



The negative ions adsorption on the ion source surface at the resonant electron capture by molecules and measurements of the ion lifetime



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ABSTRACT

Using a static sector magnetic mass spectrometer MI-1201B, modified to record negative ions for resonant electron capture by molecules, the ion extraction time from the ionization chamber combined with this apparatus was measured up to several hundred microseconds. It was shown that the values obtained are anomalously great and they arise from the fact that part of the formed in the ionization chamber ions are adsorbed onto the chamber's surface. Those of them that were produced initially in the gas phase, namely the transient negative ions, with a finite lifetime ($\sim 1 \mu\text{s}$ and greater), transform themselves, on the surface, into 'infinitely living' (stable) ions. As a result, the contribution of the stable ions to the total ion beam falsifies the ion's lifetime measurements performed with this instrument.

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1. Introduction

Transient negative ion formation during the resonance capture of slow (0–15 eV) electrons by molecules is a process of fundamental importance and has been studied intensively in the last few decades [1–3]. In this area of research, the average lifetime of the negative molecular ions ($^{\text{a}}\text{M}^-$), with respect to the additional electron autodetachment (τ_{a}), is one of the key parameters [4]. This ion characteristic can be measured by different experimental techniques. However, discrepancies exist between the reported lifetimes. One of the most fundamental examples is the lifetime of the ions of hexafluoride (SF_6), which is a widely studied compound, commonly employed for determination of the captured electron energy (E_{el}). In different experiments, $\tau_{\text{a}}(\text{SF}_6^-)$ was found to lie in a large range, extending from a few microseconds to a few milliseconds, and even to seconds. For instance, $\tau_{\text{a}}(\text{SF}_6^-)$ was found to be 10, 25, 68, 2, and 19 μs in [5–9], respectively, using time-of-flight (TOF) mass spectrometry. Ion cyclotron resonance (ICR) gave 500 μs [10] and values from 10 to 50 ms [11]. Cryogenic ion-beam trap measurements presented values from 10 to 100 ms [12], the electrostatic ion storage ring ELISA measured it to be between

100 μs and a few seconds [13], and a Penning ion trap, in combination with K/SF_6 , gave values of 1–10 ms [14,15]. Therefore, a clear spread in the reported values of $\tau_{\text{a}}(\text{SF}_6^-)$ exists in all experiments, but the most pronounced difference occurs between the data obtained from the TOF method and those measured with ion trap techniques. This disagreement has received significant consideration, which has led to a definite explanation. This explanation constitutes a statement that the SF_6^- ions are formed in the ionization chamber (IC) with different internal energies, i.e. with different vibrational populations. As a result, the ions are formed in a number of autoionizing states, characterized by different ion lifetimes, and τ_{a} is measured to be higher or lower depending on the time interval (Δt , 'time window') elapsed between the generation of the ions in the IC and their arrival at the detection system. As a result, the spread of measured τ_{a} values arises because of unequal Δt values in the different techniques. To date, this explanation has been universally accepted and is accompanied by detailed consideration of the reasons for the diversity in the ion internal energies. It was adduced that this diversity is caused by variations in E_{el} and the thermal vibrational energy of the parent molecule before the electron capture, with a subsequent influence on the ion state in some cases of residual gas collisions and radiative stabilization [10–13,15]. The competition between the direct autodetachment provided by the active vibrational modes and the delayed autodetachment caused by the internal vibrational relaxation (IVR) in the ions was also noted [14]. Alternatively, some doubts have been cast upon the involvement of E_{el} , because it was shown, for instance, in [14] that

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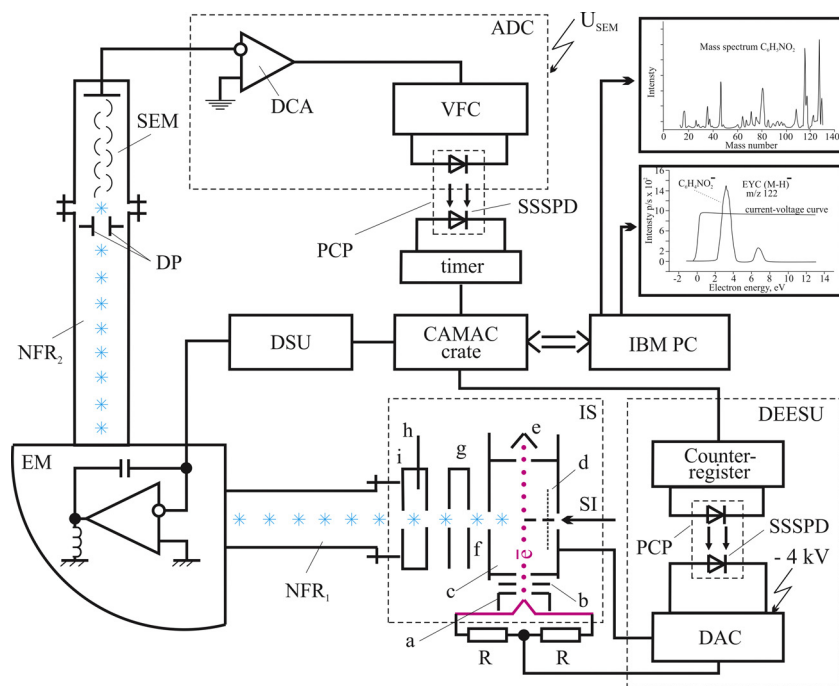


Fig. 1. Scheme of MI-1201B static magnetic sector mass spectrometer modified for recording negative ions in regime of resonant electron capture. CAMAC: computer application for measurement and control; DAC: digital-to-analog converter; DCA: direct current amplifier; DEESU: discrete electron energy scanning unit; DP: deflecting plates; DSU: digital scanning unit; EM: electromagnet; IS: ion source; NFR: nonfield region; PCP: photon-coupled pair; SEM: secondary electron multiplier; SI: sample inlet; SSSPD: silicon small-sized photodiode; VFC: frequency-to-voltage converter. (a, i) Lens, see t_{ext} .

$\tau_a(\text{SF}_6^-)$ was $\sim 19 \mu\text{s}$ and was independent of E_{el} . Thus, one can conclude that the temperature population of the parent molecule is, obviously, the main source of the ion internal energy variations [15–17]. As a result, if experiments are carried out at the same temperature, the spread in τ_a should mainly arise from differences in Δt , and the spread of τ_a should be insignificant if the Δt values are similar. This latter point is, obviously, the situation for the TOF technique, where the scattering of both Δt and $\tau_a(\text{SF}_6^-)$ are minor [5–9]. Simultaneously, there is a problem which arises when $\tau_a(\text{aM}^-)$ is measured with another beam instrument, namely the MI-1201B static magnetic sector mass spectrometer (SMS), modified to record the negative ions in the regime of the resonant electron capture. This instrument is used in the present paper. It has a vague similarity to TOF and SMS with respect to Δt ; therefore, it gives $\tau_a(\text{aM}^-)$ values on a similar level to TOF. However, the problem exists with SMS, because in a number of cases, we obtained a rather perceptible scatter in the measurements of $\tau_a(\text{aM}^-)$, when the SMS experiments were carried out at the same Δt and at the same temperature. It is thought that the MI-120B static magnetic sector mass spectrometer has constructional features that create one more experimental effect, leading to additional scattering of $\tau_a(\text{aM}^-)$. It is likely that the problem is related with the ion source of SMS, or, more exactly, with the IC placed in the ion source and the problem can be determined from measurement of the extraction time (t_{ext}) of the ions from the IC. The value of t_{ext} from the IC is the most unknown parameter of SMS, as it has never been identified experimentally in combination with τ_a . Therefore, t_{ext} was measured in this paper with the example of SF_6 , simultaneously with τ_a at various experimental conditions. In doing so, searches were carried out to find the dependence of t_{ext} on these conditions and a relationship between t_{ext} and τ_a , as well as for the unknown instrumental feature associated with the scattering of τ_a in the case of SMS. The methods of τ_a and t_{ext} , measurements have been described elsewhere [2, 18]. In addition, these methods are presented below in the experimental section of the present paper, along with a description of the SMS.

2. Experimental

2.1. Negative ion mass spectrometer (SMS) in the regime of the resonance electron capture

The scheme of the MI-1201B static magnetic sector mass spectrometer (JSC Selmi, Ukraine) used herein, modified to record negative ions in the regime of the resonant electron capture, is shown in Fig. 1. The instrument in general was described earlier [2]. In this paper, the ion source, where monoenergetic electrons and negative ions are formed, is depicted in Fig. 1 by the dashed rectangle marked as 'IS'. The electrons are emitted from the heated tungsten cathode in the cathode chamber 'a'. Then the electrons come through the screening diaphragm-lens 'b', which has a minor positive potential, into the IC, 'c'. The electrons obtain the desired energy (0–15 eV) by varying the potential difference between the cathode middle point and the IC box. These variations are carried out with a computer-controlled digital-to-analog converter (DAC) which is a part of a discrete electron energy scanning unit (DEESU). The system gives the distribution of 0.3–0.4 eV at a current of $1 \mu\text{A}$ (defined by the half-height of the effective yield curve – EYC – of SF_6^-). Electrons passing through the chamber are collected on the electron collector 'e'. The negative ions are generated in the IC ('c'), repelled from it by the electrode 'd', extracted from the chamber 'c' by the positive potential of the lens 'g', focused by the lens 'f', pass through the corrected electrode 'h' and are accelerated to the magnet-analyzer by the lens 'i' which is the exit slit of the ion source. The ion acceleration occurs on the path to the lens 'h' by a high negative potential ($U_{\text{acc}} = -4 \text{ kV}$ in the present study) applied on the ion source as a whole, except the lens 'h', which is kept under the ground potential. Further, the ions enter into the first nonfilled region (NFR₁) of the analyzer tube and go through the mass analyzing electromagnet (EM). The magnet field of the EM is varied using IBM PC through a digital scanning unit (DSU). Then, the ions go into the second nonfilled region (NFR₂) and are recorded by the secondary electron multiplier (SEM), the last

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