Low temperature behaviour of collisions between antiprotonic helium and hydrogenic molecules and an indication of the Wigner threshold law

B. Juhašsz a,*,1, D. Barna b,2, J. Eades b, H. Fuhrmann c, R.S. Hayano b, M. Hori d, D. Horváth a,e, H.A. Torii f, E. Widmann b,1, H. Yamaguchi b, T. Yamazaki g, J. Zmeskal c

a Institute of Nuclear Research of the Hungarian Academy of Sciences, H-4026 Debrecen, Hungary
b Department of Physics, University of Tokyo, 7-3-1 Hongo, Bunkyo-ku, Tokyo 113-0033, Japan
c Stefan Meyer Institut für subatomare Physik, Boltzmanngasse 3, A-1090 Vienna, Austria
d CERN, CH-1211 Geneva 23, Switzerland
e KFKI Research Institute for Particle and Nuclear Physics, H-1325 Budapest, Hungary
f Institute of Physics, University of Tokyo, 3-8-1 Komaba, Meguro-ku, Tokyo 153-8902, Japan
g RIKEN, Wako, Saitama 351-0198, Japan

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Abstract

The temperature dependence of the quenching cross section of two long-lived metastable states of antiprotonic helium, in collisions with hydrogen and deuterium molecules, has been measured. In the case of the state \((n, l) = (37,34)\), the cross section decreases with decreasing temperature until it levels off below \(\sim 30\, \text{K}\), showing a quantum tunneling effect with a small activation barrier. In the case of the state \((39,35)\), on the other hand, for which the lack of an activation barrier was predicted theoretically, the quenching cross section increases slightly with decreasing temperature, indicating the \(1/\nu\) Wigner threshold law of exothermic reactions involving neutral particles.

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1. Introduction

Antiprotonic helium is an exotic metastable atom produced in a helium medium, when an incoming antiproton \((\bar{p})\) is captured by a helium atom, replacing one of its electrons to form \(\bar{pHe}^+ \equiv \bar{p} - e^- - He^{2+}\), with large antiproton principal and orbital quantum numbers \((n, l) \sim (39,38)\) (cf. [1]). In the past decade, collisions between antiprotonic helium atoms and hydrogenic molecules \((H_2\) and \(D_2)\) at low temperatures \((<100\, \text{K})\) have been studied. The overall lifetime \((\sim 4\, \mu\text{s})\) of the metastable states of \(\bar{pHe}^+\) was found to decrease when the \(\bar{pHe}^+\) atoms collide with \(H_2/D_2\) molecules [2-4]. With the advent of laser spectroscopy, the interactions of individual metastable states with \(H_2/D_2\) molecules were successfully investigated. The quenching phenomenon thus revealed is of great interest because; (i) strong quenching persists at low temperatures below \(30\, \text{K}\), (ii) the quenching cross section \(\sigma_q\) strongly depends on the quantum numbers \((n, l)\) of the \(\bar{pHe}^+\) atom [5-7], and (iii) \(\sigma_q\) is usually a factor of \(\sim 1.5\) smaller for \(D_2\) than for \(H_2\) [8,9].

From the physicochemical point of view, an antiprotonic helium atom resembles an exotic hydrogen atom in which the electron in its 1s orbit is bound to a \([\bar{pHe}^+](n,l)\) ‘nucleus’, which has an \((n,l)\)-dependent effective charge...
The barrier height, the pre-exponential cross section and the tunneling cross section of an antiprotonic state can be obtained by measuring \( \sigma^{(n,l)}_q \) at different temperatures and fitting Eq. (3) to the data. We already made such measurements on the antiprotonic states \((n,l) = (38,37)\) and \((37,34)\), and concluded that \( \sigma^{(n,l)}_q \) is the dominant term at temperatures below 30 K, and the temperature dependence of \( \sigma^{(n,l)}_q \) does follow Eq. (3) in the case of these two states [13]. Now we report the continuation of these investigations; we measured the quenching cross section of the state \((37,34)\) at more temperature points and made a detailed temperature scan of the state \((39,35)\). Fig. 2 shows these states in the energy level diagram of \( \text{pHe}^+ \). Previously, the latter state has only been studied at 30 K, where quenching cross sections of \((29.2 \pm 1.1) \times 10^{-16} \text{cm}^2\) and \((30.8 \pm 1.4) \times 10^{-16} \text{cm}^2\) have been obtained for \( \text{H}_2 \) and \( \text{D}_2 \), respectively [8]. These values are close to the geometrical cross section \( \sigma_{\text{geom}} = 21 \times 10^{-16} \text{cm}^2 \) of the \( \text{H}_2 – \text{He} \) and \( \text{D}_2 – \text{He} \) collisions [14]. This, and also the calculations of Sauge and Valiron, suggested that there is no activation barrier for the state \((39,35)\) and thus all collisions result in quenching. Due to the lack of the activation barrier, one might expect a temperature-independent quenching cross section.

2. Experimental setup and method

The measurements were carried out at CERN’s Antiproton Decelerator (AD). The experimental setup was almost identical to our previous measurements [13,15,16]. Antiprotonic helium atoms were created by stopping pulses of \( \sim 3 \times 10^7 \) antiprotons, with an energy of 5.3 MeV, in a cryogenic helium target every \( \sim 100 \) s. After slowing down by collisions with helium atoms, about 3% of the antiprotons are captured into various long-lived metastable states with \( n = 36-40 \) and \( l = n - 1 \). The formed \( \text{pHe}^+ \) atoms rapidly reach thermal equilibrium with the surrounding helium atoms. The subsequent annihilations of the captured antiprotons (mainly into energetic charged pions) were observed as a light pulse, lasting several microseconds, in two adjacent acrylic Cherenkov counters. The light was collected by two gateable fine-mesh photomultipliers, the
outputs of which were recorded using a digital oscilloscope as an ‘analog delayed annihilation time spectrum’, which is essentially the annihilation rate of the antiprotons versus time. The gas target consisted of ⁴He (purity: 99.9996%) to which hydrogen (purity: 99.999%) or deuterium (purity: 99.8%) was premixed at molar concentrations of 30–10000 ppm, with a nominal relative accuracy of 2%. The final relative accuracy was calculated as the quadratic sum of the nominal 2% error and a ‘fluctuation error’, which originated from absorption and desorption of H₂/D₂ molecules on the target chamber surfaces. The latter was assigned the following conservative values based on offline tests with a gas chromatograph [17]:

- 10 ppm admixtures: 10%, 8%, and 4% at 15, 17, and 20 K, respectively.
- 30 ppm admixtures: 5%, 4%, and 2% at 15, 17, and 20 K, respectively.
- 100 ppm admixtures: 2% and 1%, at 15 and 17 K, respectively.

The assigned fluctuation error was 0% at temperatures above 20 K or at concentrations above 100 ppm.

The quenching cross section of a metastable antiprotonic state can be obtained by measuring its decay rate γ at different admixture (hydrogen or deuterium) concentrations. Assuming that the antiprotonic helium atoms are quenched in binary collisions with the admixture molecules, this can be expressed in terms of the state-dependent quenching cross section σ_q as

\[ \gamma = \gamma_0 + \gamma_{He} + n_{adm}v_{th}\sigma_q^{(n,l)}, \]

where γ_0 is the ‘intrinsic’ decay rate of the state in a collision-free environment (this is calculated theoretically), γ_{He} is the decay rate caused by collisions with helium atoms [this is negligibly small for all states except (37,34)], n_{adm} is the absolute number density of the admixture molecules, and v_{th} = \sqrt{8k_B T/(\pi M_{red})} is the average relative thermal velocity, with M_{red} being the reduced mass of the colliding species [18]. If we measure γ at different admixture number densities at a given temperature, plot γ' = γ - γ_{He} versus n_{adm}v_{th} and fit a straight line to the data points, then the tangent of this line will give σ_q^{(n,l)} at the given temperature. The number density n_{adm} of the H₂ and D₂ molecules was varied either by changing the gas mixture i.e. the admixture concentration or by changing the pressure of the gas target.

To measure the decay rate γ of a metastable state, we induced resonant laser transitions (see Fig. 2) between this state and a short-lived (≤ ns) state with a laser pulse, after the arrival of each antiproton pulse. The laser pulse was produced by a dye laser pumped by a Nd:YAG laser, and it was directed into the helium target where the antiprotonic helium atoms formed. This resulted in a sharp peak in the annihilation time spectrum. The area of this peak is proportional to the antiproton population of the parent state. The decay rate can then be obtained by measuring the population as a function of time. A detailed description of this ‘t1-scan’ method can be found in our previous publication [13].

The analysis of the collected data was done very similarly to our previous measurements [9]. However, this time the quenching caused by helium atoms was also taken into account in the case of the state (37,34) [19], as it was mentioned above. In the case of the state (39,35), collisions with helium atoms do not cause quenching [20].

The rate constant k, which is usually used to describe the rate of a reaction, can be expressed using the velocity-dependent reactional cross section \( \sigma(v) \) as \( k = \int \sigma(v) v f(v) dv \), where \( f(v) \) is the Maxwell–Boltzmann velocity distribution. For a given temperature, one can find an average cross section \( \langle \sigma \rangle \) such that \( k = \int \sigma(v) v f(v) dv = \langle \sigma \rangle v_{th} \). In our case, the measured quenching cross section \( \sigma_q^{(n,l)} \) is such an average cross section.

3. Results and discussion

Fig. 3 shows the newly and previously [7,8,13] measured quenching cross sections of the antiprotonic states (37,34) and (39,35) versus the inverse temperature (Arrhenius plot). It can be clearly seen from the figure that the quenching cross section of the state (37,34) follows Eq. (3). Previously, the cross sections with H₂ and D₂ were known only at three temperatures: 30, 100, and 300 K [8,13], which was not enough for a reliable fit of Eq. (3). However, with the

![Fig. 3. Arrhenius plot of the newly and previously measured quenching cross sections of the states (37,34) (triangles) and (39,35) (circles). The plotted curve is the result of fitting Eq. (3) to the data points of (37,34) with H₂. For a discussion on the large error of the data point of the state (39,35) with D₂ at 60 K, see Ref. [19].](image-url)
three new data points with H\textsubscript{2} (at 15, 20, and 60 K), which
align quite well with the old data points, it became possible to
fit Eq. (3) to the data of the state (37, 34) with H\textsubscript{2}. This fit
(see the curve in Fig. 3) yields the following results:

\[ E_{\text{th}}^{(37,34)} = (16.8 \pm 3.1) \times 10^{-3} \text{eV}, \quad \sigma_{\text{q}}^{(37,34)} = (6.2 \pm 1.9) \times 10^{-16} \text{cm}^2, \text{ and } \sigma_{\text{r}}^{(37,34)} = (1.10 \pm 0.05) \times 10^{-16} \text{cm}^2. \]

These values are quite close to the ones obtained previously by
simultaneously fitting the data of the states (37, 34) and
(38, 37) [13].

The quenching cross section of the state (39,35), however,
shows a completely different behaviour: there is no
Arrhenius term, and the cross section increases slightly
with decreasing temperature, both with H\textsubscript{2} and D\textsubscript{2}, con-
trary to the naive assumption that it should be tempera-
ture-independent and close to the geometrical cross
section. In fact, at the lowest temperature (15 K), the mea-
sured \( \sigma_{\text{q}} \) is almost twice as large as the geometrical cross
section. To exclude the possibility that this increase was
caused by some measurement error, \( \sigma_{\text{q}} \) with D\textsubscript{2} was re-
measured at 30 K; this was already measured before [8].
Both the old and the new values are plotted in Fig. 3 at
30 K (upper and lower empty circles, respectively). They
agree reasonably well.

There is also a similar increase of \( \sigma_{\text{q}} \) below 20 K in case
of the state (37, 34) with H\textsubscript{2} (see Fig. 3), but that is much
less significant and it might just be statistical scattering of
the data points.

One might think that the increase in the measured \( \sigma_{\text{q}} \) is
just a temperature-dependent quenching caused by helium
atoms. However, previous measurements showed that
helium atoms do not cause any significant quenching at
6 K [20], and other measurements [19] on the population
decay rate of the state (39,35) at 15 and 30 K indicated that
quenching by helium is also negligible at these tempera-
tures. Contamination by oxygen or nitrogen molecules –
which can also cause strong quenching [4,21] – is unlikely
to cause an increase of the measured cross section either.
This is because the maximum (saturation) vapour pressure
of these gases is negligibly small at such low temperatures
(20 K and below).

Fig. 4 shows the quenching cross section of the state
(39,35) with D\textsubscript{2} as a function of the inverse collisional velocity \( 1/v_{\text{th}} \). The data points are quite well aligned on a
straight line (the correlation factor of the points is 0.849).
Namely, they satisfy the following relation:

\[ \sigma_{\text{q}}^{(39,35)} = \sigma_{\text{q}i} + \alpha \frac{1}{v_{\text{th}}}. \]  

The result of a fit to this relation is also drawn in Fig. 4.
The velocity-independent (i.e. energy-independent) term is
\( \sigma_{\text{q}i} = (13.0 \pm 6.3) \times 10^{-16} \text{cm}^2 \), while the tangent of the line is
\( \alpha = (8.7 \pm 3.4) \times 10^{-13} \text{cm}^2 \text{m/s} \). This fit has a reduced
chi-square \( \chi_{\text{red}}^2 = 0.87 \), while a fit of a constant line has
\( \chi_{\text{red}}^2 = 1.83 \).

Such a cross section dependence on the inverse velocity
is indicative of the Wigner threshold law which states –
among other things – that in case of an exothermic reaction

involving neutral particles near a threshold, the cross sec-
tion is expected to follow an \( 1/v \) dependence in the low tem-
perature limit [22,23].

There was insufficient time to measure the quenching
cross section above 60 K, therefore it is not known whether
it keeps a \( 1/v \) dependence at high temperatures, or on the
contrary, levels off around the geometric cross section.

The Wigner threshold law should be revealed more
dominantly at lower temperatures. Unfortunately, we
could not measure the quenching cross section below
15 K, because at such low temperatures the hydrogen and
the deuterium molecules can freeze out from the admixture,
completely spoiling the experiment.

There are no theoretical calculations on the pH\textsuperscript{+}-H\textsubscript{2} or
pHe\textsuperscript{+}-D\textsubscript{2} systems which would investigate the temperature
dependence in detail. Such calculations are therefore
awaited. There are, however, calculations on similar sys-
tems. One calculation on the H + H\textsubscript{2} and similar reactions
showed that the rate constants \( k \sim \sigma v \) of the \( \text{D} + \text{H}_{2} \rightarrow \text{DH} + \text{H} \) and the \( \text{D} + \text{DH} \rightarrow \text{D}_{2} + \text{H} \) exothermic exchange reactions are independent of the temperature below \( \sim 10 \) K (i.e. the cross section is proportional to \( 1/v \)) [24,25]. Recent
measurements on these reactions in solids, in the tempera-
ture range of 4–7 K, agreed with these calculations [26].
Another calculation on the relaxation of vibrationally
excited H\textsubscript{2} molecules in collisions with \( ^{3}\text{He} \) and \( ^{4}\text{He} \) atoms
predicted a \( 1/v \) dependence of the cross section of this pro-
cess below \( \sim 30 \) K [27], whereas the cross section was mea-
sured only down to 50 K, above which an Arrhenius-type
temperature dependence was seen [28].

Further experimental data are also available on similar
systems. Rate constants of the reactions of muonium
(Mu) with the halogen gases F\textsubscript{2}, Cl\textsubscript{2}, and Br\textsubscript{2} have been
measured in a wide (but relatively high) temperature range
The rate constant in the $\text{Mu} + \text{Br}_2$ reaction was found to increase slightly with decreasing temperature, indicating a large increase of the cross section. On the other hand, the $\text{Mu} + \text{F}_2$ and $\text{Mu} + \text{Cl}_2$ reactions had decreasing rate constants, and also decreasing cross sections. Recently, in an experiment where H atoms were added to sprayed thin films of $\text{C}_2\text{H}_2$ and $\text{C}_2\text{H}_4$ to form $\text{C}_2\text{H}_6$, an enhancement in the yield was observed at temperatures below 20 K. However, this is most likely ascribed to possible increase of H atoms on the films at low temperatures, and may not be relevant to the Wigner threshold law.

In summary, the present laser-tagged measurements of the quenching cross sections of individual metastable states of antiprotonic helium, in collisions with $\text{H}_2$ and $\text{D}_2$ at low temperatures ($15–60$ K), have shown the following results.

(i) The quenching of the state $(n, l) = (37,34)$ follows the Arrhenius temperature dependence above $50$ K, which is explained by the activation barriers predicted by Sauge and Valiron [12], and levels off below $30$ K, indicating a quantum tunneling effect.

(ii) On the other hand, the quenching of the state $(39,35)$ shows no such Arrhenius-type temperature dependence, and a large cross section, of the order of the geometrical cross section, is observed at low temperatures. This can be understood by the predicted absence of the activation barrier. The cross section increases gradually with the decrease of the temperature below $30$ K, showing a $1/v$ behaviour, which is consistent with the Wigner threshold law.

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