Rapid Communications

Question of why metastable muonic helium is not quenched

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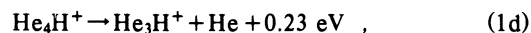*Theoretical Division, Los Alamos National Laboratory, University of California, Los Alamos, New Mexico 87545*
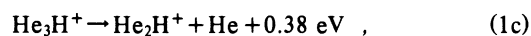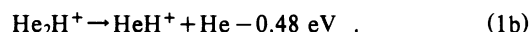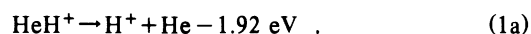(Received 13 July 1981)

The ion-clustering mechanism which is the explanation usually invoked for the long collisional lifetime of metastable muonic helium is examined. Based on current knowledge of similar clusters, this mechanism does not work.

During the last decade, metastable muonic helium has proved to be a cooperative species for experimental investigation.[1] Currently, related measurements on metastable muonic hydrogen are being attempted.[2] For Lamb-shift-type experiments the lifetime of the metastable muonic state is an important consideration. The dominant disappearance mechanisms for the isolated metastable muonic helium ion are muon decay (4.5×10^5 sec^{-1}) and two-photon emission (1.1×10^5 sec^{-1}). Additional quenching by external interactions may be expected via the Stark and the Auger effects. However, the experiments, performed with 7–50 atm of helium, have seen no decisive evidence of any pressure-dependent quenching. In particular, the earliest experiment (at 7 atm) established a quite low level of 2×10^3 sec^{-1} on the Stark quenching rate.[1(a)] If it is assumed that the metastable muonic helium species is $[\alpha\mu^-(2s)]^+$, designated He$_\mu{}^+(2s)$, this limit is in drastic disagreement with theoretical calculations.[3–5] The theoretical collisional Stark quenching rate[5] at 7 atm is 1.0×10^6 sec^{-1}. The explanation[4] which has come to be generally accepted is that helium atoms form a cluster with the muonic ion; the resulting electric field at the site of the ion is supposed to be very weak for symmetry reasons, thus obviating the Stark mixing effect. This argument has always been presented qualitatively and so it seems appropriate to investigate its merits in light of current knowledge about such clusters.

Because of the small size of the muonic orbital, the interaction of normal He atoms with He$_\mu{}^+$ is essentially the same as with H$^+$ except at very small internuclear distances.[5] Milleur *et al.*[6] have made a theoretical study of He$_n$H$^+$ complexes and found the

relative stabilities to be as follows:

$$\text{HeH}^+ \rightarrow \text{H}^+ + \text{He} - 1.92 \text{ eV} \quad , \tag{1a}$$

$$\text{He}_2\text{H}^+ \rightarrow \text{HeH}^+ + \text{He} - 0.48 \text{ eV} \quad , \tag{1b}$$

$$\text{He}_3\text{H}^+ \rightarrow \text{He}_2\text{H}^+ + \text{He} + 0.38 \text{ eV} \quad , \tag{1c}$$

$$\text{He}_4\text{H}^+ \rightarrow \text{He}_3\text{H}^+ + \text{He} + 0.23 \text{ eV} \quad , \tag{1d}$$

where He$_2$H$^+$ is linear, He$_3$H$^+$ is planar (D_{3h}), and He$_4$H$^+$ is tetrahedral. Hence the He$_2$H$^+$ complex is predicted to be most stable. This conclusion is not in disagreement with the observation by Atkins[7] of very large ion clusters in liquid helium since we are not concerned with the additional atoms which may be bound at larger distances by long-range forces. That is, the electric field at the site of the central atom (H$^+$ or He$_\mu{}^+$) is primarily affected by the nearby atoms.

We thus may expect the He$_\mu{}^+$ ion to combine with two He atoms if the ion exists long enough for the third-order kinetics to occur. For this purpose it is instructive to consider the analogous situation in the high-pressure helium afterglow; the masses are almost the same and the binding energies are similar.[8] The three-body rate constant[9] for formation of He$_2{}^+$ is about 6×10^{-32} cm^6/sec and subsequent formation of He$_3{}^+$ is even faster.[10] Hence at pressures greater than about 3 Torr the conversion of the He$_\mu{}^+$ ion to He$_2$He$_\mu{}^+$ may be rapid enough to inhibit *collisional* Stark quenching. At such pressures recent work suggests that molecular-ion formation may be augmented by four-body conversion reactions.[9]

Supposing now that the He$_\mu{}^+(2s)$ ion actually does reside in a complex He$_n$He$_\mu{}^+$, with $n = 1$ or 2, we

need not be further concerned with quenching collisions but we must estimate the rate of Stark transitions induced by internal molecular vibrations. Adiabatic treatment of the nuclear motion is justified since $2\Delta/(h\nu_{osc}) \geq 6$, where $\Delta(=0.027$ a.u.$)$ is *half* the average $2s$-$2p$ muonic atomic splitting and ν_{osc} is the classical vibrational frequency. The wave function Ψ for motion on the potential energy surface V is calculated quantum mechanically and the transition rate is then obtained by perturbation theory,

$$\gamma_{St} = \langle \Psi | A(\vec{R}) | \Psi \rangle \ . \tag{2}$$

The transition rate at a fixed nuclear configuration (designated $\vec{R}$) is[3]

$$A(\vec{R}) = \gamma_0 \left| \frac{\epsilon(\vec{R}) - \Delta}{2\epsilon(\vec{R})} \right| \ , \tag{3}$$

assuming $\gamma_0 \ll \epsilon(\vec{R})/\hbar$, where $\gamma_0(=2.0 \times 10^{12}$ sec$^{-1})$ is the $2p$ radiative transition rate and we have defined

$$\epsilon(\vec{R}) = [\Delta^2 + S^2(\vec{R})]^{1/2} \ . \tag{4}$$

Assuming that the Hellmann-Feynman theorem is applicable and that the molecular He_μ^+ retains a net charge of nearly $+1$, the first-order Stark energy is

$$S(\vec{R}) = \frac{3a_\mu}{Z} |\nabla V(\vec{R})| \ . \tag{5}$$

Here $a_\mu(=5.0 \times 10^{-3}a_0)$ is the muonic Bohr radius and $Z(=2)$ is the nuclear charge. The gradient of the potential is taken with respect to the Cartesian coordinates of the muonic atom. The evaluation of $A(R)$ by Eqs. (3)$-$(5) was explicitly checked in the case of $HeHe_\mu^+$ and found to yield results within about 2% of the *ab initio* results[5] except close to the

minimum of the potential curve.

If the vibrational energy is small, a normal-coordinate description, with each mode described by a harmonic-oscillator wave function, should be adequate. This approximation is also simplified by retaining only the first-order term in the expansion of A in powers of S^2. Before proceeding to $He_2He_\mu^+$ let us first compare the results for $HeHe_\mu^+$ obtained with (1) the *ab initio* potential,[5] (2) a Morse potential $(D_e = 2.00$ eV, $R_e = 1.47a_0$, and $\beta = 1.47a_0^{-1}$ fit to the *ab initio* potential), and (3) a harmonic-oscillator potential $(\omega = 2030$ cm$^{-1})$. In the harmonic-oscillator approximation we get the simple expression

$$\gamma_{St}^{HO}(1) = \hbar \left[\frac{\gamma_0}{4\Delta^2} \right] \left[\frac{3a_\mu}{Z} \right]^2 (2\pi c)^3 \left[\frac{mM}{m+M} \right] \omega^3(v + \tfrac{1}{2}) \ , \tag{6}$$

where M is the mass of He_μ^+, m is the mass of He, ω is the vibrational frequency (in wave number units), and v is the vibrational quantum number. Hence

$$\gamma_{St}^{HO}(HeHe_\mu^+) = 1.15 \times 10^8(v + 0.5) \ , \tag{6a}$$

expressed in units of sec^{-1}. It can be seen in Table I that the harmonic-oscillator approximation is good for the lowest few vibrational states and thus we can apply it with confidence to the larger cluster.

For a linear symmetric triatomic molecule, oriented in the direction of the x axis, the above assumptions give the electric field on the central atom,

$$\frac{1}{e} \nabla V(\vec{R}) = \frac{(2\pi c)^2}{e} \left[\frac{2mM}{2m+M} \right]^{1/2}$$

$$\times (\omega_3^2 \xi_3 \hat{x} + \omega_2^2 \xi_{2a} \hat{y} + \omega_2^2 \xi_{2b} \hat{z}) \ , \tag{7}$$

TABLE I. Stark quenching in $HeHe_\mu^+$.

v	*Ab initio*	$\gamma_{St}(sec^{-1})$ Morse	Harmonic oscillator
0	5.7×10^7	5.9×10^7	5.8×10^7
1	1.5×10^8	1.5×10^8	1.7×10^8
2	2.3×10^8	2.3×10^8	2.9×10^8
3	3.0×10^8	2.8×10^8	4.0×10^8
4	3.4×10^8	3.2×10^8	5.2×10^8
5	3.6×10^8	3.4×10^8	
6	3.8×10^8	3.5×10^8	
7	3.8×10^8	3.5×10^8	
8	3.7×10^8	3.3×10^8	
9	3.4×10^8	3.1×10^8	
10	3.0×10^8	2.7×10^8	
11	2.4×10^8	2.3×10^8	
12	1.7×10^8	1.9×10^8	
13	1.1×10^8	1.3×10^8	
14	4.9×10^7	7.9×10^7	
15	1.4×10^7	2.2×10^7	

in terms of the normal coordinate ξ_i, where the asymmetric stretching mode is designated by a subscript 3 and the bending mode by 2. The symmetric stretch makes no contribution since the field on the central atom vanishes by symmetry in that mode. Evaluation of Eq. (2) in the harmonic-oscillator approximation then yields

$$\gamma_{St}^{HO}(2) = \langle \Psi_1^{(v_1)} \Psi_{2a}^{(v_{2a})} \Psi_{2b}^{(v_{2b})} \Psi_3^{(v_3)} | A(\vec{R}) | \Psi_1^{(v_1)} \Psi_{2a}^{(v_{2a})} \Psi_{2b}^{(v_{2b})} \Psi_3^{(v_3)} \rangle$$

$$= \hbar \left[\frac{\gamma_0}{4\Delta^2} \right] \left[\frac{3a_\mu}{Z} \right]^2 (2\pi c)^3 \left[\frac{2mM}{2m+M} \right] [\omega_3^3(v_3 + \tfrac{1}{2}) + \omega_2^3(v_{2a} + v_{2b} + 1)] \ . \tag{8}$$

Other than the different vibrational frequencies, this expression differs from Eq. (6) only by the mass factor which depends on the transformation between normal and Cartesian coordinates. The vibrational frequencies for $He_2He_\mu^+$, determined from the frequencies given by Milleur et al.[6] for He_2H^+, are $\omega_2 = 511$ cm^{-1} and $\omega_3 = 855$ cm^{-1}. The result is

$$\gamma_{St}^{HO}(He_2He_\mu^+)$$

$$= 1.16 \times 10^7 [v_3 + 0.5 + 0.213(v_{2a} + v_{2b} + 1)] \ . \tag{8a}$$

in units of sec^{-1}. In the ground vibrational state this gives a quenching rate of 8×10^6 sec^{-1}. This rate is more than three orders of magnitude faster than the experimental upper limit for Stark quenching.[1(a)]

We may conclude that the clustering mechanism apparently does not account for the observed long lifetime of metastable muonic helium. In contrast, it seems to assure rapid quenching. On the other hand, one might note the ω^3 dependence of the quenching rate in Eq. (8) and speculate that the theoretical-structure calculations missed some stable complexes of the type He_nH^+ with $n \geqslant 3$. If larger complexes do exist they can be expected to have smaller vibrational frequencies; however, an order-of-magnitude decrease appears unlikely.

Another explanation, which has been considered for the long lifetime of metastable muonic helium, is that an electron may recombine with He_μ^+ to form a neutral atom, which would neither be collisionally quenched nor form clusters. This mechanism was rejected because of the lack of electrons and the rapid internal Auger process which would occur if the electron is in a $1s$ orbital.[1(c),5] The latter objection is dispelled if the electron is in a highly excited orbital,[11] but this possibility does not appear to be viable either since a diffuse electron could not be expected to prevent Stark mixing collisions.

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[8]The dissociation energy of He_2^+ is 2.47 eV [B. Liu, Phys. Rev. Lett. **27**, 1251 (1971)]; He_3^+ is linear and bound relative to He_2^+ by about 0.17 eV [P. L. Patterson, J. Chem. Phys. **48**, 3625 (1968); C. Vauge and J. L. Whitten, Chem. Phys. Lett. **13**, 541 (1972)]; much smaller currents of He_4^+ have also been observed but the molecule is not characterized [see, e.g., H. Helm, J. Phys. B **9**, 1171 (1976)].

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[11]Unpublished calculations by the author indicate that for $n \geq 5$ the average internal Auger rate would be less than the Auger rate (7.5×10^4 sec^{-1}) found in Ref. 1(b).