

Vibrational autodetachment of highly excited SF₆⁻ in a cryogenic electrostatic ion beam trap

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SF₆ is a widely used gaseous dielectric with insulating properties due to both its large cross section for electron capture and the stability of the formed anions. We have therefore measured the vibrational autodetachment rate from SF₆⁻ anions produced in a Cs sputter ion source at high rotational and vibrational excitation [1]. The ions were stored at 6 keV kinetic energy in an electrostatic ion beam trap and the rate of neutral particles emerging from the trap were measured as a function of storage time. The trap was cooled to <15 K by a closed-cycle helium refrigeration system. The extremely low pressure (few 10⁻¹² mbar) achieved by cryopumping results in a very low background of collision-induced charge exchange reactions. This enabled us to follow the emission rate over five orders of magnitude and for up to 100 ms, which leaves sufficient time for adiabatic detachment. We have compared the experimental data to a statistical rate model developed from semi-empirical detachment rates [2] that account for intra-molecular vibrational relaxation as well as for the threshold behaviour of electron emission, together with calculations [3] of the vibrational structure of the anion and the neutral molecule, and have also included the competing relaxation channel of radiative cooling.

Assuming thermal populations of the rotational and vibrational levels of the anion we have fitted this model to the rates to extract initial rotational and vibrational temperatures of the ions under different operating conditions of the ion source. In all cases the statistical rate model offers a very good reproduction of the experimental data, over the full range of observed rates and times. The temperatures are found to generally increase with the pressure of the SF₆ precursor gas in the ion source, which suggests that the vibrational levels are populated in ion-neutral collisions during ion acceleration. In order to also obtain information about fundamental molecular properties from the experiment, we have repeated this procedure for different combinations of values for the adiabatic electron affinity of SF₆ and of the radiative cooling time constant of the anion. From this procedure, we are able to deduce the adiabatic electron affinity of SF₆ and the radiative stabilization rate of excited SF₆⁻ near the detachment threshold.

References:

[1] S. Menk et al, in preparation.

[2] J. Troe, T. M. Miller and A. A. Viggiano, J. Chem. Phys. **136** 121102 (2012).

[3] W. Eisfeld, J. Chem. Phys. **134** 054303 (2011).