Collisional Quenching of Metastable Hydrogen Atoms*

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We have measured the cross section for the quenching (i.e., depopulation) of the $2s$ (metastable) state of atomic hydrogen by collisions with atoms of helium and argon. The energy range covered was 0.25–30.0 keV. The experimental technique was to measure the attenuation of a beam passing through the various gases, the detector being a Lyman-α counter viewing a section of the beam where a dc electric field mixed $2s$ and $2p$ states, which essentially converted metastable atoms into Lyman-α photons. For helium as target gas, the cross section was also calculated using a pseudopotential and the eikonal approximation. For helium, the measured cross section falls rapidly as the energy increases from 0.25 to 2.5 keV and remains roughly constant at $7.5\pi a_0^2$ from 2.5 to 30 keV. The theoretical curve is about a factor of 4 below the measured value. Implications of this discrepancy are discussed in the text. For argon, the measured cross section increases from $20\pi a_0^2$ to $27\pi a_0^2$ as the energy increases from 1 to 30 keV.

I. INTRODUCTION

We have measured the total cross section σ_Q for the quenching of metastable hydrogen atoms in collisions with atoms of helium and argon, and have calculated this cross section for helium. "Quenching" means any process that removes atoms from the $2s$ state, so that the total cross section may be thought of as a sum of partial ones for stripping (ionization of the metastable atom), pickup (formation of H^-), and deexcitation (transition to other states such as $2p$, followed by decay to the ground state). The cross section for elastic scattering is not included in that for quenching. The energy range covered is 0.25–30 keV, corresponding to relative velocities from 0.1 to 1.1 a. u.

Our main purpose in this work has been the classical one of comparing theory with experiment. There are very few cases of atomic collisions where there are available both a reliable (to 10% or better) absolute measurement of an inelastic cross section and an equally reliable calculation from first principles. Our results fall short of this ideal in that the estimated error in our cross sections is 20% instead of 10%, and in that for practical reasons the target atoms were chosen to be more complicated than could be handled from first principles only.

Some insight into the relative importance of various quenching mechanisms can be gained from the adiabatic criterion. Let v (in a. u.) be the relative velocity of the two atoms, and ϵ be the change (in a. u.) in the internal energy of the colliding pair. According to the adiabatic criterion, a given quenching mechanism, corresponding to a certain ϵ, would be negligibly important at velocities such that $\epsilon/v \gg 1$.

At thermal energies, the dominant quenching mechanism should be $2s$-$2p$ transitions, since even for $2s$-$3p$ transitions the parameter ϵ/v is much more than 1 ($\epsilon/v = 70$ at 0.025 eV). At ~100 eV, ϵ/v becomes 1 for $n = 2$ to 3 transitions, and these should become important. At slightly higher energies other transitions to higher bound states should begin to play a role. At ~400 eV, ϵ/v is 1 for transitions between $n = 2$ and the ionization limit so at energies higher than this, stripping (ionization of the metastable) could become important. Similarly ~13 keV is the expected threshold for a $2s$-$2p$ transition in the hydrogen atom, accompanied by excitation of the helium to a low-lying level.

At thermal energies, measurements of quenching have been reported by Fite *et al.*,[1] Comes and Wenning,[2] and Kass and Williams.[3] At higher energies, overlapping with our energy range, measurements have been reported by Spiess *et al.*,[4] (2.5 keV), Gilbody *et al.*[5] (10–30 keV), and Dose *et al.*[6] (2–60 keV). Their results for helium and argon are shown in Figs 2 and 3. For completeness we should also mention Ref. 7 (Byron *et al.*), which is a preliminary report on results more fully presented here.

Spiess[4] and Gilbody *et al.*[5] measured not only the total quenching cross section, but also that part of it due to stripping (ionization of the metastable). As expected from the adiabatic criterion, they found that stripping was an important quenching mechanism, typically accounting for more than half of the total cross section.

On the theoretical side, calculations of quenching cross sections have been reported by Gersten,[8] Byron and Gersten,[9] and by Byron *et al.*[7] Gersten's paper[8] considers quenching by molecules; Byron and Gersten[9] deal with helium, but their method is intended for energies somewhat lower than our lower limit of 0.25 keV.

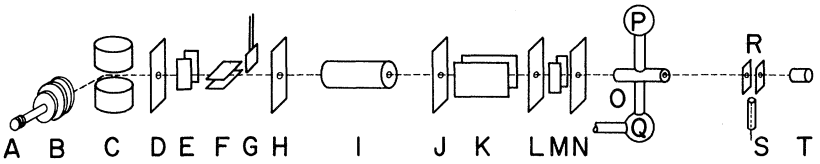

FIG. 1. Schematic diagram of the apparatus. For explanation of letters see Sec. II.

II. APPARATUS

Conceptually, the experiment was very simple. We measured the attenuation of a beam of metastables upon passage through a gas cell target, and inferred the quenching cross section from the pressure dependence of this attenuation. A schematic diagram of the apparatus is shown in Fig. 1. Briefly, protons from an ion source A were converted into metastable atoms at the cesium cell I. The attenuation of the metastable beam was measured by a detector consisting of quench plates R and geiger counter S. A more detailed description follows.

Ions (H^+, H_2^+, H_3^+) were extracted from a commercial (Ortec) rf ion source A, focused by an einzel lens B, and bent through 30° by a magnet C to select protons. The extracted ions typically had an energy of 3.0 keV, and never more than 4.5 keV. To obtain higher energies, the beam was accelerated by an electric field at D. To obtain lower energies, either the gap was used as a decelerator, or the ions were extracted from the source at a lower voltage.

Steering plates E and F were used to trim the direction of the beam which then passed through a cell I containing cesium vapor. The cesium cell was 15 cm long. Its entrance and exit apertures were 1 cm in diameter. A typical operating pressure was 6×10^{-4} torr (the vapor pressure of cesium at 100 °C). Water-cooled snouts (not shown in Fig. 1) collected cesium that otherwise would have condensed in other parts of the apparatus. Liquid-cesium metal was loaded into the cell under an argon blanket and atmospheric pressure, a 2-g charge lasting several months. We found it important to pump out the argon either with a sorption pump or well-trapped mechanical pump. Otherwise, pump oil condensed on the originally shiny cesium surface, forming a dark crust which required over heating to break, and which reformed each time the cesium was cooled.

Two metal plates at K (clearing field) provided a transverse dc electric field which swept out charged beam components which were collected at the next aperture L. The electric field E required to deflect protons is given by Et^2 = constant, where t is the time atoms spend in the field, and the constant depends on geometry. Metastables are quenched by this field, the fraction quenched depending on the combination E^2t. Hence by making the field long enough, it is possible to arrange that the attenuation of metastables by the field be as small as desired. In our case, the quenching was only a few percent at 3 keV, and increased to about 50% at 30 keV.

The neutral beam issuing from the aperture L consisted of hydrogen atoms in the ground state, in the metastable state, and possibly in higher excited states, some of which might be sufficiently long lived to travel all the way down the rest of the apparatus. Plates M were used to prequench the beam by applying a (transverse) field strong enough to convert all metastable atoms into ground-state atoms. The prequench field was used to prove we had metastables and to discrimate against ground-state atoms, as described in Sec. III. The field is expected to have little effect on atoms in highly excited states, because the Lamb shift and fine-structure splittings in these are so small that even the clearing field K saturated all transitions. That is, the relative populations of atoms in various highly excited states should have been independent of a further increase in field at M. On the other hand, the prequench field M was downstream from the clearing field K, and one might argue that, because of the elapsed time M might have had some small effect. To check this hypothesis, rather involved but straightforward calculations would have to be carried out. These have not yet been done.

After leaving the prequench region, the beam passed through the gas cell O, which was 16.2 cm long. The entrance aperture, of 1 mm diameter, and the exit aperture, 4.8 mm in diameter, could each be moved under vacuum in the horizontal and vertical directions. The purpose of this arrangement was to ensure that the metastable beam was passing down the symmetry axis of the gas cell.

The effective length of the gas cell was estimated to differ from its geometric length by less than 0.5%, and hence this correction was ignored. As has been shown by Howard,[10] on the axis of a circular aperture the increase in number density outside is exactly balanced by the decrease inside. There remain corrections due to the finite wall thickness and to the increase in base pressure of the adjoining vacuum chamber. It is these that were estimated to be less than 0.5%.

The pressure in the gas cell was measured by a capacitance manometer P (MKS Baratron) which

was connected to the cell by a "dead" line, i.e., one in which gas was not flowing so that no pressure drops occurred along it. After all data had been taken, the Baratron was recalibrated by its manufacturers. The error in the absolute-pressure calibration was taken to be less than 0.3%, which is negligible.

Gas was fed into the cell through a commercial leak valve Q (made by Varian Corp.) across which the pressure dropped from atmospheric to that in the gas cell. Between the gas tank and the leak valve, the gas was filtered by passing through about 2 m of $\frac{1}{4}$-in. copper line held at a low temperature (liquid nitrogen for helium, dry ice and acetone for argon). At low pressures, immediately after the leak valve, the gas was again filtered by passage through a titanium sublimation pump.

At R, metastable atoms leaving the gas cell passed through a longitudinal dc electric field which mixed the $2s_{1/2}$ and $2p_{1/2}$ states so that some fraction (depending on the field strength) decayed with emission of a Lyman-α photon. This photon was detected by an iodine-helium geiger counter S whose count rate was then proportional to the current of metastable atoms leaving the gas cell. Since the quenching cross section could be calculated from the *fractional* decrease in count rate with increasing pressure, it was not necessary to know the magnitude of this proportionality constant. (For the same reason, it was not necessary to know the details of the angular distribution into which the Lyman-α photons were emitted.)

The iodine-helium counter was similar to that described by Brackmann *et al.*[11] and Dose.[6] Ours was constructed of stainless steel with the central wire 1.5 mm in diameter and the outer shell 2.5 cm in diameter. Windows and feedthroughs were fastened in place with outgassed Epoxy (Armstrong C7 with activator W). There was provision for periodic refilling of the counter. The iodine crystals were contained in a reservoir which was part of the helium feedline. We found it important to polish the central wire (or else the output pulses could be large, jagged, and followed by satellite pulses), to keep iodine crystals away from direct contact with the Epoxy (or else the Epoxy would be attacked and leaks develop), and to keep the iodine reservoir at least approximately temperature controlled (or else the counter efficiency would vary with room temperature). Our reservoir was water cooled, its temperature remaining constant to 0.5 °C or better over 6 h. For stability, it was important to bake the counter (several hours at 120 °C, while being pumped by a trapped mechanical pump). When viewed directly on an oscilloscope, through a suitable blocking capacitor but without any preamplifier, the output pulses had an amplitude of 5 V, and a risetime of less than 0.1 μsec. The dead time of the counter was 30 μsec, and typical counting rates were 10^3 sec^{-1}. All observed count rates were corrected for dead time, the correction at most amounting to 5%.

The total neutral current was monitored at T by measuring the current of secondary electrons ejected from a stainless-steel post biased at -22 V with respect to a grounded shield around it. Typical currents ranged from 0.1 to 10×10^{-10} A, depending on the beam energy. The base pressure in the vacuum system was 2×10^{-7} Torr.

III. EXPERIMENTAL METHOD AND RESULTS

A. Method of Analysis

The beam contained not only metastable, but also ground-state, atoms. Although the detector R, S was for all practical purposes totally insensitive to the ground-state component, this component gave trouble in that it produced metastables in the gas cell through the reaction H $(1s)+X\rightarrow$ H $(2s)+X'$ (where X, X' denote states of the target atom). At a given gas-cell pressure p, signals proportional to the ground-state current could be eliminated by taking the difference $\Delta(p)$ between the signal $S(p)$ (in counts per sec) with the prequench field off, and the signal $S_0(p)$ observed with the prequench field turned on hard enough to convert all metastable atoms to ground-state atoms. More specifically, if n is the number density of target atoms in the gas cell and l its length, then for sufficiently small n, S and S_0 are given by the equations

$$\begin{aligned} S(p) &= I^* \ (1 - n\sigma_Q l) + I_0 n\sigma_{1s\,2s} l \ , \\ S_0(p) &= (I^* + I_0) n\sigma_{1s\,2s} l \ , \end{aligned} \tag{1}$$

where I^* is the product of metastable current (in atoms/sec) and detector efficiency, while I_0 is the similar product for ground-state atoms. The count rates $S(p)$ and $S_0(p)$ depend on both I^* and I_0. However, their difference $\Delta(p)$ is proportional to I^* only:

$$\Delta(p) \equiv S(p) - S_0(p) = I^* \ [1 - n \ (\sigma_Q + \sigma_{1s2s}) \, l] \ , \tag{2}$$

and the ratio $\Delta(p)/\Delta(0)$ is independent of I^*:

$$\Delta(p)/\Delta(0) = 1 - n \ (\sigma_Q + \sigma_{1s\,2s}) l \ . \tag{3}$$

Equation (3), of course, only holds for pressures low enough that the second term on the right-hand side is small compared to the first. At higher pressures, the graph of $\Delta(p)/\Delta(0)$ deviates from a straight line. If *only* quenching processes were to take place, and there were no mechanisms by which a metastable could be created in the gas cell, then the logarithm of the ratio would be linear in pressure. This of course is not true, but we did find that the graph of log $[\Delta(p)/\Delta(0)]$ was straight over a larger pressure range [typically (0-140)

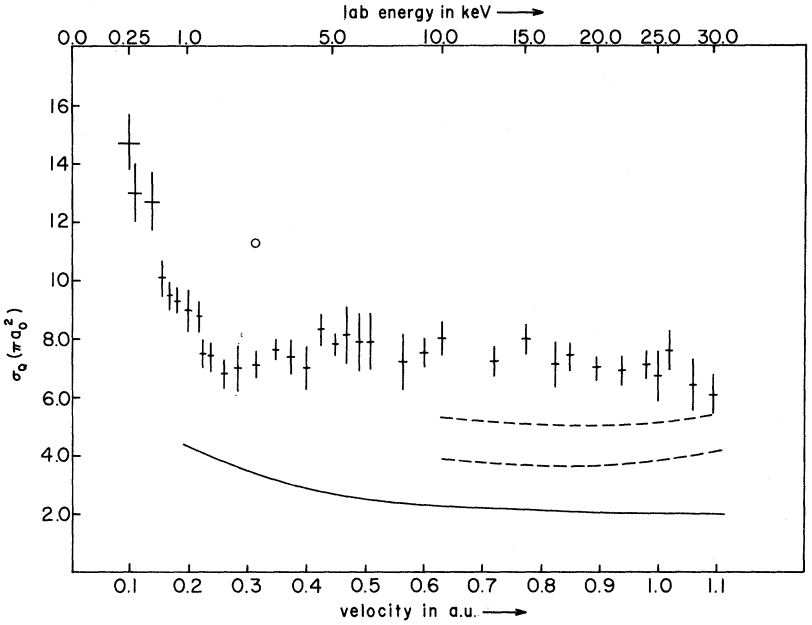

FIG. 2. Cross section for the quenching of metastable hydrogen atoms by helium. The results of Gilbody *et al.* (Ref. 5) lie between the two dotted lines. The single open circle is a measurement of Spiess (Ref. 4). The solid curve was calculated using a helium pseudopotential as described in Sec. IV.

$\times 10^{-5}$ Torr] than was a linear plot. Since $\log(1-x) \approx x$ for $x \ll 1$, the cross sections inferred from linear and logarithmic plots are identical, so we used the logarithmic plot.

There is another quantity with dimensions of a cross section which may be inferred from our data. This is $\sigma_{1s\,2s} I_T/I^*$, where $I_T = I_0 + I^*$, and it may be found from the slope (at zero pressure) of a graph in which $S_0(p)/\Delta(0)$ is plotted against pressure. However, since this quantity depends on I_T/I^*, which is energy dependent, it is of little fundamental interest and we do not present it.

B. Background Gas

A number of assumptions are implicit in Eqs. (1)–(3). One of these is that the background pressure is zero. It may be shown that when Eqs. (1) are modified by the addition of terms which take into account the effect of the background gas in quenching metastables [cross section $(\sigma_Q)_B$] and in

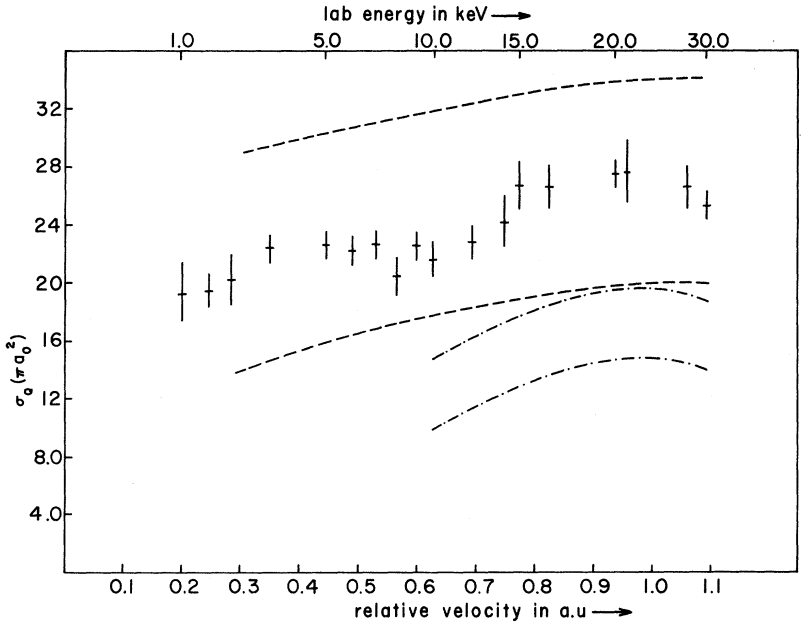

FIG. 3. Cross section for the quenching of metastable hydrogen by argon. The results of Gilbody *et al.* (Ref. 5) lie between the two dot-dashed curves, those of Dose *et al.* (Ref. 6) between the two dashed curves.

exciting atoms from the $1s$ state to the $2s$ state [cross section $(\sigma_{1s\,2s})_B$] then the cross section inferred from Eq. (3) is not $\sigma_Q+\sigma_{1s\,2s}$, but this quantity multiplied by the correction factor $1+n_BL\times(\sigma_Q+\sigma_{1s\,2s})_B$. ($n_B$ is the number density of the background gas and $L=75$ cm is the distance from the prequench plates to the detector.) By neglecting $(\sigma_{1s\,2s})_B$ in comparison with $(\sigma_Q)_B$ and taking the latter to be 3×10^{-15} cm^2 (which is the largest quenching cross section observed by Dose[6] for water vapor), the correction involved was found to be less than 1% and was neglected.

C. Elastic Scattering

The most serious source of uncertainty in our measurements is probably elastic scattering. The geometry of gas cell and detector was such that metastable atoms elastically scattered by less than 18 mrad reached the detector, while those scattered through larger angles hit metal surfaces and hence made the measured value of σ_Q larger than it should be. It was estimated that this effect increased the measured value of σ_Q by less than $0.6\pi a_0^2$ (which is less than the error quoted for σ_Q in Figs. 2 and 3).

In estimating this correction, it was assumed that for angles greater than 10 mrad the differential cross section for elastic scattering of metastables by argon and helium is the same as that due to a potential of the form α/r^s, with α and s being constants. This form has been found to be adequate for many atom-atom collisions at energies of several hundred eV to a few keV.[12] α was estimated from a rough measurement of the differential cross section. By scanning the beam just before it reached the detector, it was found that at 9.0 keV, with helium as target, the differential cross section at 10 mrad was $(1000\pm500)a_0^2$. The cross section was smaller or about the same for other energies, and for argon. (a_0 is the Bohr radius.) By using explicit expressions[12] for the differential cross section as a function of energy, angle, and the parameter s, the correction due to elastic scattering was found to be at most $0.6\pi a_0^2$. (The parameter s was varied from 4 to infinity, the energy from 0.3 to 30 keV.)

Another test of elastic-scattering corrections was made by measuring σ_Q with a detector aperture made smaller by a factor 10. Within our quoted errors, the value of σ_Q was unaffected. However, this is not a reliable test since most of the elastic-scattering correction is due to very-wide-angle scattering. The results presented in Figs. 2 and 3 have not been corrected for elastic scattering.

D. Highly Excited States

We also have implicitly made some assumptions about the possible presence of long-lived highly excited states of hydrogen in the beam. Such states have been observed by Dixon and Harrison,[13] who, however, used a thin cesium target (whereas ours is thick). A sufficiently long-lived atom might decay to the $2p$ state in front of the detector and be counted. We have some evidence that under certain conditions such decays were seen, but the observations were not conclusive. If such long-lived excited states were present in the beam, then the right-hand sides of Eqs. (1) would have to be modified by the addition of terms, which might be pressure dependent. In light of the arguments presented in Sec. II these terms would be the same for both $S(p)$ and $S_0(p)$, thus leaving the difference $\Delta(p)$ unaffected, i.e., the count rate due to long-lived excited states would be independent of prequench voltage and hence would cancel.

E. Results

The measurements described above yield values of $\sigma_Q+\sigma_{1s\,2s}$ of order $8\pi a_0^2$. This is much greater than the maximum value of $0.3\pi a_0^2$ reported by Orbeli *et al.*[14] for $\sigma_{1s\,2s}$ in helium and argon, so we have really measured σ_Q only. The results are presented in Figs. 2 and 3.

The error bars shown are essentially a measure of the reproducibility, i.e., they cover the results of several different measurements made over a period of weeks under various conditions of cesium temperature, beam focus, magnitude of neutral current, etc. The error estimated from the scatter of points in one pressure curve was typically somewhat smaller (by about a factor of 2) than the error bars in Figs. 2 and 3. Both the fluctuations during one run, and from run to run, are believed to be due partially to drifts in beam intensity during one run, and (probably more importantly) due to fluctuations in the ratio I_T/I^* (i.e., the fraction of the total beam that was in the metastable state). The sources of these fluctuations are as yet incompletely understood.

IV. THEORY AND DISCUSSION

In a previous publication,[7] preliminary experimental results were analyzed by using a pseudopotential to replace the interaction of the noble-gas atom with the electron of the incident metastable atom. This pseudopotential was derived from perturbation theory and included, in effect, a Hartree short-range part due to the noble-gas charge cloud and a long-range polarization term. However, Byron and Gersten[9] in analyzing the low-energy quenching of metastables showed that the effect of exchange was important and in order to circumvent difficulties in calculating its effect tried to construct a noble-gas pseudopotential directly from electron noble-gas low-energy elastic scattering. For helium they used a crude δ-function potential based on the scattering length at zero energy. Fortunately

very elaborate pseudopotentials are starting to become available.[15] They have been obtained by fitting low-energy scattering of electrons by noble gases to a potential containing a number of adjustable parameters. The solid curve in Fig. 2 shows the result of using the helium pseudopotential of Ref. 15 in the eikonal formalism of Ref. 7. The "improved" pseudopotential gives results for the quenching cross section which are a factor of 3 or 4 below the experimental results of this paper.

In addition, our experimental results differ from those of Spiess *et al.*[4] and Gilbody *et al.*,[5] as shown in Fig. 2. In their paper, Spiess *et al.* quote no error for their value of $11\pi a_0^2$ at 2.5 keV (open circle on Fig. 2), but state that they are in agreement with an earlier published value[7] of $8\pi a_0^2$.

However, the disagreement between our results and those of Gilbody *et al.* is outside all error bars. We are not able to resolve this discrepancy and can only offer the following comments: There may be an error in measurement of target-gas pressure. We relied on a capacitance manometer which was calibrated by the manufacturer against a dead-weight tester. Gilbody *et al.* effectively used a liquid-nitrogen–trapped McLeod gauge. (Actually they measured target-gas thickness by using charge-exchange cross sections measured by Stier and Barnett.[16] Stier and Barnett measured their gas pressures with a liquid-nitrogen–trapped McLeod gauge.)

The discrepancy might conceivably be due to elastic scattering, which tends to make the measured quenching cross section greater than the true one. We checked on this by making a rough measurement of the differential cross section at one (large) angle and estimated the contribution from angles greater than this (Sec. III C). Gilbody *et al.*[5] do not describe their procedures in detail, but state that an increase in the angular acceptance of their detector produced an insignificant change in recorded signals.

Although it seems quite unlikely that a flux of highly excited hydrogen atoms could affect measurements of quenching cross sections, we did not spend much time systematically looking for such a flux. It may be noted that we used cesium to produce metastables, while Gilbody *et al.* used hydrogen.

From the theoretical point of view, there are several possible reasons for a discrepancy between theory and experiment. First, it may be that pseudopotential methods are simply not adequate for analyzing phenomena which depend on the small distance form of the pseudopotential. (Distances from about 1.5 a.u. to 2.5 a.u. are important in quenching by helium, as may be seen from the experimental results of Fig. 2.) Second, perhaps one is pushing too far the requirement $\Delta E/v < 1$, which is central to the many-body eikonal method. (Here ΔE is the energy difference between "important" states in the hydrogen atom and v is the incident velocity, in a.u.) We feel that it is unlikely that either of the above possibilities could lead to errors as large as a factor of 3 or 4.

Finally, the neglect of the cross-polarization term at small distances may be seriously in error. Roughly speaking, the polarization contribution to the many-body potential is really of the form

$$V_{\rm pol} = -\tfrac{1}{2}\alpha[\vec{R}/R^3 - (\vec{R}-\vec{\rho})/|\vec{R}-\vec{\rho}|^3]^2, \tag{4}$$

where R is the internuclear separation, $\vec{\rho}$ is the internal coordinate of the hydrogen atom, and α is the polarizability of the target (all in a.u.). We have used just the term proportional to $1/|\vec{R}-\vec{\rho}|^4$, suitably cut off. The neglect of the term proportional to $1/R^4$ is certainly correct if one is not concerned with angular distributions, but the cross-term could conceivably play a significant role at small distances, even though it is rather unimportant at intermediate distances.[9] Of course, at large distances this cross term is very important, canceling the leading $1/R^4$ dependence of the direct polarization terms so that the $1/R^6$ van der Waals behavior is obtained.

If this final difficulty is the main one, then it may be that argon, krypton, and xenon are better candidates for a pseudopotential treatment than are helium and neon, since larger distances (of order 4 a.u.) should dominate the problem (for argon, see Fig. 3). Since for these three heavy noble gases the polarizabilities are much larger than in helium and neon, we might expect that at distances like 4 a.u. the polarization term in the pseudopotential will dominate and that the cross term will not be of major importance.[9] If we replace $V_{\rm eff}$ in Eq. (3) of Ref. 7 by just

$$V_{\rm eff} = -\tfrac{1}{2}\alpha|\vec{R}-\vec{\rho}|^{-4}, \tag{5}$$

then by using the optical theorem, the *total* cross section (elastic plus all inelastic) can be evaluated analytically. One obtains

$$\begin{aligned}\sigma_{\rm tot} &= 4\pi a_0^2 \int_0^\infty [1-\cos(\pi\alpha/4vb^3)]\,b\,db \\ &= 3\Gamma(\tfrac{4}{3})(\pi\alpha/4v)^{2/3}\pi a_0^2 \\ &= 2.3(\alpha/v)^{2/3}\pi a_0^2,\end{aligned} \tag{6}$$

where v, α, and b are in a.u. Since the elastic cross section is typically about 10% or less of the total cross section under these conditions, $\sigma_{\rm tot}$ can be taken to be nearly equal to σ_Q, i.e., we write

$$\sigma_Q \simeq 2.3(\alpha/v)^{2/3}\pi a_0^2. \tag{7}$$

There are several circumstances which make a direct application of this result not entirely free of ambiguity. First, for Ar, Kr, and Xe the ener-

gy difference between $H(2s)+N(g)$ and $H(1s)+N(e)$ (where N is a noble gas, g is a ground state, and e is some excited state) is of order of a few volts. Using the ionization potential as a mean excitation energy, the difference ranges from 5.5 eV in argon to 2 eV in xenon. Thus, the adiabatic criterion suggests that for velocities much greater than about 0.2 a.u., i.e., energies in our experiment greater than about 1 keV, such excitation of target with deexcitation of $H(2s)$ will not be hindered significantly. However, Orbeli *et al.*[14] have measured the similar process $H(1s)+N(g)\rightarrow H(2s)+N(e)$ summed over all final states of the noble-gas target and found that in the region where this process is adiabatically unhindered, cross sections are of order $0.3\pi a_0^2$ for Ar, Kr, and Xe. Thus, we expect that in our experiment the process of target excitation plus metastable deexcitation will be at most a few πa_0^2, which is negligible compared to the cross sections we find in argon.

Perhaps more important, however, is the process $H(2s)+N(g)\rightarrow H(e)+N(e)$, where $H(e)$ represents any of the "nearby" excited states of hydrogen ($2p$, $3s$, $3p$, $3d$, ...) and $N(e)$ represents any excited state of the noble-gas target. In argon, for example, the energy difference between typical initial and final states is about 0.7 a.u., whereas in xenon it is about 0.5 a.u. This means that for incident velocities above about 0.5 a.u. (i.e., incident energies of about 6 keV) the whole range of quenching with target excitation will come rapidly into play.

On the other hand, if one goes to much lower values of v, one must be careful not to go to such low values that the adiabatic criterion excludes a large number of important hydrogen excited states (the target remaining in the ground state). For a v of 0.3–0.4 a.u. all of the hydrogen bound states and the low continuum are accessible. To see if the formula of Eq. (7) gives a sensible result here, let us take $v=0.4$ a.u. (incident energy of 4 keV). For argon, $\alpha=11$ a.u., so $\sigma_Q\simeq 21\pi a_0^2$, which is in good agreement with our observations. More to the point than numerical agreement, however, is the fact that this result indicates a sensible magnitude. Equation (7) also suggests that in this general energy region, Kr should give a quenching cross section about 30% larger, while Xe should give a result about 60% larger. Below $v\simeq 0.4$, Eq. (7) will predict results which are too large, and above $v=0.4$, Eq. (7) will give results which are too small.

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[1]W. L. Fite, R. T. Brackmann, D. G. Hummer, and R. F. Stebbings, Phys. Rev. 116, 363 (1959).

[2]F. J. Comes and U. Wenning, Z. Naturforsch. 24, 587 (1969).

[3]R. S. Kass and W. L. Williams, Phys. Rev. Letters 27, 473 (1971).

[4]G. Spiess, A. Valance, and P. Pradel, in *Abstracts of Papers of the VII International Conference on the Physics of Electronic and Atomic Collisions* (North-Holland, Amsterdam, 1971), p. 1083.

[5]H. B. Gilbody, R. Browning, R. M. Reynolds, and G. I. Riddell, J. Phys. B 4, 94 (1971).

[6]V. Dose, V. Meyer, and M. Salzmann, J. Phys. B 2, 1357 (1969).

[7]F. W. Byron, Jr., R. V. Krotkov, and J. A. Medeiros, Phys. Rev. Letters 24, 83 (1970).

[8]J. I. Gersten, J. Chem. Phys. 51, 637 (1969).

[9]F. W. Byron, Jr. and J. I. Gersten, Phys. Rev. A 3, 620 (1971).

[10]W. M. Howard, Phys. Fluids 4, 521 (1961).

[11]R. T. Brackmann, W. L. Fite, and K. E. Hagen, Rev. Sci. Instr. 29, 125 (1958).

[12]I. Amdur and J. E. Jordan, Advan. Chem. Phys. 10, 29 (1966).

[13]A. J. Dixon and M. F. A. Harrison, in Ref. 4, p. p. 892.

[14]A. L. Orbeli, E. P. Andreev, V. A. Ankudinov, and V. M. Dukelskii, Zh. Eksperim. i Teor. Fiz. 57, 108 (1969). [Sov. Phys. JETP 30, 63 (1970)].

[15]C. Bottcher, J. Phys. B 4, 1140 (1971).

[16]P. M. Stier and C. F. Barnett, Phys. Rev. 103, 896 (1956).