curve) obtained when integrating the  $Ba^+$  ion signal of Fig. S1 over the wavenumber of the pump laser. The two Gaussian distributions which fit this velocity are shown in the same panel as dashed blue and red curves together with the fit (full green curve). When the  $Ba^+$  ion signal of Fig. S1 is integrated over the two Gaussian distributions, action spectra are obtained. They are shown in the bottom panel of Fig. S2 (full curves with the color of the corresponding velocity dis-

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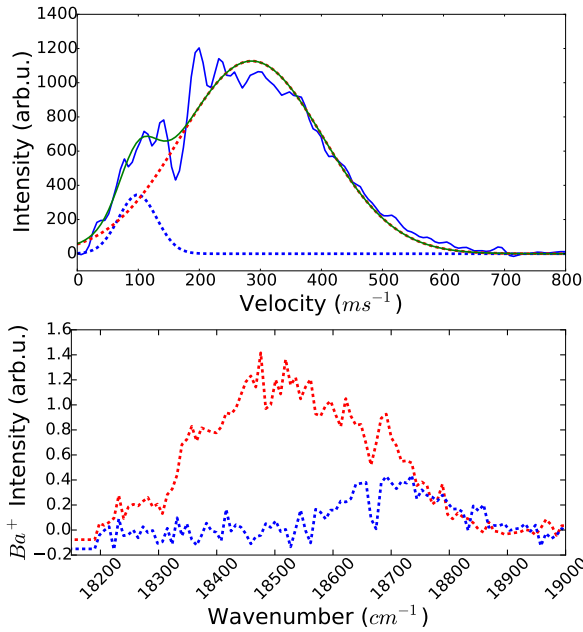


FIG. S2: (Top) Measured  $Ba^+$  velocity distributions (blue full curve) and its fit (green full curve) by two Gaussian distributions (blue and red dashed curves). (Bottom) Action spectra (blue and red full curves) corresponding to the two Gaussian velocity distributions shown as blue and red dashed curves in the top panel (see text for details).

tribution).

The action spectrum shown in blue in the present Fig. S2 is that shown in Fig. 2 of the main paper. The corresponding atoms have an average velocity of  $98 m.s^{-1}$ , *i.e.* an average kinetic energy of 0.007 eV. This action spectrum is discussed in the main paper (Sec. 4.3) as documenting barium atoms that are ejected out of the cluster after they have been excited on the cluster by the pump laser in a one-photon process.

The other action spectrum, shown in red in the present Fig. S2) documents barium atoms which are ejected with a much larger velocity ( $286 m.s^{-1}$ ). Its shape is very close

to that of the  $\Sigma$ -like band, which is shown in Fig. 5 (top panel) of the main paper. This signal is therefore associated with the excitation of Ba atoms that are bound to the argon cluster, but unlike those documented by the blue action spectrum they are not ejected off the cluster prior ionization in a one photon process. Two very different processes may be invoked to interpret the red action spectrum.

The first possible interpretation of the red action spectrum follows the observation reported by Loginov and Drabbels where  $Ba^+$  ions are ejected after ionization of Ba on the droplet [1]. Here, because of unfavorable energetics, a 2-photon ionization by the probe laser is required. As in the helium droplet experiment, the exact nature of the ejection process is not clearly identified yet.

The second possible interpretation of the red action spectrum involves a sequential 3-step process. In the first step, barium is excited to the  $\Sigma$ -like state by the pump laser. Then it undergoes a non-adiabatic energy transfer to a lower electronic state and its recoil energy is large enough to push it off the cluster. Such a process where the initial excitation of barium is transferred to the  $6s6p^3P_{0,1,2}$  triplet has been documented already [2]. Absorption of a single probe photon (3.50 eV) by  $Ba(6s6p^3P_2)$  provides barium with 5.18 eV energy, not enough for ionization. The observation of  $Ba^+$  ions thus require a 2-photon ionization. The corresponding photoelectrons are expected with a 3.45 eV energy. Unfortunately the latter is too large to be detected in the present experiment. This precludes discriminating between the two processes that have just been proposed to interpret the action spectrum shown in red in Fig. S2).

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- [1] E. Loginov and M. Drabbels, *J. Chem. Phys.*, 2012, **136**, 154302.
  - [2] M. Osborne, M.-A. Gaveau, C. Ge, O. Sublemontier, J.-M. Mestdagh and J. P. Visticot, *J. Chem. Phys.*, 1997, **106**, 1449.