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Survival Probability in Dissociative Attachment*

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The survival probability in dissociative attachment is investigated with special attention to the (e,H_2) system. It is shown that the simple expression for the dissociative-attachment cross section, as given by the product of a capture cross section and a survival probability, is equivalent to the s-wave approximation for the $g\rightarrow g$ dissociative attachment. This expression, however, does not constitute an approximation for the $g\rightarrow u$ dissociative attachment, since the parity of the initial rotational states of H_2 is always opposite to that of the relative angular momentum states of H and H^- and the capture cross section appearing in the simple expression is identically zero. According to the Kronig selection rules and the symmetry requirements, only odd partial waves of the incident electron may contribute to the $g\rightarrow u$ dissociative attachment in the (e,H_2) system. Consequently, the lowest contributing partial wave is not the s wave but the p wave of the incident electron. This, then, destroys the simple proportional dependence of the cross section on the survival probability. However, one may still express the cross section as a sum of products of a capture cross section and a survival probability for the various contributing angular momentum states of the constituent nuclei. The dependence of the survival probability on the angular momentum states of the constituent nuclei is also investigated for the (e,H_2) system. It is observed that for the $g\rightarrow u$ dissociative attachment the survival probability depends strongly on the angular momentum states. This arises because the $g\rightarrow u$ dissociative attachment occurs at such a low energy that variations in the centrifugal barrier become comparable with the breakup energy of the constituent atoms. This then suggests a strong temperature dependence for the $g\rightarrow u$ dissociative attachment in the (e,H_2) system. For the $g\rightarrow g$ dissociative attachment, such dependence is much weaker since here the process becomes significant at a somewhat higher energy and the variation in centrifugal energy is overshadowed by the large break-up energy of the constituent atoms. The validity of the commonly adopted approximation for survival probability (involving the auto-ionization width and relative velocity of the nuclei) is also examined.

I. INTRODUCTION

In 1951 Holstein[1] suggested that the cross section for dissociative attachment may be represented as

$$\sigma=\sigma^{(c)}p\,, \tag{1.1}$$

where $\sigma^{(c)}$ is the cross section for capturing of the incident electron by the target molecule AB into a repulsive state of the molecular ion AB^- at a fixed nuclear configuration and, p is the survival probability that the electron will remain captured while the molecular ion is dissociating in the repulsive field. From this intuitive picture Holstein deduced that the survival probability is given by the expression

$$p=e^{-\Gamma\tau/\hbar}\,, \tag{1.2}$$

where Γ is the auto-ionization width of the molecular ion state in question and τ is the time required for the constituent nuclei to move apart at a distance greater than R_c (Fig. 1). It was later shown by Bardsley, Herzenberg, and Mandl[2] that within the framework of the Born-Oppenheimer separation approximation, and with the molecular rotation neglected, the cross section for dissociative attachment may indeed be written in the form given by Eq. (1.1) with the survival probability given by

$$p=\exp\left\{-\lim_{R\to\infty}\int_{R_0}^{R}\frac{\Gamma(R')dR'}{v(R')}\right\}$$
$$=\exp\left\{-\lim_{R\to\infty}\int_{t(R_0)}^{t(R)}\Gamma[t(R')]dt(R')\right\}\,, \tag{1.3}$$

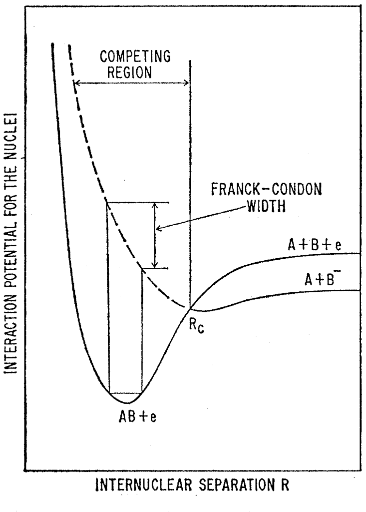

FIG. 1. Schematic diagram of interaction potentials for dissociative attachment of the Franck-Condon type. The dashed curve represents the real part of the interaction potential. For simplicity, the splitting of the interaction potential due to the local electric field along the internuclear axis is not shown here.

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[1] T. Holstein, Phys. Rev. 84, 1073 (1951).

[2] J. N. Bardsley, A. Herzenberg, and F. Mandl, in *Atomic Collision Processes*, edited by M. R. C. McDowell (North-Holland Publishing Company, Amsterdam, 1964), p. 415.

where R_0 is the classical turning point and $v(R)$ is the relative velocity of the nuclei at a separation distance R. This confirms the approximate validity of the expression for survival probability as given by Eq. (1.2). Recently, the simple expression for the cross section [Eq. (1.1)] has been used for studying the isotope effect[3] and for estimating the width[4] with the survival probability given either by Eq. (1.3) or (1.2).

In this paper we show that the initial molecular rotational states which were neglected in deriving Eq. (1.1) are of importance in dissociative attachment for the (e,H_2) system considered here. This importance arises in two specific ways, namely through the dependence of the survival probability on the relative angular momentum states of the constituent nuclei[5] and through the Kronig selection rules[6] between participating states of $AB+e$ and $A+B^-$. We will show in Sec. II that the simple product expression for the cross section [Eq. (1.1)] with p given by Eq. (2.18) constitutes an approximation only for the *gerade*-to-*gerade* $(g\rightarrow g)$ dissociative attachment of the s-wave electrons. For the *gerade*-to-*ungerade* $(g\rightarrow u)$ dissociative attachment this approximation is identically zero since according to the Kronig selection rules and the symmetry requirements only odd partial waves of the incident electron may contribute in this case. Consequently, the lowest contributing partial wave is the p wave. This then destroys the simple proportional dependence of the cross section on the survival probability as given by Eq. (1.1).

It has been observed experimentally that H^- ions in the scattering system (e,H_2) first appear at electron energies around 3.75 eV, near the theoretical threshold for the production of H^- ions.[7] At higher energies, around 10 eV and above, further peaks with greater magnitude are observed in the cross section for H^- production.[8] The first peak around 3.75 eV has been attributed to the $g\rightarrow u$ dissociative attachment involving the lowest auto-ionization ${}^2\Sigma_u{}^+$ state of $H_2{}^-$. The peak at energies around 10 eV comes from the $g\rightarrow g$ dissociative attachment due to the lowest auto-ionization ${}^2\Sigma_g{}^+$ state of $H_2{}^-$. Since the $g\rightarrow u$ dissociative attachment occurs at such low energy, one would then expect that the variation in the centrifugal barrier associated with the relative partial waves of H and H^- would become significant for the imaginary parts of their radial phase shift. This implies that the survival probability, being exponentially dependent on the imaginary parts of the phase shift, may become strongly dependent on the relative angular momentum of the constituent nuclei. For the $g\rightarrow g$ dissociative attachment, on the other hand, the J dependence on the survival probability can be quite different since here the process becomes significant at somewhat higher energy. The variation in centrifugal energy is overshadowed by the large breakup energy of the constituent atoms. One would then expect a weaker J dependence of the survival probability.

A numerical study of the survival probability is carried out for the (e,H_2) system at electron energies near the threshold for the $g\rightarrow u$ dissociative attachment and at somewhat higher energies for the $g\rightarrow g$ dissociative attachment, using an expression for the survival probability given in Sec. II. Indeed, it is observed that for the $g\rightarrow u$ case the survival probability depends significantly on the angular momentum states of the constituent nuclei, and for the $g\rightarrow g$ case this J dependence is much weaker. This then suggests a strong temperature dependence for the $g\rightarrow u$ dissociative attachment in the (e,H_2) system just due to the initial distribution of rotational states. Of course, the initial distribution of vibrational states also contributes to the temperature dependence. Unfortunately, no measurement on the temperature dependence has been carried out for this system. Recently, temperature dependence of dissociative attachment has been measured[9] for the (e,O_2) system where one observes a strong temperature dependence in which the threshold is shifted to lower energy and the cross section itself is enhanced as temperature increases. It should be noted that recently O'Malley[10] has, in a parametrization study, reached the conclusion that this behavior for the (e,O_2) system can be explained as due to the initial distribution of vibrational states.

It is also observed from the present study that the approximate expression [Eq. (1.2)] for the survival probability is not valid in this case for either the study of isotope effect or the estimation of width. This is simply because the intuitive expression for the survival probability is valid only for cases where

$$\mu v(R)^2 > \Gamma(R) \tag{1.4}$$

as also pointed out by Bardsley *et al.*[2] If the criterion given by Eq. (1.4) is not met, the use of Eq. (1.2) or Eq. (1.3) for studying the isotope effect or estimating the width[3,4] can be very misleading. In addition, there should be a factor in the survival probability which comes from the imaginary part of the interaction potential at the classical turning point. This factor, which depends critically on the differences of the slopes

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[3] Yu. N. Demkov, Phys. Letters **15**, 235 (1965).

[4] G. J. Schulz and R. K. Asundi, Abstract of the 19th Gaseous Electronic Conference, Atlanta, Georgia, 1966, p. 41 (unpublished).

[5] J. C. Y. Chen, Phys. Rev. **156**, 12 (1967).

[6] G. Herzberg, *Spectra of Diatomic Molecules* (D. Van Nostrand Company, Inc., Princeton, New Jersey, 1950), 2nd ed., p. 416.

[7] G. J. Schulz and R. K. Asundi, Phys. Rev. Letters **15**, 946 (1965).

[8] See, for example, D. Rapp, T. E. Sharp, and D. D. Briglia, Phys. Rev. Letters **14**, 533 (1965); G. J. Schulz, Phys. Rev. **113**, 816 (1959).

[9] W. L. Fite, R. T. Brachman, and U. R. Henderson, in *Proceedings of the Fourth International Conference on the Physics of Electronic and Atomic Collisions, Quebec, 1965* (Science Bookcrafters, Hastings-on-Hudson, New York, 1965), p. 100; R. F. Stebbings, M. A. Fineman, J. W. McGowan, B. A. Turner, and F. A. Wolf, Defense Atomic Support Agency Report No. DASA-GA-6699, 1965 (unpublished).

[10] T. F. O'Malley, Phys. Rev. **155**, 59 (1967).

of the real and imaginary parts of the interaction potential at the classical turning point, is usually different from unity.

II. EXPRESSION FOR THE SURVIVAL PROBABILITY

In the Born-Oppenheimer separation approximation, it can be shown that the strong coupling between the electronic motion in the scattering channel and dissociative-attachment channel results in a complex potential, $V(\mathbf{R})-\frac{1}{2}i\Gamma(\mathbf{R})$, for the constituent nuclei.[5] The nuclear motion is described by the equation

$$\{K_{\mathbf{R}}+V(\mathbf{R})-\tfrac{1}{2}i\Gamma(\mathbf{R})\}\Xi(\mathbf{R})=E\Xi(\mathbf{R}), \qquad (2.1)$$

where $K_{\mathbf{R}}$ is the relative kinetic-energy operator of the nuclei. The same equation can also be derived[2,11] by considering the dissociating negative molecular-ion state as a resonance state for a fixed nuclear configuration in the open-channel segment of the Hilbert space. Since the potential is non-Hermitian, no source term in Eq. (2.1) is necessary.

For dissociative attachment in the (e,H_2) system near the threshold, the relevant negative-ion state is the $^2\Sigma_u{}^+$ state of $\mathrm{H}_2{}^-$. It has been found recently that $\Gamma(R)$ for the $^2\Sigma_u{}^+$ state of $\mathrm{H}_2{}^-$ is very large[12] ($\Gamma\cong4.5$ eV in the Franck-Condon region of the ground state of the H_2 molecule). As a consequence, it is no longer constructive to consider the nonstationary character [measured by $\Gamma(R)$] to be small so that $V(R)$ may be treated as a stationary potential curve for describing the nuclear motion.[5] To treat the problem properly we must consider both $V(R)$ and $\Gamma(R)$ with equal footing.

After angular decomposition, Eq. (2.1) may be rewritten as a pair of coupled equations (in atomic units)

$$\left\{\frac{d^2}{dR^2}-\frac{J(J+1)}{R^2}+2\mu[E-V(R)]\right\}\eta_J(R)=\mu\Gamma(R)\zeta_J(R), \qquad (2.2a)$$

$$\left\{\frac{d^2}{dR^2}-\frac{J(J+1)}{R^2}+2\mu[E-V(R)]\right\}\zeta_J(R)=-\mu\Gamma(R)\eta_J(R), \qquad (2.2b)$$

where μ is the reduced mass and where we have assumed that the complex potential is a function of radial coordinate R only. For a continuum solution of Eq. (2.1), we impose the boundary conditions that the solutions, η_J and ζ_J, vanish at $R=0$ and have the asymptotic forms

$$\eta_J(R)\underset{R\to\infty}{\longrightarrow}k^{-1}\cosh\delta_J{}^{(2)}\sin(kR-\tfrac{1}{2}J\pi+\delta_J{}^{(1)}), \qquad (2.3a)$$

$$\zeta_J(R)\underset{R\to\infty}{\longrightarrow}k^{-1}\sinh\delta_J{}^{(2)}\cos(kR-\tfrac{1}{2}J\pi+\delta_J{}^{(1)}), \qquad (2.3b)$$

$$k=\{2\mu[E_e-D_0+E_a+\epsilon_v+\epsilon_L]\}^{1/2}=(2\mu E)^{1/2}, \qquad (2.4)$$

where k is the wave number of the relative motion of H and H^-, E_e is the incident electron energy, D_0 is the dissociation energy of H_2, E_a is the electron affinity of H, and ϵ_v and ϵ_L are, respectively, the initial vibrational and rotational energies of H_2. Clearly Eq. (2.3) implies that

$$\xi_J(R)=\eta_J(R)+i\zeta_J(R)\underset{R\to\infty}{\longrightarrow}k^{-1}\sin(kR-\tfrac{1}{2}J\pi+\delta_J), \qquad (2.5)$$

where

$$\delta_J=\delta_J{}^{(1)}+i\delta_J{}^{(2)} \qquad (2.6)$$

is the complex phase shift and $\xi_J(R)$ is the radial part of $\Xi(\mathbf{R})$. Note that here we deal with Σ states; thus the angular states are properly described by the simple rotator wave functions.

Since $V(R)$ is a slowly varying potential, i.e.,

$$\frac{1}{k}\frac{d\ln V}{dR}\ll1, \qquad (2.7)$$

the nuclear motion can be described semiclassically. We solve Eq. (2.2) by the JWKB approximation.[13] The proper solution for $\xi_J(R)$ at the right-hand side of the turning point R_0 is[5]

$$\xi_J(R)=\kappa_J(R)^{-1/2}\sin\left\{\tfrac{1}{4}\pi-\mathfrak{D}_J{}^{(+)}(R_0)+i\mathfrak{D}_J{}^{(-)}(R_0)+\int_{R_0}^{R}\kappa_J(R')dR'\right\}, \qquad (2.8)$$

with

$$\kappa_J(R)=\{2\mu[E-\mathfrak{U}_J(R)+i\tfrac{1}{2}\Gamma(R)]\}^{1/2}, \qquad (2.9)$$

$$\mathfrak{U}_J(R)=V(R)+(J+\tfrac{1}{2})^2/(2\mu R^2), \qquad (2.10)$$

$$\mathfrak{D}_J{}^{(\pm)}(R_0)=-\frac{(2\mu)^{1/2}}{6}\Gamma(R_0)^{3/2}\left\{\left(\frac{d\mathfrak{U}_J}{dR}\pm\frac{1}{2}\frac{d\Gamma}{dR}\right)\Big/\left[\left(\frac{d\mathfrak{U}_J}{dR}\right)^2+\frac{1}{4}\left(\frac{d\Gamma}{dR}\right)^2\right]\right\}_{R=R_0}, \qquad (2.11)$$

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[11] T. F. O'Malley, Phys. Rev. **150**, 14 (1966). It should be noted that the result derived in this reference does not apply to dissociative attachment for homonuclear diatomic molecules A_2, in which the *gerade* (or *ungerade*) system of the initial Σ target state is not conserved into the dissociative-attachment channel. This is because the parity of the rotational state L of the target molecule A_2 is always opposite to that of the relative angular-momentum state J of A and A^-. Thus the δ function δ_{LJ} which is used in deriving the results can never be unity and consequently the expression for the T matrix [Eq. (5.9b) of the reference] is identically zero; see also J. C. Y. Chen [Phys. Rev. **148**, 66 (1966)] for the discussion on the fixed-nuclear-configuration approximation.

[12] J. N. Bardsley, A. Herzenberg, and F. Mandl, Proc. Phys. Soc. (London) **89**, 305 (1966), Figs. 3 and 4, curves labeled A, and Figs. 9 and 10.

[13] See, for example, N. F. Mott and H. S. W. Massey, *The Theory of Atomic Collisions* (Clarendon Press, Oxford, England, 1965), 3rd ed.; R. E. Langer, Phys. Rev. **51**, 669 (1937).

where $\mathfrak{D}_J^{(\pm)}$ arises from the imaginary part of the potential in matching with the Airy solution at the turning point R_0. Note that $\mathfrak{D}_J^{(\pm)}(R_0)$ is implicitly energy-dependent since the classical turning point R_0 is energy-dependent. The phase shift can now be obtained from the asymptotic behavior of Eq. (2.8):

$$\delta_J^{(1)}=(J+\tfrac{1}{2})\tfrac{1}{2}\pi-kR_0+\mathfrak{D}_J^{(+)}(R_0)+\lim_{R\to\infty}\int_{R_0}^{R}\mathrm{Re}[\kappa_J(R')-k]dR', \quad (2.12a)$$

$$\delta_J^{(2)}=\mathfrak{D}_J^{(-)}(R_0)+\lim_{R\to\infty}\int_{R_0}^{R}\mathrm{Im}[\kappa_J(R')]dR'. \quad (2.12b)$$

If we expand $\kappa_J(R)$ assuming $\Gamma(R)$ is small, we obtain

$$\delta_J^{(2)}=\mathfrak{D}_J^{(-)}(R_0)+\lim_{R\to\infty}\frac{1}{2}\int_{R_0}^{R}\frac{\Gamma(R')dR'}{v_J(R')} \quad (2.13)$$

with

$$v_J(R)=\{(2/\mu)[E-V(R)]-[(J+\tfrac{1}{2})/(\mu R)]^2\}^{1/2}, \quad (2.14)$$

where $v_J(R)$ is the relative velocity of the nuclei at separation R. The magnitude of $\mathfrak{D}_J^{(-)}$ depends strongly on the differences in the slopes of the real and imaginary parts of the potential at the classical turning point R_0. Essentially, the same results have been derived before neglecting the J dependence.[2] They are given here to establish our notation and to demonstrate the explicit J dependence.

For given initial vibrational and rotational states, v and L, of the target molecule, the cross section for dissociative attachment is given by

$$\sigma_{vL}=\frac{1}{2L+1}\sum_{M_L}\int|\sum_J \alpha_{J,vLM_L}e^{i\delta_J}|^2 d\Omega_f. \quad (2.15)$$

Now, if the multipoles generated by the electronic distribution are neglected in the effective potential for the nuclear motion, the integration over nucelar angular states in α_{J,vLM_L} results in a δ function $\hat{\delta}_{JL}$. The cross section then takes the form[2,11]

$$\sigma_{vL}\cong\sigma_{L,vL}^{(c)}P_L(g\to g) \quad (2.16)$$

with

$$P_L(g\to g)=p_L, \quad (2.17)$$

where $\sigma_{L,vL}^{(c)}$ is defined by Eq. (2.16) and may be interpreted as the capture cross section and p_L is the survival probability

$$p_L=\exp\{-2\delta_L^{(2)}\}. \quad (2.18)$$

Equations (2.16) and (2.17) imply that to calculate the survival probability for a given rotational state L of the target one needs only to calculate the corresponding imaginary part of the phase shift [Eq. (2.12b)].

The approximate expression for the cross section given by Eq. (2.16) is equivalent to the s-wave approximation for the incident electron, since more explicitly α_{J,vLM_L} in Eq. (2.15) can be written for the present case as

$$\alpha_{J,vLM_L}=\langle\xi_J(R)|\lambda_{JLM_L}(k_i,R,\Omega_f)|\chi_v(R)\rangle \quad (2.19)$$

with

$$\lambda_{JLM_L}=2\left(\frac{\mu k}{k_i}\right)^{1/2}\times\sum_{l,m}\sum_{M_J}i^{-J}\langle Y_{JM_J}(\hat{R})|Y_{lm}(\hat{R})|Y_{LM_L}(\hat{R})\rangle\times Y_{JM_J}(\Omega_f)\lambda_l'(R)Y_{lm}^*(\Omega_i), \quad (2.20)$$

where k_i and l are the wave number and angular momentum of the incident electron, $\lambda_l'(R)$ is the electronic matrix element, χ_v is the initial vibrational wave function, and Ω_i is the incident direction. For s-wave electrons $l=0$, integration over $\hat{R}$ of the spherical harmonics in Eq. (2.19) results in a δ function $\hat{\delta}_{JL}$ and, consequently, Eq. (2.16). The s-wave approximation does not, however, in general, satisfy the Kronig selection rules.[6] In particular, we have the selection rule

$$\Delta K=0, \quad (2.21)$$

where K is the total orbital angular momentum of the system. In the present case $\Delta\Lambda=0$; thus Eq. (2.21) implies that the relation

$$|L-l|\leq J\leq L+l \quad (2.22)$$

must be satisfied.

The target molecule H_2 is initially in a *gerade* electronic state, and the rotational states L can be either even or odd, depending on whether the constituent protons are in singlet or triplet nuclear spin states. On the other hand, the electronic state for the dissociative-attachment channel may have an *ungerade* symmetry. Consequently, the parity for the angular momentum states of H and H^- is just opposite to that of the rotational states of H_2; namely, J is odd for singlet and even for triplet nuclear spin states. Thus, independent of nuclear spin multiplicities, J and L always have opposite parity. Consequently, from the selection rule [Eq. (2.22)], only odd l's are allowed. This implies that if the target molecule is in a *gerade* state, s-wave electrons cannot give rise to dissociative attachment in the *ungerade* mode. The approximate expression for the cross section [Eq. (2.16)] is then identically zero. This is also apparent from the δ function $\hat{\delta}_{JL}$, since L and J always have opposite parity. The above conclusions apply to all similar dissociative attachment (as well as dissociative recombination) processes involving homonuclear molecules, independent of whether the nuclei are fermions or bosons. If they are bosons, the parity of the angular states in relation to the nuclear spin symmetry is just reversed from that for fermions discussed above.

The fact that L and J always have opposite parity nevertheless remains.

Equation (2.16), however, constitutes an approximation for dissociative attachment in which the *gerade* (or *ungerade*) symmetry of the target molecule (not including the symmetry of the incident electron) is conserved into the dissociative-attachment channel. In this case, the incident electron must have *gerade* symmetry for the *gerade* (or *ungerade*) target molecule.

For the case in which the *gerade* symmetry of the target ${}^1\Sigma_g{}^+$ state of H_2 is not conserved[14] into the dissociative-attachment channel (which is represented by the ${}^2\Sigma_u{}^+$ state of $H_2{}^-$), the lowest contributing partial wave of the incident electron is the p wave. Taking $l=1$, the selection rule given by Eq. (2.22) allows J to take up the two values $L+1$ and $L-1$ ($J=L$ is eliminated by their parities). The cross section for dissociative attachment is then given by the approximation

$$\sigma_{vL} \cong \sigma_{L+1,vL}{}^{(c)} C_{L+1} e^{-2\delta_{L+1}{}^{(2)}} + \sigma_{L-1,vL}{}^{(c)} C_{L-1} e^{-2\delta_{L-1}{}^{(2)}}, \quad (2.23)$$

where the $C_{L\pm 1}$'s are related to the Clebsh-Gordan coefficients

$$C_{L\pm 1} = [2(L\pm 1)+1] \begin{pmatrix} L\pm 1 & 1 & L \\ 0 & 0 & 0 \end{pmatrix}^2 . \quad (2.24)$$

Equation (2.23) may also be written in the form given by Eq. (2.16) under certain conditions. If the classical turning point R_0 does not change appreciably with J for ξ_J, we may replace $\langle \xi_{L\pm 1}(R) | \lambda_1'(R) | \chi_v(R) \rangle$ by $\langle \xi_L(R) | \lambda_1'(R) | \chi_v(R) \rangle$ and obtain from Eq. (2.24)

$$\sigma_{vL} \cong \sigma_{L,vL}{}^{(c)} P_L(g \rightarrow u) \quad (2.25)$$

with $P_L(g \rightarrow u)$ given in terms of the survival probabilities

$$P_L(g \rightarrow u) = C_{L+1} p_{L+1} + C_{L-1} p_{L-1}. \quad (2.26)$$

Note that for the case $L=0$, Eqs. (2.23) and (2.26) reduce to one term since by the selection rule [Eq. (2.22)], the $L-1$ term is not allowed.

III. RESULTS FOR THE SURVIVAL PROBABILITY

The survival probability $p_L = \exp\{-2\delta_L{}^{(2)}\}$ with $\delta_L{}^{(2)}$ given by Eq. (2.12b) is evaluated numerically for the (e,H_2) system using the recently obtained values for $V(R)$ and $\Gamma(R)$ as a function of R.[12] The calculated survival probability in the $g \rightarrow u$ dissociative attachment is plotted in Fig. 2 as a function of the initial rotational state L for H_2 and its various isotopes for the electron energy near threshold. One of the important features of the result is the strong dependence of the survival probability on the rotational state L. Examining Fig. 2 it is seen that variation in p_L can be easily of one order of magnitude. The L dependence of survival probability comes, of course, from the J dependence of the imaginary phase shift $\delta_J{}^{(2)}$. Examining Eqs. (2.9), (2.10), and (2.12b), it is apparent that the J dependence is most pronounced at energies where E is comparable with the centrifugal energy, $(J+\frac{1}{2})^2/2\mu R^2$, for R close to the classical turning point R_0. For the present case, the $g \rightarrow u$ dissociative-attachment cross section has a peak near the threshold energy and thus the energy E for the relative motion of H and H^- is small. The centrifugal energy is comparable to the relative kinetic energy E and consequently the J dependence is strong. For the the case of $g \rightarrow g$ dissociative attachment, the J dependence of the survival probability is less pronounced (see Fig. 3), since here the energy is large in comparison with the centrifugal energy and the J dependence is somewhat overshadowed. However, the dependence can be significant if the relative kinetic energy of the constituent nuclei is small.

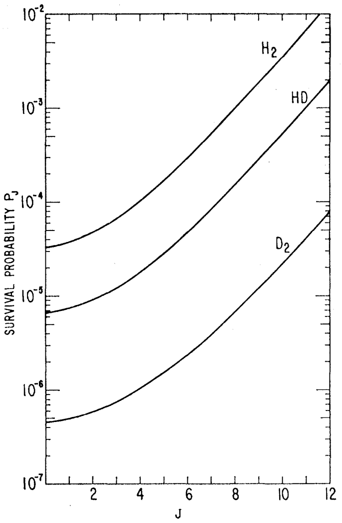

FIG. 2. Dependence of the survival probability on the nuclear angular momentum state L for the $g \rightarrow u$ dissociative attachment of H_2, HD, and D_2 at an incident electron energy of 3.775 eV.

We have tabulated in Tables I and II the calculated values of p_L for several values of L using Eqs. (1.3) and (2.18) with the imaginary phase shift given by Eq. (2.12b). It is observed that, for the $g \rightarrow u$ case, the approximation given by Eq. (1.3) underestimates the survival probability easily by two orders of magnitude. For the $g \rightarrow g$ case, this approximation is reasonable. Assuming for the moment that the mass dependence in the capture cross section can be neglected, the ratio of cross sections for different isotopes is given simply for each L by the ratio of survival probabilities; for example, in the $g \rightarrow u$ case,

$$\frac{\sigma_{vL}(E,H_2)}{\sigma_{vL}(E,HD)} \cong C_{L+1}\frac{p_{L+1}H_2}{p_L HD} + C_{L-1}\frac{p_{L-1}H_2}{p_L HD} . \quad (3.1)$$

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[14] The *gerade* or *ungerade* symmetry of the total initial system, i.e., the target H_2 molecule plus the incident electron should, of course, be conserved into the dissociative-attachment channel, $H+H^-$.

TABLE I. Survival probability in dissociative attachment of H_2 and its isotopes for the $g \rightarrow u$ case.

	H_2		HD		D_2		HT		T_2	
	JWKB[a]	Γ/v[b]	JWKB[a]	Γ/v[b]	JWKB[a]	Γ/v[b]	JWKB[a]	Γ/v[b]	JWKB[a]	Γ/v[b]
0	3.292×10^{-5}	5.547×10^{-8}	6.649×10^{-6}	4.179×10^{-9}	4.555×10^{-7}	5.477×10^{-11}	3.229×10^{-6}	1.300×10^{-9}	1.730×10^{-8}	2.762×10^{-13}
1	3.706×10^{-5}	6.148×10^{-8}	7.370×10^{-6}	4.568×10^{-9}	4.956×10^{-7}	5.891×10^{-11}	3.558×10^{-6}	1.414×10^{-9}	1.853×10^{-8}	2.932×10^{-13}
2	4.676×10^{-5}	7.543×10^{-8}	9.027×10^{-6}	5.456×10^{-9}	5.856×10^{-7}	6.812×10^{-11}	4.310×10^{-6}	1.671×10^{-9}	2.125×10^{-8}	3.302×10^{-13}
3	6.549×10^{-5}	1.022×10^{-7}	1.214×10^{-5}	7.105×10^{-9}	7.489×10^{-7}	8.461×10^{-11}	5.708×10^{-6}	2.145×10^{-9}	2.604×10^{-8}	3.944×10^{-13}
4	1.005×10^{-4}	1.524×10^{-7}	1.778×10^{-5}	1.007×10^{-8}	1.031×10^{-6}	1.127×10^{-10}	8.205×10^{-6}	2.983×10^{-9}	3.399×10^{-8}	4.993×10^{-13}
5	1.664×10^{-4}	2.494×10^{-7}	2.803×10^{-5}	1.549×10^{-8}	1.520×10^{-6}	1.609×10^{-10}	1.268×10^{-5}	4.484×10^{-9}	4.710×10^{-8}	6.692×10^{-13}
6	2.922×10^{-4}	4.459×10^{-7}	4.702×10^{-5}	2.580×10^{-8}	2.381×10^{-6}	2.456×10^{-10}	2.086×10^{-5}	7.270×10^{-9}	6.901×10^{-8}	9.489×10^{-13}
7	5.353×10^{-4}	8.686×10^{-7}	8.285×10^{-5}	4.640×10^{-8}	3.931×10^{-6}	4.000×10^{-10}	3.610×10^{-5}	1.268×10^{-8}	1.064×10^{-7}	1.422×10^{-12}
8	1.006×10^{-3}	1.837×10^{-6}	1.514×10^{-4}	8.984×10^{-8}	6.781×10^{-6}	6.943×10^{-10}	6.499×10^{-5}	2.375×10^{-8}	1.715×10^{-7}	2.248×10^{-12}
9	1.913×10^{-3}	4.208×10^{-6}	2.835×10^{-4}	1.869×10^{-7}	1.212×10^{-5}	1.282×10^{-9}	1.204×10^{-4}	4.764×10^{-8}	2.880×10^{-7}	3.745×10^{-12}
10	3.637×10^{-3}	1.040×10^{-5}	5.379×10^{-4}	4.168×10^{-7}	2.227×10^{-5}	2.512×10^{-9}	2.270×10^{-4}	1.021×10^{-7}	5.002×10^{-7}	6.571×10^{-12}
11	6.849×10^{-3}	2.769×10^{-5}	1.024×10^{-3}	9.943×10^{-7}	4.169×10^{-5}	5.220×10^{-9}	4.316×10^{-4}	2.336×10^{-7}	8.940×10^{-7}	1.212×10^{-11}
12	1.270×10^{-2}	7.901×10^{-5}	1.942×10^{-3}	2.532×10^{-6}	7.896×10^{-5}	1.148×10^{-8}	8.213×10^{-4}	5.691×10^{-7}	1.635×10^{-6}	2.349×10^{-11}

[a] Calculated according to Eqs. (2.18) and (2.12b) at an incident electron energy of 3.775 eV.
[b] Calculated according to Eq. (1.3) at an incident electron energy of 3.775 eV.

In Table III, the isotope effect as given by the ratio formula is tabulated for the $g \rightarrow u$ case, for several values of L using Eqs. (1.3) and (2.18) with the imaginary phase shift given by Eq. (2.12b). As expected, Eq. (1.3) predicts a quite different isotope effect from that predicted by using Eq. (2.12b) for the phase shift. This then demonstrates that the intuitive expressions [Eqs. (1.1) and (1.2)] are not valid for studying the observed isotope effect[3] unless the criterion Eq. (1.4) is satisfied. In addition, there is the problem of thermal averaging since the measured cross section is an averaged quantity over the initial distribution of vibrational and rotational states. One expects that the strong L dependence of the survival probability in the $g \rightarrow u$ case would have a profound effect on the temperature dependence of the cross section.

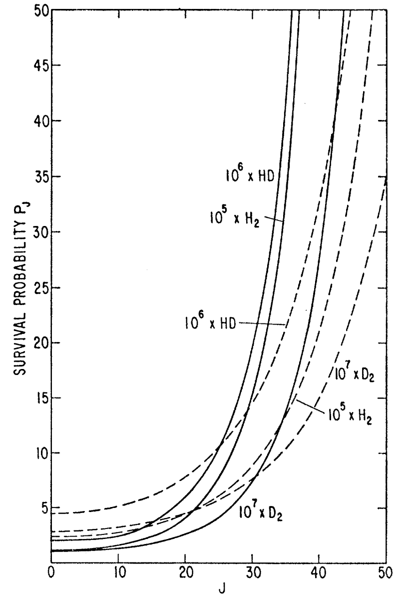

FIG. 3. Dependence of the survival probability on the nuclear angular momentum state L for the $g \rightarrow g$ dissociative attachment of H_2, HD, and D_2. The solid curves and dashed curves are for incident electron energies of 7.0 and 10.2 eV, respectively.

Assuming that the experimental measurement of the cross section for dissociative attachment is carried out under thermal equilibrium conditions, the thermal-averaged cross section is

$$\sigma(T,E)=Z^{-1}\sum_{v}\sum_{L}{}' \, 2(2L+1)\sigma_{vL}(E)e^{-(\epsilon_v+\epsilon_L)/KT}, \qquad (3.2)$$

where Z^{-1} is the partition function of the target molecule which normalizes the thermal distribution function and where the prime on the sum over L indicates a restriction in the L sum imposed by the symmetry requirement of the total target molecular wave function in interchanging the constituent nuclei when they are identical. Thus, in the case of H_2 and D_2, for example, we have, respectively,

$$\sum_{L}{}'=\sum_{\text{even }L}+3\sum_{\text{odd }L}, \quad \sum_{L}{}'=3\sum_{\text{odd }L}+6\sum_{\text{even }L}.$$

The lower limits in sums over v and L are restricted by the threshold energy [see Eq. (2.4)]:

$$E_e+\epsilon_v+\epsilon_L \geq 3.75 \text{ eV}. \qquad (3.3)$$

In writing down the thermal-averaged cross section [Eq. (3.2)], we have assumed that the temperature is not too high so that excited electronic states are not populated. (There is no low-lying excited electronic state in the hydrogen molecule.)

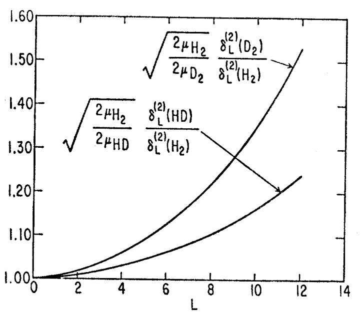

FIG. 4. Isotope effect on the imaginary phase shift for the $g \rightarrow u$ dissociative attachment at $E_e=3.775$ eV [Eq. (2.14)] as a function of nuclear angular momentum state L.

TABLE II. Survival probability in dissociative attachment of H_2 and its isotopes for the $g \rightarrow g$ case.

L	H_2 JWKB[a]	H_2 Γ/v[b]	HD JWKB[a]	HD Γ/v[b]	D_2 JWKB[a]	D_2 Γ/v[b]
0	2.298×10^{-5}	3.037×10^{-5}	4.410×10^{-6}	6.084×10^{-6}	2.769×10^{-7}	4.107×10^{-7}
1	2.306×10^{-5}	3.047×10^{-5}	4.423×10^{-6}	6.102×10^{-6}	2.776×10^{-7}	4.117×10^{-7}
2	2.321×10^{-5}	3.068×10^{-5}	4.448×10^{-6}	6.138×10^{-6}	2.789×10^{-7}	4.137×10^{-7}
3	2.345×10^{-5}	3.100×10^{-5}	4.488×10^{-6}	6.192×10^{-6}	2.809×10^{-7}	4.167×10^{-7}
4	2.377×10^{-5}	3.142×10^{-5}	4.540×10^{-6}	6.265×10^{-6}	2.836×10^{-7}	4.207×10^{-7}
5	2.417×10^{-5}	3.195×10^{-5}	4.606×10^{-6}	6.357×10^{-6}	2.870×10^{-7}	4.257×10^{-7}
6	2.465×10^{-5}	3.259×10^{-5}	4.687×10^{-6}	6.469×10^{-6}	2.911×10^{-7}	4.319×10^{-7}
7	2.523×10^{-5}	3.336×10^{-5}	4.782×10^{-6}	6.601×10^{-6}	2.960×10^{-7}	4.391×10^{-7}
8	2.590×10^{-5}	3.425×10^{-5}	4.893×10^{-6}	6.755×10^{-6}	3.016×10^{-7}	4.475×10^{-7}
9	2.667×10^{-5}	3.527×10^{-5}	5.019×10^{-6}	6.931×10^{-6}	3.080×10^{-7}	4.571×10^{-7}
10	2.754×10^{-5}	3.643×10^{-5}	5.163×10^{-6}	7.130×10^{-6}	3.153×10^{-7}	4.679×10^{-7}
11	2.853×10^{-5}	3.774×10^{-5}	5.325×10^{-6}	7.355×10^{-6}	3.234×10^{-7}	4.801×10^{-7}
12	2.963×10^{-5}	3.921×10^{-5}	5.506×10^{-6}	7.606×10^{-6}	3.325×10^{-7}	4.936×10^{-7}

[a] Calculated according to Eqs. (2.18) and (2.12b) at an incident electron energy of 10.2 eV.
[b] Calculated according to Eq. (1.3) at an incident electron energy of 10.2 eV.

The approximate stability of the cross-section ratio [Eq. (3.1)] with respect to the rotational state L as exhibited in Table III does not necessarily imply that thermal averaging is not important, since the thermal average is carried out before the ratio is taken. To calculate the observed isotope effect as given by the ratios of the experimental cross sections, an average of the theoretical cross section over the initial distribution of states is necessary. Hence, for example,

$$\frac{\bar{\sigma}_{\mathrm{H}_2}(T,E)}{\bar{\sigma}_{\mathrm{HD}}(T,E)}=\left(\frac{Z_{\mathrm{HD}}}{Z_{\mathrm{H}_2}}\right)\times\frac{\sum_v \sum'_L (2L+1)\sigma_{vL}(E,H_2)e^{-[\epsilon_v(\mathrm{H}_2)+\epsilon_L(\mathrm{H}_2)]/KT}}{\sum_{v',L'} (2L'+1)\sigma_{v'L'}(E,HD)e^{-[\epsilon_{v'}(\mathrm{HD})+\epsilon_{L'}(\mathrm{HD})]/KT}}. \tag{3.4}$$

Since the mass dependence of the capture cross section is[15]

$$\sigma_{L,vL}{}^{(c)}\propto\mu^{1/4}, \tag{3.5}$$

Eq. (3.4) takes the approximate form for low temperature:

$$\frac{\bar{\sigma}_{\mathrm{H}_2}(T,E)}{\bar{\sigma}_{\mathrm{HD}}(T,E)}\cong\left(\frac{\mu_{\mathrm{H}_2}}{\mu_{\mathrm{HD}}}\right)^{1/4}\frac{\bar{P}(\mathrm{H}_2)}{\bar{P}(\mathrm{HD})} \tag{3.6}$$

with

$$\bar{P}(\mathrm{H}_2)\equiv\frac{\sum'_L (2L+1)\{C_{L+1}p_{L+1}{}^{\mathrm{H}_2}+C_{L-1}p_{L-1}{}^{\mathrm{H}_2}\}e^{-\epsilon_L/KT}}{\sum'_{L'} (2L'+1)e^{-\epsilon_{L'}/KT}} \tag{3.7a}$$

and

$$\bar{P}(\mathrm{HD})\equiv\frac{\sum_L (2L+1)p_L{}^{\mathrm{HD}}e^{-\epsilon_L/KT}}{\sum_{L'} (2L'+1)e^{-\epsilon_{L'}/KT}}, \tag{3.7b}$$

where we have assumed that at low temperature only the ground vibrational state is of importance. One of the interesting features of Eqs. (3.4) and (3.6) is the prediction of the temperature dependence of the isotope effect which can be easily observed experimentally. The calculated isotope cross-section ratio as given by Eq. (3.6) is smaller than that observed. In the $g \rightarrow u$ case, the calculated values for $\bar{\sigma}_{\mathrm{H}_2}/\bar{\sigma}_{\mathrm{HD}}$ and $\bar{\sigma}_{\mathrm{H}_2}/\bar{\sigma}_{\mathrm{D}_2}$ are 5.01 and 55.85, whereas the observed values are about 7.62 and 200, respectively. The accuracy of our result is, of course, bounded by the JWKB approximation and by the accuracy of the adopted theoretical values for Γ and V.[12] Judging from these values, it seems to suggest that the discrepancy between the theoretical results and the

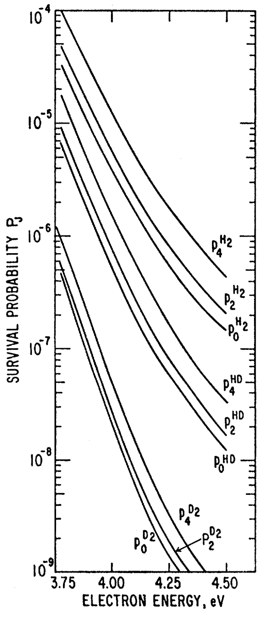

FIG. 5. Dependence of the survival probability on the incident electron energy for the $g \rightarrow u$ dissociative attachment of H_2, HD, and D_2.

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[15] The $\mu^{1/4}$ mass dependence may not constitute a good approximation for the capture cross section since the similarity transformation by mass scaling in the nuclear overlap integral does not factor out the mass dependence completely. Consequently, the continuum nuclear wave function has an inherent mass dependence which can be enhanced by a large imaginary component of the complex potential. {*Note added in proof*. The observation that the $\mu^{1/4}$ mass dependence may not constitute a good approximation for the capture cross section has recently been verified numerically [J. C. Y. Chen and J. L. Peacher (to be published)]. An easy way to see this is to examine the coefficients in the asymptotic forms of the continuum nuclear wave function as given by Eqs. (2.3) and (2.5), since both $\cosh\delta_J{}^{(2)}$ and $\sinh\delta_J{}^{(2)}$ can be expressed in terms of the survival probability [Eq. (2.18)].}

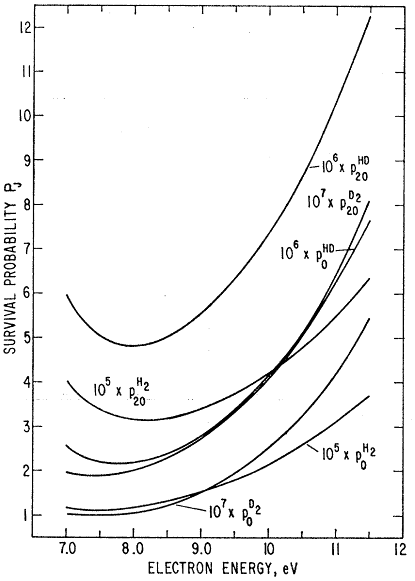

FIG. 6. Dependence of the survival probability on the incident electron energy for the $g \rightarrow g$ dissociative attachment of H_2, HD, and D_2.

experimental results are significant. This point will be discussed in Sec. IV.

TABLE III. Isotope effect due to the survival probability in the $g \rightarrow u$ dissociative attachment.[a]

	$\sigma_{vL}(E,\mathrm{H}_2)/\sigma_{vL}(E,\mathrm{HD})$[b]		$\sigma_{vL}(E,\mathrm{H}_2/)\sigma_{vL}(E,\mathrm{D}_2)$[c]	
L	JWKB[d]	Γ/v[e]	JWKB[d]	Γ/v[e]
0	4.951	13.28	72.26	1012.8
1	5.718	15.06	77.73	1080.0
2	5.995	15.75	83.57	1155.8
3	6.380	16.81	92.19	1275.7
4	6.837	18.27	103.23	1448.5
5	7.317	20.17	116.02	1687.5
6	7.763	22.59	129.52	2012.0
7	8.123	25.61	142.34	2449.9
8	8.353	29.35	152.98	3041.5
9	8.433	33.96	160.12	3843.7
10	8.364	39.61	162.95	4935.4
11	8.166	46.46	161.31	6422.9

[a] These are calculated at an incident electron energy of 3.775 eV.
[b] See Eq. (3.4).
[c] Approximation given by Eq. (2.27) is used here in deriving the ratio equation similar to Eq. (3.4).
[d] The survival probabilities used here are calculated using Eqs. (2.12b) and (2.18).
[e] The survival probabilities used here are calculated using Eq. (1.3).

TABLE IV. The exponential factor in the survival probability arising from the complex potential at the turning point.

	$\exp\{-2\mathfrak{D}_L^{(-)}(R_0)\}$ for $g \rightarrow u$[a]			$\exp\{-2\mathfrak{D}_L^{(-)}(R_0)\}$ for $g \rightarrow g$[b]		
L	H_2	HD	D_2	H_2	HD	D_2
0	0.634	0.590	0.523	0.3425	0.2903	0.2199
1	0.668	0.617	0.543	0.3425	0.2902	0.2199
2	0.738	0.674	0.583	0.3424	0.2902	0.2198
3	0.849	0.764	0.648	0.3423	0.2901	0.2198
4	1.008	0.893	0.741	0.3422	0.2900	0.2197
5	1.218	1.067	0.869	0.3420	0.2899	0.2196
6	1.479	1.292	1.037	0.3418	0.2897	0.2195
7	1.782	1.570	1.252	0.3415	0.2895	0.2194
8	2.105	1.894	1.521	0.3412	0.2893	0.2193
9	2.411	2.248	1.846	0.3409	0.2891	0.2192
10	2.656	2.605	2.223	0.3406	0.2888	0.2190
11	2.798	2.925	2.640	0.3403	0.2886	0.2188
12	2.814	3.168	3.075	0.3399	0.2883	0.2186

[a] Calculated at an incident electron energy of 3.775 eV.
[b] Calculated at an incident electron energy of 10.2 eV.

Recently, Schulz and Asundi[4] made use of a ratio formula derived from Eqs. (1.1) and (1.2) for estimating the width of the $^2\Sigma_u^+$ state of H_2^- by utilizing measured dissociative-attachment cross sections for various isotopes of H_2. As shown in Fig. 4, the simple relation for the imaginary phase shift

$$\delta_L^{(2)}(\mathrm{H}_2)/(2\mu_{\mathrm{H}_2})^{1/2} \cong \delta_L^{(2)}(\mathrm{HD})/(2\mu_{\mathrm{HD}})^{1/2} \cong \delta_L^{(2)}(\mathrm{D}_2)/(2\mu_{\mathrm{D}_2})^{1/2} \tag{3.8}$$

holds approximately for low values of L. Making use of Eq. (3.8), we obtain from Eq. (3.1) the approximate equations

$$\frac{\sigma_L(\mathrm{H}_2)}{\sigma_L(\mathrm{HD})} \cong \exp\left\{-2\bar{\delta}_L^{(2)}(\mathrm{H}_2)\left[1-\left(\frac{\mu_{\mathrm{HD}}}{\mu_{\mathrm{H}_2}}\right)^{1/2}\right]\right\} \tag{3.9a}$$

and

$$\frac{\sigma_L(\mathrm{H}_2)}{\sigma_L(\mathrm{D}_2)} \cong \exp\left\{-2\bar{\delta}_L^{(2)}(\mathrm{H}_2)\left[1-\left(\frac{\mu_{\mathrm{D}_2}}{\mu_{\mathrm{H}_2}}\right)^{1/2}\right]\right\}. \tag{3.9b}$$

Equations (3.9) demonstrate that by utilizing the measured values for the cross-section ratios $\sigma(\mathrm{H}_2)/\sigma(\mathrm{HD})$ and $\sigma(\mathrm{H}_2)/\sigma(\mathrm{D}_2)$ at low temperature ($T \cong 300°\mathrm{K}$), an approximate constant can be found at a given electron energy. The constant so determined, aside from the problem of thermal averaging, approximates the imaginary phase shift which is not simply related to the width unless the approximation given by Eq. (1.3) is valid. For the present case Eq. (1.3) is not valid as is shown in Table I.

Finally, the energy dependence of the survival probability is investigated. The result is shown in Figs. 5 and 6. We observe that for the $g \rightarrow u$ case the survival probability is also a sensitive function of energy. As shown in Eq. (2.11), the starting value $\mathfrak{D}_L^{(-)}(R_0)$ of the JWKB imaginary phase shift which is derived from the asymptotic phase of the Airy function is implicitly energy-dependent. This factor, i.e.,

$$\exp[-2\mathfrak{D}_L^{(-)}(R_0)],$$

in the survival probability is tabulated for several values of L in Table IV. It is observed that its contribution, in this case, is significant. It is worthwhile to note that this factor has a larger effect for the $g \rightarrow g$ case than for the $g \rightarrow u$ case. This indicates that the reasonable agreement shown in Table II with the approximation given by Eq. (1.3) is fortuitous, since more exactly this factor should be included in Eq. (1.3) as given by Eq. (2.13). The inclusion of this factor would then destroy the reasonable agreement. The magnitude of this factor depends critically on the difference of the slopes of the imaginary and real parts of the potential. It should be noted that $\mathfrak{D}_L^{(-)}(R_0)$ changes sign at some particular value of L. This implies that this factor can either increase or decrease the survival probability. Thus, the validity of the approximate expressions for the survival probability given by Eqs. (1.2) and (1.3) are limited, in addition to the criterion given by Eq. (1.4), to cases where this factor is approximately unity.

IV. DISCUSSION AND CONCLUSIONS

In this study we have demonstrated that the simple expression for the dissociative-attachment cross section, as given by the product of a capture cross section and a survival probability, is not a general expression. A general expression for the cross section can nevertheless be expressed as a sum of products of a capture cross section and a survival probability for the various contributing angular momentum states of the constituent nuclei. This unfortunately destroys the simple proportional dependence of the cross section on the survival probability. In an attempt to preserve this simple picture, we show for the (e,H_2) system that if one retains only the lowest contributing partial waves of the incident electron, one may still write the cross section as a product of two terms in which the survival probability is somewhat modified [Eqs. (2.25) and (2.26)].

The selection rule derived for the $g \rightarrow u$ dissociative attachment is of interest in that it suggests that the $^2\Sigma_u{}^+$ $H_2{}^-$ state is actually a low-lying p-wave resonance in slow electron scattering by hydrogen molecules. Experimentally this can be observed by measuring the angular dependence in resonance electron scattering by H_2 molecules or the angular distribution of the detached electron in associative detachment $H+H^- \rightarrow H_2+e$ by crossed-beam techniques at appropriate energies.

The numerical study of the survival probability carried out in Sec. III demonstrates the reliability of various approximate expressions for the survival probability. Perhaps the most interesting feature of the numerical study lies in the dependence of the survival probability on the relative angular momentum states J. We have found a strong J dependence of the survival probability for the $g \rightarrow u$ dissociative attachment, but for the $g \rightarrow g$ dissociative attachment the J dependence is much weaker. Since the J dependence is strong in the $g \rightarrow u$ case, one would then expect a significant temperature dependence of the cross section. Of course, the thermal distribution of initial vibrational states v will also give rise to a temperature dependence of the cross section through the v dependence of the capture cross section.

The detailed numerical results of our study such as the isotope effect depend critically on both the real and imaginary parts of the interaction potential. The general features of the survival probability nevertheless remain relatively insensitive to reasonable variations in the adopted potential curves. We found that the general behavior of the J dependence of the survival probability varies according to whether the real part of the potential is attractive or repulsive. For an attractive potential of the Morse type, it is possible to have a strong J dependence for the survival probability if the threshold lies within (or near) the Franck-Condon region of the target molecule. This arises because the constituent atoms may then dissociate with very low relative kinetic energy.

ACKNOWLEDGMENTS

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