Formation and disappearance rates of metastable muon- α levels in high-pressure helium targets

L. Bracci

Università di Pisa, Italy

E. Zavattini

*European Organization for Nuclear Research, Geneva, Switzerland
and Università di Trieste, Italy*

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By considering the formation of excited muonic molecular ions $[(\mu^- \text{He})_{2S}^+ \text{He}]^{*+}$ in the triplet state $a^3\Sigma^+$, it is possible to explain the presence of metastable muonic helium levels at late times, observed when negative muons are stopped in a high-pressure ($p \geq 6$ atm) helium-gas target at room temperature. A comparison of the different experimental results with the present idea is reported, and measurements to obtain independent checks of this scheme are proposed.

I. INTRODUCTION

When a negative muon slows down in a helium target, an excited muonic helium ion $(\mu^- {}^4\text{He})_n^+ = M_n^+$ is quickly formed (n is the principal muonic quantum number). It is worthwhile to remember¹ that, already for $n=10$, such a $Z=1$ system has dimensions r_{10} smaller than the hydrogen Bohr radius a_e ($r_{10} = a_e/4$). Therefore from a “chemical” point of view, soon after its formation the system M^+ will behave like an ionized hydrogen atom H^+ and will therefore undergo many of the processes that are common to H^+ ions formed in helium. For instance, through three, or more, multibody collision processes it is quite possible to obtain the formation of a muonic helium molecular ion (which for simplicity we sometimes call—incorrectly—a cluster):

(1)

The possibility of the formation of a bound helium molecular ion, containing a muonic M_n^+ system, was first suggested some time ago² as a possible explanation for the lack of $K\gamma$ x rays for the case of negative muons stopped in a liquid-helium target.³ With this complex molecular system, it was possible to emphasize the relative importance⁴ of the internal Auger deexcitations on the molecular ion’s electron, over that of radiative transitions, for muons down to $n=4$.

Cohen⁵ was the first to show that the formation of the muonic helium molecular ions in the $X^1\Sigma^+$ stable (1S) bound state through process (1) is already dominant for relatively low-pressure helium targets. In fact, he has shown that for $p > 0.1$ atm, all negative muons stopped in a helium target will decay, being bound in a molecular system. Moreover, at a sufficiently high pressure during the muon lifetime, the formation of higher-order systems such as $(M^+ \text{He}_r)^+$ ($r=1,2,\dots$) from the ground-state molecular ion $X^1\Sigma^+$ becomes dominant. Starting from

these premises, the quenching rate $q_{2S}(p)$ of the metastable (2S) muonic levels, formed by stopping negative muons in a helium-gas target, can be calculated^{5,6} as a function of the helium-gas target pressure. One of the predictions of this scheme is that for $p > 1$ atm, the 2S level lifetime turns out to be quite short (between $\tau_{2S} = 10$ and 30 ns—see Fig. 1 taken from Ref. 6) owing to the Auger transitions and Stark-mixing radiative deexcitations within the molecule itself.

On the other hand, the presence of metastable muonic helium has been seen in experiments done by stopping negative muons in high-pressure helium targets ($p \geq 7$ atm) for times comparable to the muon lifetime⁷⁻¹⁰ (see, for example, Fig. 2 taken from Ref. 8).

FIG. 1. Dependence of the quenching rate for the M_{2S}^+ muonic system on helium pressure p . The various curves correspond to the different predictions obtained using different parameters (see Ref. 6) in the calculation.

FIG. 2. The $2S_{1/2}/2P_{3/2}$ resonance signal (Ref. 8). In the abscissa, the stepping motor position (SMP); in the ordinate, the number of counts per SMP normalized to the same number of stopped negative muons. Laser pulse delay from the muon stopping time: 500 ns.

In this paper we wish to take into account also the formation of some of the low-lying excited-level bound states¹¹ of the muonic helium molecular ion during the multibody collision process (1). We will discuss the relevance of the formation of some of these excited systems to the experimental results obtained by stopping negative muons in a high-pressure helium-gas target. All these states have a formation energy threshold larger than 10 eV.

We will see that the presence of these excited molecular ion bound states (in particular, the triplet ones) explains the already mentioned presence of the $2S$ systems at high pressure. Moreover, the formation of these excited (triplet) molecules can also explain some of the results¹² on the residual polarization P_μ observed by stopping polarized negative muons in a high-pressure helium-gas target.

If we call t_{cas} the cascade time (i.e., $1/t_{\text{cas}} \geq 10^9 \text{ s}^{-1}$) then, by definition, after t_{cas} the stopped negative muons will all be bound in a system M_i^+ with $i=1S$ or $2S$. Taking into account that the formation rate of process (1) is proportional to the square of the target pressure, then for pressures in the right range of values, the formation rate of the $X^1\Sigma^+$ molecular ion will be comparable to the free-muon decay rate $\lambda_0 = 4.5 \times 10^5 \text{ s}^{-1}$ (let us call this sit-

uation "the regime of cluster formation following the M_n^+ deexcitation") and much smaller than $1/t_{\text{cas}}$. Naturally, no excited molecular ion states can be formed in this regime because the thermal energy is too small.

It is clear, however, that if the pressure becomes sufficiently high, say $p > p_{\text{crit}}$ (where p_{crit} is some critical value), the molecular formation process (1) will begin to take place during the time interval t_{cas} (let us call this high-pressure situation "the regime of cluster formation preceding the complete M_n^+ deexcitation"). Then the muonic system M_n^+ can still be in an excited state with $n \gg 2S$, and there is therefore a certain amount of energy that can be released by this system to the colliding group of particles in process (1). In this case, new channel reactions are possible. In particular, together with process (1), we can also have

where the $*$ refers to the excited states, and $m < n$ so as to take care of the energy threshold for the formation of excited clusters.

Following Refs. 5 and 6, in order to make some quantitative estimates of processes (1) and (2), we will refer to the experimental results of de Vries and Oskam¹³ for the processes

and apply these results to the case where H^+ is replaced by the muonic ion M^+ . We will see that for $p \geq p_{\text{crit}} \approx 2$ atm, the multibody processes (1) and (2) will most probably proceed at a rate much faster than $1/t_{\text{cas}}$.

Before discussing process (2) and the properties of the $(M_m^+ \text{ He})^{*+}$ systems, let us recall some experimental results.

II. GENERAL INFORMATION

In 1970, when searching for delayed x rays in the range 2–14 keV, Placci *et al.*² observed 28 ± 6 events after stopping negative muons in a 7-atm helium-gas target. They attributed these delayed x rays (nonmonochromatic but with energies in the range 0–8.2 keV) to the following second-order electromagnetic decay process due to metastable ionic systems $(\mu^- \text{ He})_{2S}^+$ formed within the helium-gas target:

The total yield ϵ_{2S}^1 , as deduced from these delayed events (counted from a time $t=580$ ns after the muon stopping time t_μ^{stop}), was found to be $\epsilon_{2S}^1 = (3.4 \pm 0.7)\%$ per stopped muon, and the time distribution of the x rays for the observed M_{2S}^+ systems showed a total disappearance rate [see note (a) of Table I] of $\lambda_{2S}^1 = 1/\tau_{2S}^1 = (0.75 \pm 0.2) \times 10^6 \text{ s}^{-1}$, i.e., near to the calculated² vacuum total disappearance rate, $\lambda_{2S}^0 = \lambda_0 + \lambda_{\gamma\gamma} = 5.56 \times 10^5 \text{ s}^{-1}$, where $\lambda_{\gamma\gamma}$ is the decay rate for the second-order process (3).

Radiative transitions as well as Auger internal transi-

tions and Stark-mixing deexcitations, together with external Auger transitions due to collisions with the surrounding helium atoms, were included in the first cascade calculations² of the μ^- reaching the $1S$ or $2S$ levels. The results showed² that indeed the M_{2S}^+ systems were initially formed with a yield per muon ranging from 3% to 8%, the higher value valid for the case in which Stark-mixing transitions are sufficiently important during the cascade.

Another very important direct and independent experimental result arrived at in Ref. 2 is that the quenching rate λ_{St} , due only to Stark-mixing deexcitation [through $2S \rightarrow 2P \rightarrow 1S + X$ (8.2 keV)], was found to be less than 10^4 s^{-1} with 90% CL, i.e., much smaller than the free muon decay rate λ_0 . It is interesting to note that this result is obtained by observing that among the delayed events (in this case, $t = 580 \text{ ns}$ after t_{μ}^{stop}), no 8.2-keV monochromatic x rays were within the statistics. Strictly speaking, this last experimental result gives two possibilities: (i) either all $2S$ states have short lifetimes with $\tau_{2S} \ll t$; or (ii) part or all of them may have a lifetime τ_{2S} comparable to λ_0 ; in this case, however, we have $\lambda_{St} \ll \lambda_0$. The observation of nonmonochromatic delayed x rays from process (3) led Placci *et al.*² to exclude alternative (i).

These results on ϵ_{2S} , λ_{St} , and τ_{2S} were at first^{14,15} rather difficult to explain theoretically; early elementary calculations on Stark-mixing collisions [with subsequent deexcitation to the $1S$ level of the muonic ion $(\mu^{-4}\text{He})^+$] against isolated helium atoms of the target, already gave a value for λ_{St} much higher than λ_0 .

In 1972–1975, Bertin *et al.* and Carboni *et al.* set up an experiment to induce $2S \rightarrow 2P$ transitions in the M_{2S}^+ with an infrared laser. Using a helium target with $30 < p < 40 \text{ atm}$, and a double-resonance method, measurements of the $2P_{3/2} \rightarrow 2S_{1/2}$ and the $2P_{1/2} \rightarrow 2S_{1/2}$ energy-level differences were performed to an accuracy of 1.5×10^{-3} . The pulsed infrared laser was fired into the target, in one case 900 ns after t_{μ}^{stop} (Ref. 7) and in the others^{8,9} 500 ns after t_{μ}^{stop} . Using the same target apparatus, a separate experiment was performed looking at x rays from the decay process (3). At these pressures, ϵ_{2S}^1 was found¹⁰ to be between 3% and 4%, and the total $2S$ level lifetime τ_{2S}^1 had values similar to the value found at 7 atm.

In all the CERN experiments, ϵ_{2S}^1 and τ_{2S}^1 were directly and independently deduced from the observed number of delayed events.

Going back briefly to the cascade computations, it is worthwhile to mention that, in 1975, Bertin *et al.*¹⁰ published an approximate rule relating the $I(K\alpha)/I(K_{\text{tot}})$ ratio [of “prompt” α x rays to all x rays (per stopped muon)] to the number $\epsilon_{2S}^{\text{tot}}$ of metastable levels formed—approximate because it assumes certain values for some of the radiative transition amplitudes and because it neglects, for instance, deexcitations through Auger processes. This relation can be written

$$\epsilon_{2S}^{\text{tot}} = [1 - I(K\alpha)/I(K_{\text{tot}})]/F, \quad (4)$$

where the quantity F has a value of 7.2 ± 0.4 ; we will come back to this relation later. Figure 3 shows the lat-

FIG. 3. Recent published results on the $I(K\alpha)/I(K_{\text{tot}})$ and $I(K\beta)/I(K_{\text{tot}})$ ratios (from Refs. 16–19).

est experimental results published on the $I(K\alpha)/I(K_{\text{tot}})$ ratio.^{16–19}

In 1981, Cohen⁵ gave a firm quantitative basis to the muonic helium molecular ion formation picture, by considering specifically the cluster $(M_{2S}^+\text{He})^+$ of process (1) formed in the $X^1\Sigma^+$ stable state and computing its formation rate. With these results he was able to give precise quantitative predictions for the quenching rate $q_{2S}(p)$, as a function of pressure, of the metastable M_{2S}^+ system formed in a helium target. With such a scheme he has shown the following.

(i) For pressures over 0.1 atm, process (1) would become dominant during the muon lifetime.

(ii) The disappearance rate due to Stark-mixing deexcitation for the metastable M_{2S}^+ system bound in the muonic helium molecular ion $X^1\Sigma^+$ is $\lambda_{St} = 6 \times 10^7 \text{ s}^{-1}$ (in the lowest vibrational state); consequently, under these conditions ($p > 0.1 \text{ atm}$), the lifetime of the $2S$ level will be extremely small.

(iii) At pressures higher than 1 atm, it is possible to have other types of higher-order stable ionic clusters, such as $(M_{2S}^+\text{He}_n)^+$ (with $n = 1, 2, 3$), where the calculated quenching rate for the $2S$ levels is also very high.

(iv) This scheme is in disagreement with the high-pressure ($p \geq 7 \text{ atm}$) experimental results; it disagrees because of the much longer $2S$ level lifetimes experimentally observed, and because of the small value seen for λ_{St} at 7 atm.

At the Swiss Institute for Nuclear Research (SIN) in 1984,¹⁶ the predictions of Cohen⁵ for the behavior of the quenching rate $q_{2S}(p)$ were quantitatively confirmed up to pressures of 600 Torr. In particular, for reaction (1) leading to the $X^1\Sigma^+$ ground state, there was found for $p \lesssim 600 \text{ Torr}$ a formation rate

$$\lambda = (4.26 \pm 0.58) \times 10^7 p^2 \text{ atm}^{-2} \text{ s}^{-1}, \quad (5)$$

in agreement with the predictions of Ref. 5, which were obtained using the experimental results from Ref. 13.

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TABLE I. Experimental results on muonic helium 2S levels for $p \geq 6$ atm.

p (atm)	Ref.	ϵ_{2S} (%)	τ_{2S} (μ S)	λ_{St} (S^{-1})
6	16,18	6	$< 25 \times 10^{-3}$	
6	18	1.5 ± 0.8	0.325 ± 0.15	
7	2	3.4 ± 0.7	$(1.5 \pm 0.4)^a$	$< 1.4 \times 10^4$
30–40	7		≥ 0.9	
30–40	8		≥ 0.5	
30–40	10	3–5	$(1.4 \pm 0.15)^a$	
40	17	7	0.040	
50	20	4 ± 1	$(1.2 \pm 0.14)^a$	

^aIn Refs. 2, 10, and 20 τ_{2S} has been given omitting the systematic corrections due to the time spread of the x-ray detector system (most of the x-ray energy is around 4 keV): these new values are the corrected ones.

atm helium-gas target to search for delayed monochromatic 8.2-keV x rays, came to the conclusion¹⁶ that in these conditions for the 2S level, $\tau_{2S} < 25$ ns is valid. This conclusion has been arrived at from the “absence” of delayed monochromatic 8.2-keV x rays (an experimental result *per se*, which is obviously in complete agreement with the result published in 1971 by Placci *et al.*² at 7 atm), and with the essential additional *a priori* assumption that the Stark-mixing radiative deexcitation λ_{St} , for all metastable systems, was much bigger than the free-muon disappearance rate λ_0 (in agreement with the above-mentioned theoretical scheme of Cohen in 1982,⁵ but in contradiction with the 1971 CERN experimental results at 7 atm). Moreover, in deducing the limit for τ_{2S} , the value of ϵ_{2S} has been fixed to that given by expression (4) (to about 6%).

Finally, in 1987 Eckause *et al.*,¹⁷ stopping negative muons in a helium target at 40 atm and searching for the nonmonochromatic x rays due to process (3), reported a superior limit for the lifetime of the 2S level: $\tau_{2S} < 40$ ns. Also this value has been obtained, not in an independent way, but by fixing, for the ϵ_{2S} population, that particular value of ϵ_{2S}^{tot} obtained using expression (4).

More recently, Menshikov *et al.*⁶ considered in more detail the formation of the various stable clusters $(M^+He_n)^+$ (with $n=1, \dots, 6$) and the various quench-

ing channels for the 2S level. For $q_{2S}(p)$, they obtained as a function of pressure, for $p \geq 1$ atm, the behavior shown in Fig. 1. According to these authors, below about 10 atm the quenching proceeds mainly at the vibrational excited levels of the $(M^+He)^+$ ion (through Stark-mixing radiative deexcitations), whereas for $p > 10$ atm it proceeds, at the ground vibrational state of the ion, through (radiationless internal) Auger deexcitations. One of the consequences is, for instance, that at 6 or 7 atm the 2S-level lifetime should be around 10 ns, and mainly through radiative deexcitation (with the emission of monochromatic 8.2-keV x rays).

The results (or limits) described above are presented in Table I together with other results; from the experiments done to measure the $2S \rightarrow 2P$ transition energies, we have deduced a value (or limit) for the 2S lifetime by assuming that $\tau_{2S} \simeq D_L$, where D_L is the delay with which the infrared laser pulse was injected into the helium-gas target where the muons were stopped.

III. EXCITED CLUSTERS

A. Characteristics

The singly charged helium-hydride molecular ion $(H^+He)^+$ has been known since a long time, and many quantum-mechanical calculations for the $X^1\Sigma^+$ ground state of this system have been performed (Refs. 5, 11, 14, 15, 20, and 21).

In an analysis of the low-lying excited states of the $(H^+He)^{*+}$ system, Michels¹¹ found quite a number of excited triplet and singlet bound states of the type Σ^+ and Π^+ . For these excited bound states, Michels calculated the different vibrational-rotational constants and the dissociation energies. Using his values, we have calculated the quantities reported in Table II, referring to the equivalent $(M^+He)^{*+}$ muonic molecular excited bound states where the H^+ has been replaced by an M^+ muonic ion.

Looking at Table II, we can see the following two important facts.

(1) The value for the normal vibration frequency ω_e for the excited states is, in general, much smaller than the corresponding value for the ground state $X^1\Sigma^+$. This means that Stark-mixing transition rates $[2S \rightarrow 2P$

TABLE II. Vibrational-rotational constants; dissociation energies and λ_{St} for low-lying excited states of the $(M^+He)^+$ muonic helium molecular ion (using the results of Ref. 11).

State	D_e^0 (eV)	r_e (bohr)	ω_e (cm^{-1})	$\lambda_{St}(0)$ (S^{-1})	Computed energies $R = \infty$ (hartrees)	Computed vibrational states
$X^1\Sigma^+$	1.850	1.444	2128		-2.8757	16
$A^1\Sigma^+$	0.0353	5.68	132	1.4×10^4	-2.5000	5
$B^1\Sigma^+$	1.051	8.486	240	7.4×10^4	-2.1428	28
$C^1\Pi^+$	0.2228	7.905	138	1.5×10^4	-2.1250	12
$a^3\Sigma^+$	0.0814	4.472	234	6.8×10^4	-2.5000	7
$b^3\Sigma^+$	0.7324	7.754	249	8.5×10^4	-2.1742	20
$c^3\Pi^+$	0.1988	7.720	141	1.6×10^4	-2.1307	12
$X^1\Sigma^+{}^a$	2.00	1.470	2030	5.8×10^7		16

^aValues computed by Cohen (Ref. 5). Results of Milleur *et al.* (Ref. 21) give $\omega_e = 2051$ cm^{-1} .

$\rightarrow 1S + X$ (8.2 keV)] in the excited molecules $(M_{2S}^+ \text{He})^{*+}$ are expected to be reduced. In fact, using the same method as that applied in Refs. 5 and 6, we found the corresponding values for λ_{St} , also reported in Table II. These values have been arrived at by using, for the ground states, the following expression,^{5,6} which was obtained by applying the harmonic-oscillator approximation (with the vibrational number $\nu=0$):

$$\lambda_{St} = \lambda_{2P} (3a_\mu / 2Z\Delta)^2 \mu \omega_e^3 (1/2), \quad (6)$$

where μ is the reduced mass, $\lambda_{2P} = 2 \times 10^{12} \text{ s}^{-1}$, $a_\mu = 2.5 \times 10^{-11} \text{ cm}$, $Z=2$, and $\Delta=0.027$, where 2Δ is the $2S$ - $2P$ energy splitting in atomic units.

(2) The internal Auger deexcitation rates for the $2S$ muonic level now become negligibly small,²² given the large radii for the excited clusters (see r_e given in Table II). This means that in these excited systems the main contribution to Auger deexcitations (if any) comes, at these pressures, from the electrons of the neighboring colliding free-helium atoms: the rate for such a process has been calculated^{6,23} and is given by

$$\lambda_A^{\text{ext}}(p) = 3.6 \times 10^3 p \text{ s}^{-1}, \quad (7)$$

where p is the pressure in atmospheres.

In fact, what we have just said for $n=2$ is valid also for many levels with $n>2$; i.e., already at pressures higher than a few atmospheres in the case of the excited clusters the external Auger processes often dominate over the internal ones. Therefore, for M_{2S}^+ systems present within excited clusters, the quenching of the $2S$ level is much reduced.

An important question at this point is, what are the total disappearance rates for these excited molecular ions? Let us discuss separately the singlet and triplet systems.

1. Singlets

Figure 4(a) shows the different channels into which the singlet molecular ions can spontaneously decay.¹¹ We have computed the decay rates λ_i within the dipole approximation, according to the formula

FIG. 4. Decay scheme of the various excited muonic helium molecular ions (from Ref. 11): (a) for the singlet states, (b) for the triplet states.

$$\lambda_i = \frac{(4/3)}{\hbar^3} (\alpha/c^2) (\Delta E_{fi})^3 d_{fi}^2 S^{-1}, \quad (8)$$

where ΔE_{fi} is the energy difference between the initial and final states, and d_{fi}^2 is the square of the transition matrix element,

$$d_{fi}^2 = 2 \left| \int \psi_f \mu_x(R) \psi_i dR \right|^2 + \left| \int \psi_f \mu_z(R) \psi_i dR \right|^2, \quad (9)$$

with $\mu(R)$ being the dipole matrix element for a given internuclear separation R . The z axis is taken along the symmetry axis joining the two nuclei.

The values for ΔE_i are derived from Ref. 11 and given in Table III. The values for d_{fi} are calculated, starting from an interpolation of the dipole matrix element $\mu(R)$, which is also reported in Ref. 11 for the relevant transitions. The internuclear wave functions $\psi(R)$ are assumed to be harmonic-oscillator wave functions centered at the equilibrium distance R_{eq} , which is given in Ref. 11 for several states of the $(\text{H}^+ \text{He})^+$ ion. Therefore, for the $(M^+ \text{He})^+$ system we have

$$\psi = N H_n(R - R_{eq}) \exp[-\omega_e(R - R_{eq})^2 m / (2\hbar)], \quad (10)$$

where, after proper scaling, the frequency ω_e is derived from the corresponding values for the $(\text{H}^+ \text{He})^+$ ion in order to take into account the different value of the reduced mass m ; N is a normalization constant, and H_n are Hermite polynomials.

The lifetime for the $i \rightarrow f$ transition is then given by

$$\tau_i = 0.94 \times 10^{-6} (\Delta E_i)^{-3} (1/d_i^2) = 1/\lambda_i, \quad (11)$$

where ΔE_i is expressed in electron volts and d_i in Bohr units.

We remark that the transition rates are very sensitive to the initial and final values n_i and n_f of the vibrational quantum number. The highest λ_i value is obtained where the overlapping of the initial and the final wave function is greatest. For the transitions we consider, this occurs at (or near) the particular values reported in Table III together with the corresponding lifetimes. Therefore the τ_i values in Table III are to be regarded as lower values.

To obtain the total disappearance rates, we have also to take into account the rates $\lambda_{i,\text{coll}}(p)$ for deexcitation of the excited singlets to the lowest singlet $X^1\Sigma^+$ state, through collisions with the neighboring free-helium atoms of the target. From table III it can be deduced that it is quite possible that all excited singlet states of Table II will be converted into the ground singlet molecular ion $X^1\Sigma^+$ within 60 ns [or even more rapidly, depending on the contribution of $\lambda_{i,\text{coll}}(p)$ at the working pressure p]. Of course, from this moment the scheme proposed in Refs. 5 and 6 has to be applied. This means that if the pressure p is not too high (but still within $p \gtrsim p_{\text{crit}}$), the group of excited singlet molecules (with a muonic $2S$ -level system trapped in them) will give rise to an "early" 8.2-keV x-ray emission, which might be distinguishable from the "prompt" x rays if $\lambda_{i,\text{coll}}(p)$ is sufficiently small. On the other hand, when p is very high ($\gtrsim 10$ atm), then, since the quenching of the $2S$ level⁵ will in this case be mainly through Auger deexcitation, there

will not be an “early” additional 8.2-keV emission. Of course, no delayed signals, at times comparable to $1/\lambda_0$, are expected from this group.

2. Triplets

Figure 4(b) represents a scheme showing the various possible radiative decays of the triplet systems.¹¹ Following the procedure explained above, we have calculated some indicative rates for the processes of Fig. 4(b): the results of these computations are also given in Table III. We see that within at least 10 ns all triplets will be in the $a^3\Sigma^+$ state; this lowest triplet state is stable.

On the other hand, it is in principle also possible for the triplet states to be converted into the ground singlet molecular ion through collision with the neighboring helium atoms, for instance, according to

However, in the following we will neglect process (12),

since simple estimates show its rate to be relatively negligible (see the Appendix).

At this point we wish to add that it is most probable that, at high pressure, higher complex molecular ion formation will take place:

etc. (where T stands for triplet), which is similar to what happens^{5,6,21} for the ground singlet $X^1\Sigma^+$ molecular ions; this important point needs further theoretical investigation. Assuming that no processes such as (13) would take place, we have made a very rough estimation of the breakup rate Λ of the excited triplet molecule due to collisions with the helium atoms of the target; we found $\Lambda \lesssim \lambda_A^{\text{ext}}(p)$ [see expression (7)].

Under these conditions, the disappearance rate for a $2S$ muonic level present inside an $a^3\Sigma^+$ triplet can therefore be written

$$\lambda_{2S}^{\text{long}} = \lambda_{2S}^0 + \lambda_A^{\text{ext}} + \lambda_{\text{st}} = 1/\tau_{2S} , \quad (14)$$

TABLE III. Lifetimes and energy differences ΔE_i for the various transitions considered.

Transition	ΔE_i (Computed) (eV)	Vibrational quantum number		Lifetime	
		n_i	n_f	τ_i (ns)	λ_i
$A^1\Sigma^+ \rightarrow X^1\Sigma^+$	12.017	0	2	13	λ_1
		1	2	29	
		6	1	60	
$C^1\Pi^+ \rightarrow X^1\Sigma^+$	22.034	8	0	2.6	λ_2
		7	0	4.3	
		5	1	14	
		6	1	16	
$B^1\Sigma^+ \rightarrow X^1\Sigma^+$	8.435	6	21	50	λ_5
		6	22,23	50	
		8	16	50	
$C^1\Pi^+ \rightarrow A^1\Sigma^+$	10.017	0	0	6	λ_4
		0	1	7	
$B^1\Sigma^+ \rightarrow A^1\Sigma^+$	20.452	3	0	62	λ_3
		11	1	150	
		17	3	220	
		20	4	390	
$C^1\Pi^+ \rightarrow B^1\Sigma^+$	1.582	1	1	100	λ_6
		5	7	400	
$c^3\Pi^+ \rightarrow b^3\Sigma^+$	1.716	0	0	60	λ_7
		2	2	70	
		3	3	90	
		4	4	120	
$c^3\Pi^+ \rightarrow a^3\Sigma^+$	8.214	2	0	6	λ_9
		0	1	17	
		1	1	14	
		12	6	12	
		0	3	19	
$b^3\Sigma^+ \rightarrow a^3\Sigma^+$	9.930	0	6	3×10^{-6}	λ_8
		8	1	10^{-2}	
		1	6	2×10^{-2}	
		3	0	2×10^{-2}	
		4	0	10^{-1}	

where λ_A^{ext} is given by expression (7), and the Stark-mixing deexcitation depends on its vibrational number ν . On the other hand, Ref. 6 shows that in a time quite small compared with $1/\lambda_0$, owing to collisions the molecular systems will quickly deexcite to the $\nu=0$ level. The corresponding $\lambda_{\text{st}}(0)$ is given in Table II. If processes such as (13) do take place, then the value of $\lambda_{\text{st}}(0)$ in Table II for the triplet $a^3\Sigma^+$ is merely indicative, and most probably represents an upper limit.

Expression (14) shows that τ_{2S} is not far from $1/\lambda_0$, in the range of tens of atmospheres, indicating that at high pressure the presence of these triplet molecular ions explains the observation of the $2S$ muonic levels at late times (at 40 atm, one gets $\tau_{2S} \approx 1.1 \mu\text{s}$).

In Table II we have also reported the corresponding computations for the $X^1\Sigma^+$ system made by other authors. A comparison between the different computations seems to indicate that for λ_{st} there might be a systematic computational error of as much as approximately 20%.

So far, we have assumed that the target helium is pure. In the case where an impurity J is present within the helium target, then, given the excess of energy available in the excited clusters (of any type), charge-transfer processes from the impurity J , such as

can take place, and if $n=2S$, the metastable level will be quickly quenched by the Auger process (the neutral ground molecule is not bound).²⁰

Let us call τ_{2S}^{long} the lifetime of an M_{2S}^+ system trapped in an excited triplet molecular ion, and τ_{2S}^{short} its lifetime when trapped in any other system. What is clear is that τ_{2S}^{long} has a value close to $1/\lambda_{2S}^0$, whereas τ_{2S}^{short} is about two orders of magnitude shorter. Moreover, if we call $\epsilon_{2S}^{\text{short}}$ and $\epsilon_{2S}^{\text{long}}$ the fractions of M_{2S}^+ "initially" formed in the excited triplet states and in other systems, respectively, then we can write

$$\epsilon_{2S}^{\text{tot}} = \epsilon_{2S}^{\text{short}} + \epsilon_{2S}^{\text{long}}. \quad (16)$$

Eventually, a rough estimate of $\epsilon_{2S}^{\text{tot}}$ can be obtained using expression (4).

At this point let us consider an experiment in which negative muons are stopped in a helium-gas target in order to look for $2S$ muonic levels. If the pressure is sufficiently high, then τ_{2S}^{short} will be within the time resolution defining the "prompt" events. Under these conditions, it is obvious that only the $\epsilon_{2S}^{\text{long}}$ component can be observed and separated [mostly through the second-order decay process (3), since very few delayed monochromatic 8.2-keV x rays are emitted at late times owing to the smallness of $\lambda_{\text{st}}]$.

This means that in this case, analyzing the observed x-ray events, one cannot use the knowledge of $\epsilon_{2S}^{\text{tot}}$ [given, for instance, by the approximating expression (4)], but must use the complete fitting expression which contains also the two independent quantities, $\epsilon_{2S}^{\text{long}}$ and τ_{2S}^{long} . Of course the τ_{2S}^{long} nonmonochromatic x-ray component can easily be confused within the x-ray background from the μ^- decay-electron's bremsstrahlung, since both contributions have similar time distributions.

B. Formation of excited molecular ions

Extrapolating the low-pressure result (5), we see that for $p > 5$ atm the stable ground system $(M_{2S}^+ \text{He})^+$ would have a formation rate that could become higher than $1/t_{\text{cas}}$. However, to be more precise—and also to take into account the possibility that $m \geq 4$ in process (1)—we will use the analysis already done by de Vries and Oskam¹³ (for the case where M_{2S}^+ is replaced by H^+).

The multibody conversion (1) or (2) can be written as a two-body reaction between X and the (mY) complex:

where R represents reaction products.

Whenever m atoms Y are within a small volume V , the rate for process (17) can be written

$$\lambda_m = \sigma_m v n^m V^{(m-1)}, \quad (18)$$

where σ_m is the effective two-body reaction, v is the relative velocity between X and the (mY) complex, and n is the density of the Y atoms. This estimate indicates that the ratio λ_{m+1}/λ_m equals nV , independent of m , provided the parameters σ_m , v , and V are reasonably constant. From Ref. 13 we get experimentally $V = 2 \times 10^{-20} \text{ cm}^3$. Therefore, for $n \geq n_{\text{crit}} = 1/V$, all multibody collision rates for $m \geq 3$ will become quite important and of the same magnitude, so that at these densities it is very likely that the formation time for clusters can be shorter than the cascade time t_{cas} ; the given value of V implies that the pressure (room temperature) corresponding to n_{crit} is $p_{\text{crit}} \approx 1.7$ atm. In the following, we will take $p_{\text{crit}} = 2$ atm.

To summarize: for this range of pressures ($p \geq p_{\text{crit}}$) we can outline the following general picture. During the multibody collision, a ground $X^1\Sigma^+$ cluster may be formed with the muon at a very high n level; however, whether it is still in the interaction volume or out of it, the ground cluster will quickly undergo the Auger process ($n \gg 2$):

with M_m^+ restarting the cycle via process (1). Eventually, with these series of sequences (with or without Stark-mixing radiative deexcitations) and if no excited molecules are formed during the multibody collisions, an $X^1\Sigma^+$ system will be formed with the negative muon in the $1S$ or $2S$ level. From here onwards, the phenomenology described in Refs. 5 and 6 will apply. When, on the other hand, an excited cluster $(M_m^+ \text{He})^{*+}$ is formed—as we know, in this system the internal Auger transitions become less probable than the external ones—there is a very good chance that the $2S$ level will be formed since, already at high n , internal radiative deexcitations can be important with respect to the external Auger processes. In this case, the enclosed M_m^+ muonic systems can cascade radiatively down to the $2S$ level relatively often (and many to the $1S$ one), while remaining within the excited cluster.

As we have already seen, the time limit within which the singlet excited molecular ion component will be

transformed into the ground molecular state is short; therefore, at the end of this time, all muons will either be in an $a^3\Sigma^+$ molecular ion or in the ground singlet $X^1\Sigma^+$ system.

This picture has an upper limiting pressure p^M , which is given by the p^M for which the external Auger rate $\lambda_A^{ext}(p^M)$ becomes much higher than the μ^- decay rate. From expression (7) for $\lambda_A^{ext}(p)$, we find that for $p \gtrsim p^M = 200$ atm, the external Auger rate is more than that of λ_{2S}^0 ; and therefore from this pressure on we expect $\tau_{2S} \rightarrow 0$.

IV. COMPARISON OF EXPERIMENTAL RESULTS

From experiments done by stopping negative muons in a high-pressure helium target, we will try to estimate the fraction of muons trapped in an excited triplet cluster, and to analyze the consistency of the various experimental results when compared with the various predictions.

A. The CERN results

In Table I it can be seen that ϵ_{2S}^{long} varies around 3.5%: we will take as indicative the value $\epsilon_{2S}^{long} = (3.4 \pm 0.7)\%$ obtained at 7 atm (where also a direct measurement of λ_{St} was possible).

To get an estimate of ϵ_{2S}^{short} , we use expression (4) together with expression (16) and the results of Fig. 3 for $p > 6$ atm. In this way we get $\epsilon_{2S}^{short} \simeq 4\%$, which indicates that it is of the same order as ϵ_{2S}^{long} . If, as the cascade calculations tend to show, the $2S$ population reflects the $1S$ population proportionally, then from this value we can estimate the fraction f ($\approx 60\%$) of negative muons that are bound in the ground $X^1\Sigma^+$ molecule at late times.

With the present suggested scheme, there is fair agreement between the experimental values for the $2S$ lifetime given in Table I and the expected one given by expression (13). Moreover, comparing the value of λ_{St} obtained at 7 atm with the corresponding value for a muon in an $a^3\Sigma^+$ system (see Table II), we see that they are close, although the agreement is still poor; in this respect we should remember that probably, at these pressures, processes such as (14) might be frequent.

B. The SIN 6-atm experiment (Ref. 16)

For this discussion (where ϵ_{2S} and τ_{2S} must be deduced from the experimental data independently) we cannot refer only to the results of von Arb *et al.* given in Ref. 16, but have to use also those of Ref. 18.

Figures 5(a) and 5(b) show the spectra of the sum $E_\gamma = E_1 + E_2$ of the energies of two x rays in coincidence, for times contained in two contiguous delayed gates $D_1 = t_2 - t_1$ and $D_2 = t_3 - t_2$ (with $t_1 = 75$ ns, $t_2 = 425$ ns, and $t_3 = 1900$ ns measured from the incoming " π^- "). The peaks at 8.2 keV (presumably) represent the contribution of metastable states decaying according to process (3).

A quantitative evaluation of the "good" events present in Fig. 5, N_1 and N_2 , made by the author of Ref. 18, has given $\epsilon_{2S}^{long} = (1.5 \pm 0.8)\%$ and $\tau_{2S}^{long} = (0.325 \pm 0.150) \mu\text{s}$.

FIG. 5. Distribution of the energy sum $E_\gamma = E_1 + E_2$ (in keV) of two x rays coincident in time. These x rays are two contiguous delayed time gates $t_2 - t_1$ and $t_3 - t_2$, where (a) $t_1 = 75$ ns, $t_2 = 425$ ns; (b) $t_1 = 425$ ns, $t_3 = 1900$ ns measured from the incoming " π^- " (data taken from Ref. 18).

(ϵ_{2S}^{long} and τ_{2S}^{long}) depend, respectively, on the sum $N = N_1 + N_2$ and on the ratio N_1/N_2 , via $\ln(N_1/N_2)$.

Clearly the presence of metastable events in these graphs is in disagreement with the expected behavior required by Fig. 1, based on the formation of $X^1\Sigma^+$ ground clusters only. In fact, at these pressures the $2S$ disappearance rate is predicted to be 10 ns. These results show that already at 6 atm, a long-lifetime component is present among the metastable systems formed.

With regard to the SIN experiment at 40 atm,¹⁷ which has reported $\tau_{2S} < 40$ ns, we have already remarked that this value has been obtained by fixing for ϵ_{2S}^{long} the value ϵ_{2S}^{tot} (about 7%): we do not agree with this procedure.

C. Residual polarization P_μ of polarized negative muons stopped in a high-pressure helium-gas target

We think that the scheme proposed in this paper gives also a "natural" explanation of why, when polarized muons are stopped in a helium-gas target at pressures p defined by $p_{crit} < p < p^M$, the residual polarization P_μ is found to be between $\frac{1}{3}$ and $\frac{1}{2}$ of the expected theoretical value ($P_\mu^{th} = 15\%$).

Table IV shows the experimental results published by Souder *et al.*^{12,24} on P_μ in helium [A refers to the neutral muonic helium atom $(\mu^- \text{He})^+ e^-$ formed by contamination of the helium target with xenon; P^A is the residual polarization of the negative muon in this system].

From this table we can see that up to 50 atm, P_μ ,

TABLE IV. Total negative muon residual polarizations (from Ref. 12).

Pressure	P_μ (%)	P_A (%)	P_{tot}	[Xe] (%)
7	9.4 ± 2.3	0	9.4 ± 2.3	0
14	5.8 ± 1.3	0	5.8 ± 1.3	0
14	6.7 ± 1.6	0	6.7 ± 1.3	0.2
14	2.9 ± 1.8	5.0 ± 0.7	7.9 ± 1.9	2.0
50	3.5 ± 2.4	0	3.5 ± 2.4	0
liquid	1 ± 2	0	1 ± 2	0

which is expected to be around 15%,¹² is about two to three times smaller; moreover, it is interesting to note that by contaminating helium with xenon, it has been possible to form the muonic helium neutral atom $(\mu^- \text{He})^+ e^-$ (see P_A in Table IV).

In discussing these types of experiments, we will be interested only in systems (the majority) where the negative muons are bound in the 1S level.

First of all, it is important to stress that the electron transfer process from xenon to the $X^1\Sigma^+$ ground cluster, with the formation of a muonic helium neutral atom (see results at 14 atm),

is forbidden because of energy conservation (the ionization energies for xenon and for the muonic helium neutral atom are 12.1 and 13.6 eV, respectively, and the ground-state cluster binding energy is about 1.9 eV). A similar situation is valid for higher molecular ions such as $(M_{1S}^+ \text{He})^+$. Therefore, to explain some of the results in Table IV, we suggest that the observed residual muon polarization P_μ could be mostly due to the decay of muons bound in the excited triplet $a^3\Sigma^+$ systems.

We believe that muons bound in the ground $(M_{1S}^+ \text{He})^+$ systems have their residual polarization $P_{\mu 1}$ appreciably reduced. Simple classical calculations show that because of the rotational excitation in the ion $(M_{1S}^+ \text{He})^+$, the magnetic field at the muon site is about 100 times stronger than in the $a^3\Sigma^+$ molecular ion; moreover, for the former, in the case of maximum rotational excitation, we obtain about 100 G.

We think that the following process is, in fact, taking place:

which is energetically possible [in process (21), * means excited in an $a^3\Sigma^+$ system].

Using the experimental values for P_{tot} given in Table IV, at 14 atm with and without xenon contamination, it is found that $P_{\mu 1} \approx 3\%$ and f is around 70%, with f being the fraction of $X^1\Sigma^+$ ground-state systems present at late times. It is interesting to note that this value does not contradict a corresponding value for f found earlier when analyzing the 2S population present after the "prompt" time.

Of course, another way is to imagine having a charge transfer process forming a muonic helium neutral atom

directly²⁵ as, for instance,

which is also energetically allowed. However, for reaction (22) to happen, there must be a cross section of at least a few times 10^{-16} cm^2 at these thermal energies, since the M_{1S}^+ system at these pressures is present for a very short time (see Sec. III B).

Our suggestion, process (21), or—more generally—process (15), can be tested, as in these cases almost any other noble gas can participate in it since energetically this is always feasible. In other words, it is possible to test the presence of excited clusters at late times, by detecting process (15) [i.e., by looking at the decrease of the residual polarization, or for the presence of the neutral $(\mu^- \text{He})^+ e^-$ atoms, or both], having chosen an element J such that process (22) cannot happen (for instance, argon) owing to the conservation of energy.

V. CONCLUSIONS

In the preceding sections we have shown that when stopping negative muons in a gas target ($p > 0.1 \text{ atm}$) there are two possible regimes.

(i) One in which, after the formation of the muonic helium ion $(\mu \text{He})^+$ (stable or metastable), an $X^1\Sigma^+$ ground (singlet) muonic helium molecular ion will be formed; the phenomenology following this process is described in Refs. 5 and 6.

(ii) A second regime in which, when $p \geq p_{\text{crit}}$, multiple collisions become so frequent that a muonic helium molecular ion cannot be formed before the $(\mu \text{He})_n^+$ excited muonic ion has time to reach the stable (1S) or metastable (2S) levels.

In the latter case, given the energy available, it is then possible that also excited muonic helium molecular ions are formed (excitation energies higher than 10 eV), and we have shown that among these excited molecular ions the lowest triplet state can be present at a late time.

We have also shown that, using the experimental results of de Vries and Oskam¹³ on the formation rate of the $(\text{H}^+ \text{He})^+$ ionic complexes, p_{crit} is between 1 and 2 atm.

From the properties of the excited molecular ions, we have deduced that if a 2S muonic level is present within an $a^3\Sigma^+$ triplet muonic molecular ion, then the disappearance rate λ_{2S} is comparable to the μ^+ disappearance rate λ_0 .

Analyzing the available experiment data on the presence of the 2S muonic level (for times greater than 300 ns from the muon stopping time t_μ^{stop}), we have estimated that the lowest excited triplet muonic helium molecular ions (1S or 2S) are probably formed in 30–40% of the stopped muons in a high-pressure helium-gas target.

The direct experimental results for the Stark deexcitation rate of the 2S muonic level (with the consequent emission of monochromatic 8.2-keV x rays) measured with a helium target at 7 atm, give limits near to, but definitely smaller than, the predicted one (see Table II).

We believe that similarly to what happens for the $X^1\Sigma^+$ ground singlet molecular ions,^{5,6} the lowest triplet level due to multiple collisions also gives rise to higher-order triplet muonic helium molecular ions (triplet clusters) in which presumably the Stark deexcitation rate of the $2S$ level is further reduced.

The presence of these excited muonic helium molecular ions can be observed independently by detecting the particular charge transfer reaction (15); in fact, this process (for instance, from an argon atom) is in many cases forbidden both to a muonic $(\mu\text{He})_{1S}^+$ ion and to the $X^1\Sigma^+$ ground singlet molecular ion, owing to energy conservation. The presence of the neutral atom from reaction (15) can be detected with the experimental procedure developed by Souder *et al.*¹² Within the scheme presented here, it is possible to explain the amount of residual muon polarization P_μ that is observed when muons are stopped in a 14-atm helium-gas target.¹² As is well known, with simple arguments one expects $P_\mu = 15\%$; experimentally, a value smaller by a factor of between 2 and 3 is found. Our interpretation is that in the $X^1\Sigma^+$ ground singlet ion, the muon depolarization is quite effective owing to the magnetic field connected with the high rotational states of the molecule, whereas it is quite negligible in the $a^3\Sigma^+$ excited triplet muonic helium molecular ion. Observing the decrease of P_μ when the helium gas target is contaminated with argon (by a few percent) is also a way of detecting process (15), and hence of detecting the presence of excited triplet muonic helium molecular ions.

We will close this paper by pointing out the possible relevance of the muonic helium excited molecular ion formation in other types of experimental situations, for example: (i) the precise determination of negative muon-nuclear capture from helium nuclei measured by stopping muons in a pure helium gas target for $p \geq 1$ atm; (ii) the interpretation of the various observed reactions when negative protons $\bar{p}$ are stopped in a helium-gas target; in fact, also in this case and for $p \geq 1$ atm, because of fast multiple collisions there might be time (before the $\bar{p}$ is captured by the helium nucleus) to form antiprotonic helium molecular ions in the excited states (singlet or triplets) $[(\bar{p}\text{He})_r^+\text{He}]^{*+}$. Given the smallness of λ_{st} (see Table II), these excited molecules, once formed, can survive for "relatively" long times, and this can give rise to capture of $\bar{p}$ from (high) atomic levels (n, l) , i.e., quite different from those that are usually found when $\bar{p}$ are stopped in H_2 , and (few) anomalously delayed annihilation stars.

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APPENDIX

To estimate the transition rate from the $c^3\Pi^+$ triplet to the $X^1\Sigma^+$ singlet state [process (14)], because of col-

lisions with helium atoms we use the impact-parameter method. The transition probability P is given by

$$P = \frac{1}{\hbar^2} \left| \int_{-\infty}^{+\infty} V_{fi} e^{i\Delta E_{fi}(t/\hbar)} dt \right|^2, \quad (\text{A1})$$

where ΔE_{fi} is the energy difference between the initial and the final state. The cross section is then given by

$$\sigma = 2\pi \int_{\rho}^{\infty} P(b) b db; \quad (\text{A2})$$

P is calculated assuming a classical trajectory for the relative motion, which is approximated as a straight line tangent to the trajectory at the point of closest approach ρ .

The interaction $\hat{V}$, which is responsible for the transition, is assumed to be the dipole interaction of the atomic electrons of the $1S(1s)^2$ helium atoms with the electric field $\mathbf{E}$ generated by the $c^3\Pi^+$ ion. This latter field is approximated with the field of a pointlike charge, so that we have

$$\hat{V} = e \frac{\mathbf{n}_b + \mathbf{v}t}{|\mathbf{n}_b + \mathbf{v}t|^3} (\mathbf{r}_1 + \mathbf{r}_2), \quad \mathbf{n} \cdot \mathbf{v} = 0, \quad (\text{A3})$$

r_1 and r_2 being the coordinates of the atomic electrons of the helium. Since the total spin is conserved, the transition is accompanied by a transition of the helium atom to a triplet state. The initial wave function is a (antisymmetrized) product of the symmetric $1(1s)^2$ electron wave function of the helium atom times the antisymmetric wave function of the $c^3\Pi^+$ ion. Similarly, the final wave function is a (antisymmetrized) product of the symmetric wave function of the $X^1\Sigma^+$ ion times a triplet wave function for the helium-atom electrons.

For the helium wave function we take

$$\psi_{\text{sing}}^A = N_S^A \exp \left[-\frac{1}{a} (r_1 + r_2) \right], \quad a = \frac{16}{27} a_0, \quad (\text{A4})$$

$$\psi_{\text{trip}}^A = N_T^A \left[\exp \left[-\frac{1}{a_0^2} (r_1 + 2r_2) \right] - \exp \left[-\frac{1}{a_0^2} (2r_1 + r_2) \right] \right], \quad (\text{A5})$$

whereas for the ion wave function we take into account the results of Ref. 11 in order to have the correct exponential behavior. We have

$$\psi_{\text{sing}}^I = N_S^I \exp \left[-2.596 \frac{1}{R_1} (r_1 + r_2) \right], \quad (\text{A6})$$

$$\psi_{\text{trip}}^I = N_T^I \left[\exp \left[-16 \frac{r_1}{R_3} - 5.008 \frac{r_1}{R_3} \right] - \exp \left[-16 \frac{r_2}{R_3} - 5.008 \frac{r_1}{R_3} \right] \right], \quad (\text{A7})$$

where $R_1 = 1.444a_0$ and $R_3 = 7.72a_0$, as given in Ref. 11. The N_j^i 's are normalization constants. The exponential decrease of the wave function is accounted for by using, for the dipole matrix element, the value obtained for the

vanishing atomic ion separation times a factor $\exp(-\lambda R)$, with $R = |\vec{v}t + \vec{n}_b|$ and $\lambda = 2.7/a_0$, in accordance with Eqs. (A3)–(A7).

In conclusion, we calculate

$$P = |\langle f | \mathbf{d} | i \rangle|^2 \frac{1}{\hbar^2} \left| \int_{-\infty}^{+\infty} \left[\frac{1}{b^2 + v^2 t^2} \{ \exp[-\lambda(b^2 + v^2 t^2)^{1/2}] \} \cos \omega_{fi} \frac{t}{\hbar} \right] dt \right|^2. \quad (\text{A8})$$

For standard conditions of pressure and temperature, we get

$$\sigma = 10^{-23} \text{ cm}^2,$$

which yields a transition rate $\lambda = 50 \text{ s}^{-1}$ and a lifetime

$$\tau = 2 \times 10^{-2} \text{ s}^{-1}.$$

This lifetime is quite long compared with all other lifetimes.

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