# Mean lifetime measurements of HeH$^{2+}$($2p\sigma$) isotopes

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The HeH$^{2+}$ molecular ion decays by an electronic dipole transition from its bound first excited state ($2p\sigma$) to the repulsive ground state ($1s\sigma$). We have calculated the mean lifetimes of the vibrational states of a few HeH$^{2+}$ isotopes and found a large isotopic effect, in particular for highly excited vibrational states, i.e., states with the same $v$ have different decay rates. The measured decay curves of $^4$HeH$^{2+}$, $^3$HeD$^{2+}$, and $^4$HeD$^{2+}$ ( i.e., the number of HeH$^{2+}$ molecular ions as a function of their flight time from the target cell where they were formed), in contrast, are similar to each other. The lack of a measurable isotopic effect is related to the HeH$^{2+}$ creation mechanism. In the charge-stripping collisions, a distribution of vibrational states is populated by vertical transitions and is thus centered around roughly the same vibrational energy and not around the same quantum number $v$. The mean lifetimes of the different isotopes as a function of their energy are surprisingly similar to each other, therefore washing out the isotopic effect. [S1050-2947(96)09606-0]

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## I. INTRODUCTION

The Schrödinger equation for one-electron molecular ions is separable in confocal elliptical coordinates within the Born-Oppenheimer approximation. Thus, molecular ions like H$_2^+$ and HeH$^{2+}$ are useful systems for improving our understanding of few-body problems. Increasing the repulsive potential term in the Hamiltonian generally results in systems without bound states which can be studied only by scattering methods. The asymmetric HeH$^{2+}$ molecular ion is an exception to this trend, because it has a bound first excited electronic state and a repulsive ground state, both shown in Fig. 1. This prediction of Bates and Carson [1] was later verified by Winter *et al.* [2], who also found additional higher lying bound excited electronic states. The lowest bound electronic state, $2p\sigma$, has a minimum 0.849 eV deep at $R_0 = 3.89$ a.u. which can sustain about 15 vibrational states.

Recently, we have reported experimental evidence of the formation of the bound $2p\sigma$ electronic state of the HeH$^{2+}$ molecular ions [3,4]. These molecular ions were produced in charge-stripping collisions of 900 keV HeH$^+$ with Ar gas. The mean lifetime of this state is short, of the order of a few ns, because it decays by dipole transition to the repulsive $1s\sigma$ ground state. The measurement of the mean lifetime of HeH$^{2+}$ is thus an experimental challenge because it travels only a few mm in one mean lifetime in the accelerators available for such studies. The calculated decay rates of these dipole transitions differ significantly from one vibrational state to another especially for highly excited states. The first mean lifetime measurements reported for this molecular ion were consistent with a wide distribution of vibrational states with different mean lifetimes as well as with a single exponential decay curve with a mean lifetime of $3.9 \pm 0.4$ ns [5].

In Sec. II of this paper we briefly describe the theoretical model used to calculate the mean lifetimes of some HeH$^{2+}$ isotopes. A few improvements of the experimental method

used previously [5] are presented in Sec. III. Measurements of the decay rates of $^4$HeH$^{2+}$, $^3$HeH$^{2+}$, and $^4$HeD$^{2+}$ isotopes are presented in Sec. IV. The measured and calculated decay rates of these isotopes are also compared to each other in the same section.

## II. THEORY

The decay of HeH$^{2+}$($2p\sigma$) molecular ions is predicted theoretically to proceed via an electronic transition to the
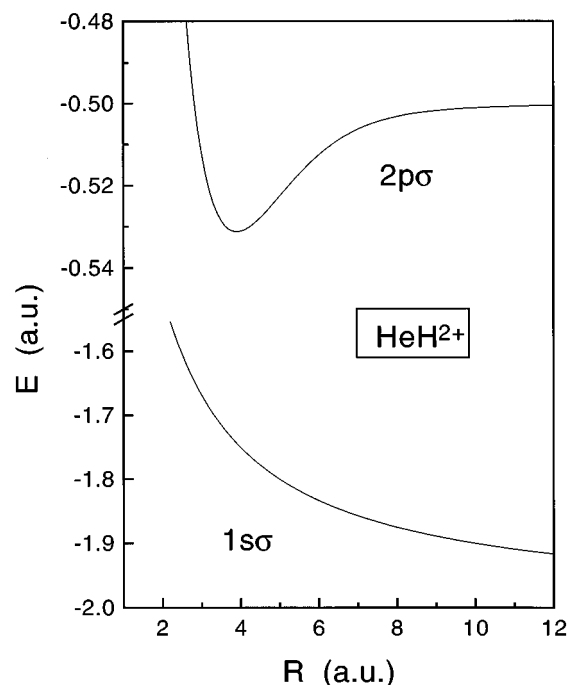


FIG. 1. Potential energy curves for the lowest states of HeH$^{2+}$ (zero corresponds to He$^{2+}$+H$^+$+$e$).

TABLE I. The mean lifetimes of the vibrational states of $^4$HeH$^{2+}$, $^3$HeD$^{2+}$, and $^4$HeD$^{2+}$.

| | $^4$HeH | | $^3$HeD | | $^4$HeD | |
|---|---|---|---|---|---|---|
| $v$ | $E_v$ (a.u.) | $\tau$ (ns) | $E_v$ (a.u.) | $\tau$ (ns) | $E_v$ (a.u.) | $\tau$ (ns) |
| 0 | -0.52911151 | 0.877 | -0.52949269 | 0.872 | -0.52957882 | 0.871 |
| 1 | -0.52509253 | 0.934 | -0.52617525 | 0.918 | -0.52642168 | 0.914 |
| 2 | -0.52131846 | 0.999 | -0.52301948 | 0.967 | -0.52340994 | 0.961 |
| 3 | -0.51789930 | 1.08 | -0.52003032 | 1.03 | -0.52054774 | 1.02 |
| 4 | -0.51454753 | 1.18 | -0.51721378 | 1.10 | -0.51784007 | 1.08 |
| 5 | -0.51157880 | 1.30 | -0.51457710 | 1.18 | -0.51529290 | 1.15 |
| 6 | -0.50891225 | 1.46 | -0.51212891 | 1.27 | -0.51291331 | 1.24 |
| 7 | -0.50657090 | 1.69 | -0.50987945 | 1.40 | -0.51070962 | 1.35 |
| 8 | -0.50458120 | 2.03 | -0.50784064 | 1.55 | -0.50869150 | 1.49 |
| 9 | -0.50297038 | 2.59 | -0.50602613 | 1.77 | -0.50687007 | 1.66 |
| 10 | -0.50175799 | 3.61 | -0.50445082 | 2.06 | -0.50525768 | 1.90 |
| 11 | -0.50093591 | 5.72 | -0.50312953 | 2.51 | -0.50386729 | 2.24 |
| 12 | -0.50044371 | 10.2 | -0.50207349 | 3.23 | -0.50271081 | 2.76 |
| 13 | -0.50018126 | 20.8 | -0.50128316 | 4.51 | -0.50179527 | 3.60 |
| 14 | -0.50005838 | 49.7 | -0.50073776 | 6.87 | -0.50111597 | 5.07 |
| 15 | -0.50001215 | 162.0 | -0.50039123 | 11.3 | -0.50064853 | 7.68 |
| 16 | -0.50000088 | 1150.0 | -0.50018690 | 20.3 | -0.50034943 | 12.6 |
| 17 | | | -0.50007656 | 40.4 | -0.50017045 | 22.1 |
| 18 | | | -0.50002452 | 95.8 | -0.50007192 | 43.0 |
| 19 | | | -0.50000500 | 316.0 | -0.50002415 | 98.6 |
| 20 | | | -0.50000034 | 2350.0 | -0.50000540 | 304.0 |
| 21 | | | | | -0.50000047 | 1860.0 |

HeH$^{2+}$($1s\sigma$) repulsive ground state which then dissociates rapidly into H$^+$ + He$^+$. These electronic transitions are referred to as vertical transitions because they are much faster than the nuclear motion. The mean lifetime of each vibrational state can be evaluated by averaging the spontaneous decay rate over all possible internuclear distances weighted by $|\psi_v(R)|^2$,

$$\tau_v^{-1} = \overline{W_{ka}^s} = \int_0^\infty \frac{2}{c^3} \omega_{ka}^2(R) f_{ka}(R) |\psi_v(R)|^2 dR, \qquad (1)$$

where $\omega_{ka}(R) = E_{2p\sigma}(R) - E_{1s\sigma}(R)$ is the transition frequency, $f_{ka}(R)$ is the oscillator strength, and $k$ and $a$ are the initial $2p\sigma$ and final $1s\sigma$ electronic states, respectively, as discussed in detail previously [5]. The mean lifetimes of a few HeH$^{2+}$ isotopes have been calculated using numerical vibrational wave functions, evaluated using the Fourier grid method [6], and the oscillator strength calculated by Arthurs et al. [7]. These values, presented in Table I, slowly increase with increasing vibrational quantum number for low values of $v$ as shown in Fig. 2, but increase rapidly for highly excited vibrational states. Furthermore, the mean lifetimes of highly excited vibrational states differ significantly from one isotope to the next. For example, the $v = 12$ state has a mean lifetime of 5.7, 2.5, and 2.2 ns for $^4$HeH$^{2+}$, $^3$HeD$^{2+}$, and $^4$HeD$^{2+}$, respectively. These differences are large enough to be detected experimentally if only this state is populated in the process used to create the HeH$^{2+}$($2p\sigma$). The previously measured mean lifetime is close to the mean lifetime of the $v = 10$ vibrational state [5] indicating that highly excited vi-

brational states play an important role. The main goal of this work was to investigate this isotopic effect.

The number of HeH$^{2+}$($2p\sigma$) molecular ions surviving after a given flight time depends on the population of its
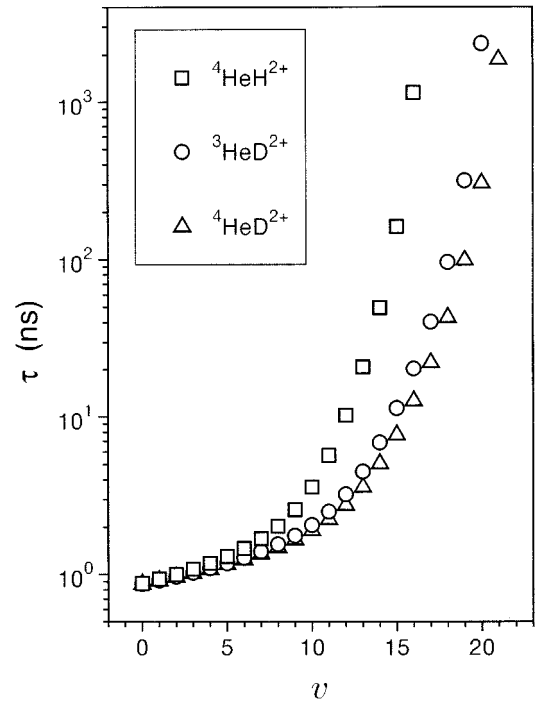


FIG. 2. The mean lifetimes of the different vibrational states of $^4$HeH$^{2+}$, $^3$HeD$^{2+}$, and $^4$HeD$^{2+}$ as a function of $v$.

vibrational states. A model was suggested to describe the whole process starting with the formation of $HeH^+$ molecular ions in the rf ion source of the accelerator, going through the $HeH^+ + Ar \rightarrow HeH^{2+}$ vertical transitions, and ending with the $2p\sigma \rightarrow 1s\sigma$ spontaneous electronic decay (see Ref. [5] for details). The fraction of $HeH^{2+}$ molecular ions as a function of their flight time is given by

$$\frac{N(t)}{N_0} = \sum_{v_f} \sum_{v_i} P_0 e^{-(E_{v_i} - E_0)/kT_{eff}}$$

$$\times \left| \int_0^\infty \psi_{v_i}^*(R)\psi_{v_f}(R)dR \right|^2 e^{-t/\tau_{v_f}}. \qquad (2)$$

The first term is the initial Boltzmann distribution of $HeH^+$ vibrational states with an effective temperature $T_{eff}$ which depends on the operating conditions of the ion source and is typically a few thousand degrees ($T_{eff}$ is the free parameter of the model). The second term is the charge-stripping transition probabilities given approximately by the square of the Franck-Condon overlap integrals (the vibrational wave functions of the electronic ground state of $HeH^+$ were calculated using the potential energy curve reported by Kołos and Peek [8]). The last term is the exponential decay of each vibrational state, where $\tau_v$, calculated using Eq. (1), are given in Table I.

### III. EXPERIMENTAL METHOD

The experimental method used for determining the decay rate of the $HeH^{2+}$ molecular ion is similar to the one used in the first mean lifetime measurements [5], and thus will be described only briefly. A 900 keV $HeH^+$ beam from the Technion Van de Graaff accelerator was directed through a differentially pumped target cell containing a thin Ar gas target where charge-stripping collisions took place. At this beam energy, the ions' speed is about 6 mm/ns. The ions produced in these collisions were analyzed within a few ns by a strong permanent magnet. In contrast to the previous experimental setup, this magnet is fixed, and the target cell is mounted on a translational stage. Thus, the trajectories of the ions passing through the analyzer do not change when the distance between the target cell and the analyzer is varied. Furthermore, the target cell length was reduced from 6 to 3 mm to improve the definition of the moment of creation of the $HeH^{2+}$, and the range of distances between the target cell and the analyzing magnet was increased, especially adding shorter distances, as shown in Fig. 3. All these changes have been made to enable the determination of deviations from a single exponential decay curve and determine how large is the isotopic effect. In addition to these changes, we have improved the method used for normalization in order to reduce the scatter in the data. Specifically, the $HeH^+$ rate was monitored directly instead of the neutral fragments used previously. This was accomplished by measuring the He fragments which were Rutherford scattered by 90° from a thin gold foil placed in the beam trajectory after the magnet. The number of $HeH^{2+}$ ions that passed the analyzer was determined from the number of ''fragment-fragment'' coincidences measured because no $HeH^{2+}$ can reach the detector
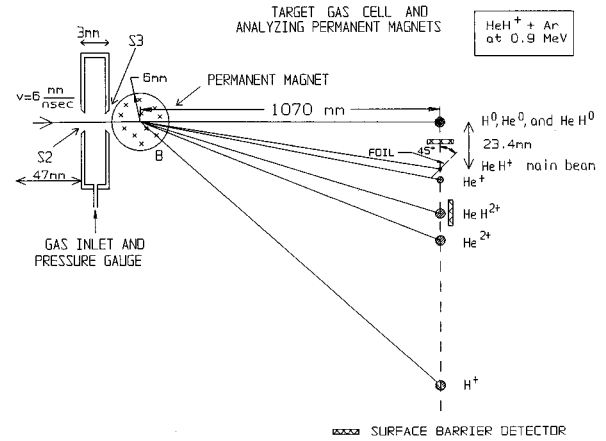


FIG. 3. A schematic view of the experimental setup.

before dissociating (see our previous publications for further experimental details [3–5]).

The number of $H^+ + He^+$ coincidence events, normalized to a constant number of helium fragments scattered off the thin gold foil, was measured as a function of their flight time from the moment of their creation all the way to the analyzing magnet exit. These coincidence events are the number of $HeH^{2+}$ molecular ions which survived all the way up to the magnet exit. The results of these measurements for the $^4HeH^{2+}$, $^3HeD^{2+}$, and $^4HeD^{2+}$ isotopes are presented in the following section. The $^3HeH^{2+}$ isotope was not measured because its parent molecular ion, namely, the $^3HeH^+$, cannot be separated from the $^4He^+$ atomic beam.

### IV. RESULTS AND DISCUSSION

The mean lifetime of the $HeH^{2+}$ molecular ions was evaluated from the direct measurement of their yield as a function of the flight time from the target cell to the exit of the analyzing magnet. It was shown previously that in the collisions forming the molecular ion of interest, i.e., $HeH^+ + Ar \rightarrow HeH^{2+}$ at 900 keV, only the bound $2p\sigma$ electronic state of $HeH^{2+}$ is populated [4]. This electronic state of $HeH^{2+}$ decays via an electronic transition to the $1s\sigma$ repulsive ground state which then dissociates rapidly into $H^+$ $+He^+$. A single exponential decay, with a mean lifetime of $3.9\pm0.4$ ns, was consistent with the data of Ref. [5]. The refined measurements we present here of the number of $^4HeH^{2+}$, $^3HeD^{2+}$, and $^4HeD^{2+}$ molecular ions as a function of their flight time are shown in Fig. 4. It can be clearly seen that the data for each isotope cannot be described by an exponential decay with a single mean lifetime. The model calculations, on the other hand, are in good agreement with the data. The effective temperature in the ion source (the free parameter of this model) was found to be about 8900 K for $^4HeH^{2+}$, 11 000 K for $^3HeD^{2+}$, and 5300 K for $^4HeD^{2+}$, all of which are well within the range of temperatures typical for this ion source. The data presented in this paper clearly indicates the existence of many decaying states with significantly different mean lifetimes. This deviation from a single exponential decay curve was not seen in the previous measurement because of the short range of flight times measured and the normalization method used.
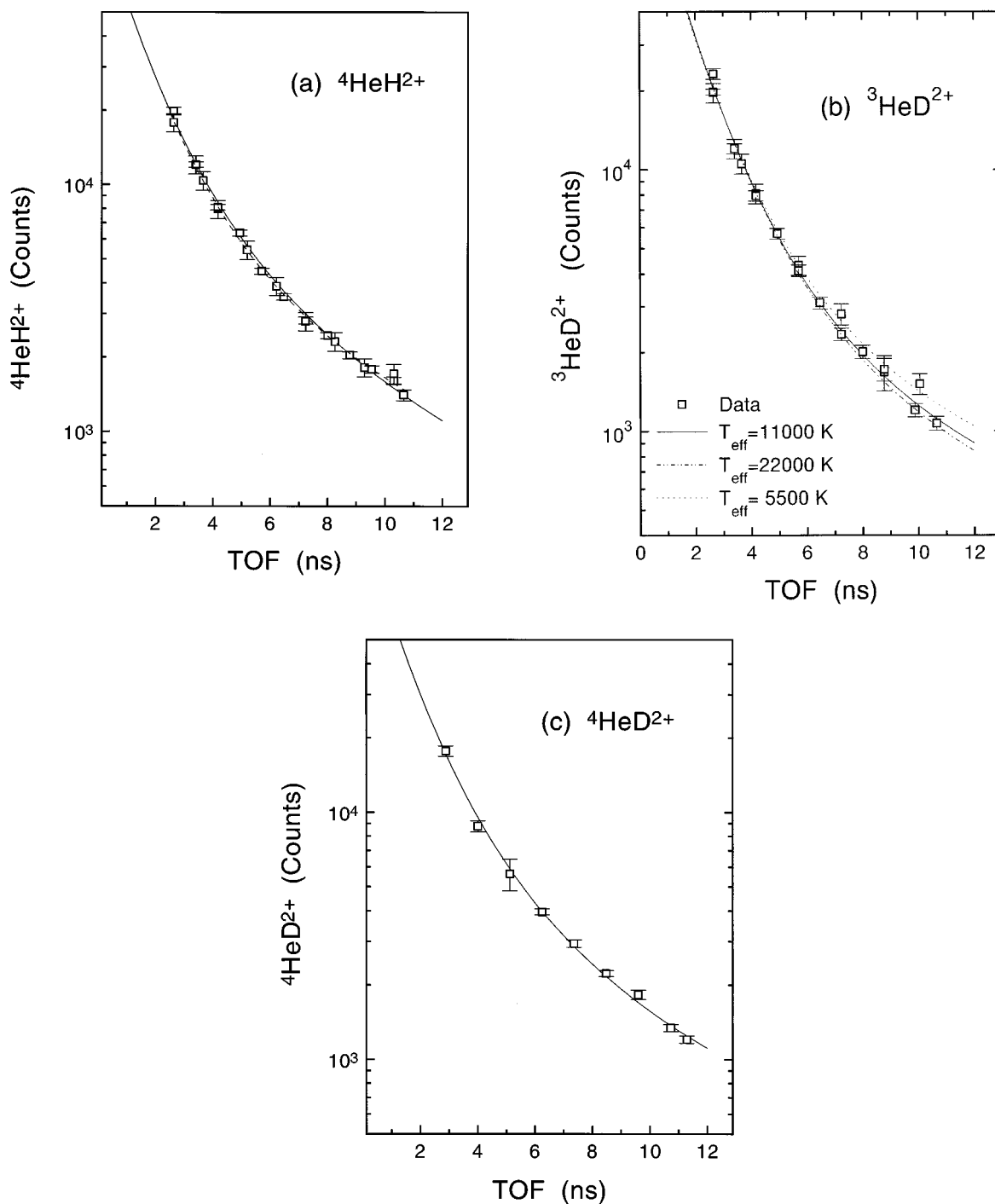
FIG. 4. The number of (a) $^4$HeH$^{2+}$, (b) $^3$HeD$^{2+}$, and (c) $^4$HeD$^{2+}$ as a function of their flight time from the target cell. The lines are a fit of the model described in the text to the data. In (a) the solid line is for $l=0$ and the dashed line is for $l=10$. In (b) the solid line is the best fit, while the dashed lines are for effective temperatures a factor of 2 higher and lower.

Similar measurements and model calculations have been conducted for other isotopes of HeH$^{2+}$. The results are shown for $^4$HeH$^{2+}$, $^3$HeD$^{2+}$, and $^4$HeD$^{2+}$ in Fig. 4. The model calculations and measurements compare well for all isotopes measured. The increased reduced mass lowers the vibrational energy levels and shrinks the distribution of internuclear distances, thus resulting in shorter mean lifetimes for the same vibrational state with larger reduced mass. However, this large effect shown in Fig. 2 causes no significant difference in the decay rates of the different isotopes.

The reason for the similarity between the decay curves of all HeH$^{2+}$ isotopes stems from the fact that the vertical transitions creating them preferentially populate vibrational states which are peaked around a certain value of vibrational energy and not states with the same vibrational quantum numbers, $v$, as demonstrated in Fig. 5. This peak value of the vibrational energy is determined by the population of vibrational states of the parent HeH$^+$ molecular ion and the Franck-Condon factors. Vibrational states with similar energies but different vibrational quantum numbers $v$ fall, sur-
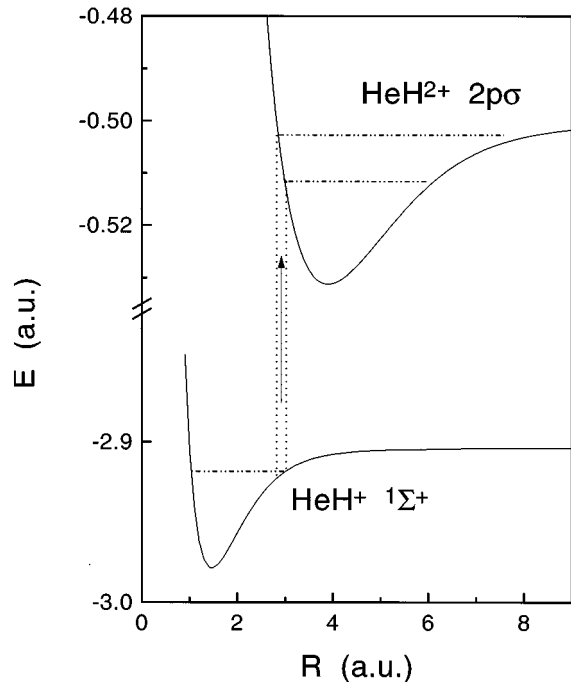
FIG. 5. The potential energy curves for the electronic ground state of HeH$^+$ (from Ref. [8]) and the lowest bound state of HeH$^{2+}$ (from our previous work Ref. [5]).

prisingly, on a ''universal'' curve plotted in Fig. 6. The number of nodes of the vibrational wave function has only a minor effect on the calculated mean lifetimes. Further theoretical work is needed to find the origin for this ''universal'' behavior.

The effective translational, vibrational, and rotational temperatures of the plasma are important parameters affected by the production mechanisms of the singly charged ions in ion sources. For example, Kanter *et al.* [9] showed that the internuclear distance of HeH$^+$ molecular ions is larger on the average in their rf source than in their duoplasmatron indicating a lower vibrational or rotational temperature (or both) in the latter. One might be tempted to think that mean lifetime measurements can also be used to evaluate the effective vibrational temperature of HeH$^+$ and other molecular ions. However, this is not possible because of the following two reasons: (i) the model fit to the decay curve is not that sensitive to the value of the effective vibrational temperature, as can be seen in Fig. 4(b). The range of possible vibrational temperatures can be narrowed by extending the measurements to longer flight times where the decay rate is more sensitive to the effective temperature. However, such measurements are not easy because of the very small rate. Furthermore, (ii) the HeH$^+$ molecular ions might be also rotationally excited. Ketterle *et al.* [10] reported similar effective rotational and vibrational temperatures (of about 3000K) for HeH$^+$ isotopes produced in their discharge ion source. We have also carried out the model calculations for $^4$HeH$^{2+}$ molecular ions with $l=10$. The angular momentum slightly stretches them (as reported by Kanter *et al.* [9]) causing an increase of the mean lifetimes. The decay curve of HeH$^{2+}$ with $l=10$ is also in good agreement with the data [see dashed line in Fig. 4(a)]. Even though this is a small effect for $l\sim10$, the effective temperature ($T_{eff}\sim$ 2000 K) is sig-
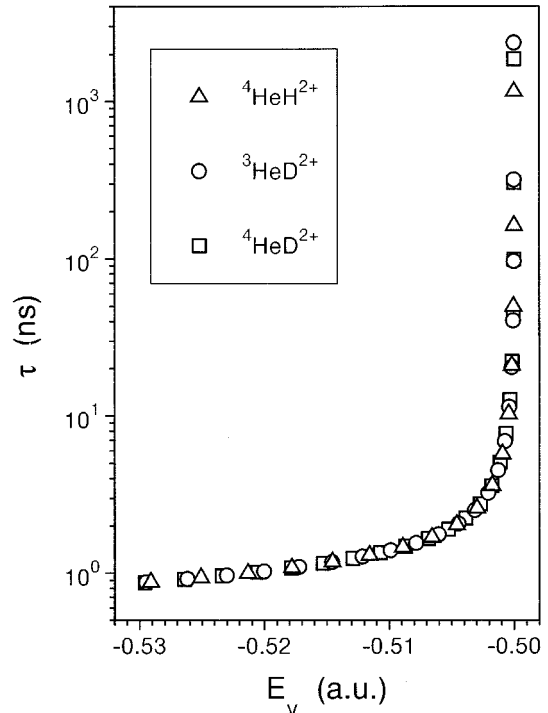


FIG. 6. The mean lifetimes of the different vibrational states, $v$, of $^4$HeH$^{2+}$, $^3$HeD$^{2+}$, and $^4$HeD$^{2+}$ as a function of $E_v$.

nificantly smaller than that of the $l=0$ case. Thus, decay rate measurements cannot be used to determine the plasma temperature in the source unless they are coupled with additional measurements such as internuclear distance measurements.

## V. SUMMARY

The number of HeH$^{2+}$ molecular ions was measured as a function of their flight time using the ''fragment-fragment'' coincidence method. The data is consistent with the decay of a wide distribution of vibrational states bound in the $2p\sigma$ potential energy curve of HeH$^{2+}$. The calculated mean lifetimes of vibrational states of different isotopes were found to depend on the reduced mass. The measured decay curves of a few different isotopes of HeH$^{2+}$, however, look alike. This similarity is caused by the creation mechanism of these molecular ions which populates a distribution of vibrational states centered around the same vibrational energy, and vibrational states with comparable energies have similar mean lifetimes for this molecular ion. In order to see the theoretically predicted large isotopic effect the vibrational state of the HeH$^{2+}$ molecular ions have to be either prepared or measured. This leaves significant experimental challenges for the future.

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