

Dissociative electron attachment to vibrationally and rotationally excited H₂ and HF molecules

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(Received 25 October 1999; published 6 April 2000)

The vibrational and rotational state dependence of dissociative attachment (DA) in low-energy e -H₂ and e -HF collisions is studied within the framework of nonlocal resonance theory. The dynamics of nuclear motion in the nonlocal complex potential is treated by the quasiclassical approach. The resonance energy, width function, and level-shift function are taken from our previous calculations based on the projection operator formalism. Results for all vibrational states up to the dissociative attachment thresholds and selected rotational states are presented. Our cross sections for H₂ exhibit a plateau structure in the energy range above the dissociation threshold, and this structure gets more pronounced with higher v or J . The dissociative attachment cross section for vibrationally and rotationally hot HF ($T=1150$ K) agrees well with available experimental data. For H₂ in thermal equilibrium at 1400 K, the agreement between experiment and theory is less satisfactory.

PACS number(s): 34.80.Ht

I. INTRODUCTION

Dissociative attachment (DA) in low-energy electron molecule scattering has been a subject of many experimental and theoretical investigations. Theoretically this process can be interpreted in terms of formation and dissociation of a temporary negative molecular ion [1,2]. The resonance energy and width were usually obtained by either fitting a parametrized width function to the experimental data [3], or by *ab initio* calculations [4]. The resonance energy obtained in the former case can then be adopted as a local complex potential energy for the nuclear motion to calculate the DA cross section [5]. In the latter case, Feshbach's projection operator formalism is usually used to separate the resonance state from the background scattering, and the nuclear dynamics is governed by the resultant energy-dependent, nonlocal complex potential [6,7]. It was pointed out that the local theory fails severely when the resonance that drives the DA process is broad [8,9]. Later on, however, Atems and Wadehra [10] and Hickman [11] showed that special versions of local theory applied to DA to the H₂ molecule can give results which are close to those of the nonlocal theory. It is not clear, though, to what extent these methods can be generalized for molecules other than H₂. In addition, both papers use a separable expression for the nonlocal potential. The separability appears due to inclusion of only open channels in the expression for the nonlocal complex potential. Whereas this procedure is justified for the imaginary part of the potential, it cannot generally be used for the real part. Moreover, it is known [4,12,13] that contribution of the closed channels corresponding to excitation of discrete vibrational states and even vibrational continuum can be very important. When the electron energy approaches the dissociation limit, the use of the separable approximation becomes even more questionable [14].

Therefore, for comparison of the local and nonlocal versions of the resonance theory of DA it is important to use the *exact* formulation of the nonlocal theory and the general version of the local theory without model assumption regarding the coupling parameter or width. So far this kind of compari-

son has been done only for attachment to H₂ in its ground rovibrational state [8]. The present work extends this type of calculations to rovibrationally excited H₂ molecule and the HF molecule.

DA to H₂ in the energy range up to 5 eV has been studied intensively in the past three decades [15–19]. Measurements of the absolute cross section for attachment to the ground state ($v=J=0$) were reported by Schultz and Asundi in 1967 [15]. Allan and Wong [16] showed that the cross section increases dramatically as the internal energy of H₂ increases, and this behavior was explained by the analysis of Wadehra and Bardsley based on the local complex potential model [3]. Several nonlocal calculations were performed more recently. Whereas Mündel *et al.* [8] and Gallup *et al.* [20] developed methods allowing complete inclusion of vibrational dynamics, Atems and Wadehra [10] and Hickman [11] included only open channels in the expression for the nonlocal potential. DA to vibrationally excited H₂ has been studied by several nonlocal resonance theories [10,11,20], and also by nonresonant effective range theory [21]. Nonlocal calculations of DA cross section to H₂ in thermal equilibrium at 1400 K were published recently [22].

HF is another interesting target for understanding the DA process. It possesses a supercritical electron dipole moment which causes the resonance width to be finite at zero electron energy. Resonant R -matrix calculations and nonlocal calculations based on the projection operator approach have been done for DA to HF molecule [23,20]. The cross section for the ground state of HF, and the vibrational enhancement were found to agree well with experiment.

In this paper we present the DA cross section for vibrationally and rotationally excited H₂ and HF. The next section outlines our theory, with emphasis on the treatment of the nuclear dynamics, Secs. III and IV give results for H₂ and HF, respectively, and Sec. V contains concluding remarks.

II. THEORY

We follow the quasiclassical version of the nonlocal resonance theory [24] for calculation of dissociative electron at-

tachment cross sections. The method is based on the Feshbach projection operator formalism [25] and has been applied to DA from low-lying vibrational states of H_2 and HF molecules. In this work we calculate DA from higher excited vibrational states and extend the theory to include rotational motion. We will only outline our treatment of the nuclear dynamics here. The description of the neutral molecule and the resonance state is identical to that used in Ref. [20]. In our treatment of rotational motion we assume that the formation of the negative-ion resonance is dominated by one partial wave with angular momentum 0 (HF molecule) or 1 (H_2 molecule), so that the rotational quantum number J can be treated as a conserved quantity during the DA process.

The Schrödinger equation describing the nuclear motion in the temporary negative ion state with total energy E and reduced mass M is [4]:

$$(E - H_d)|\Psi_{\mathbf{k}_i E}\rangle - F|\Psi_{\mathbf{k}_i E}\rangle = V_{b\mathbf{k}_i}|v_i J\rangle, \quad (1)$$

where $|\Psi_{\mathbf{k}_i E}\rangle$ is the wave function in the dissociating channel satisfying the outgoing wave boundary conditions, H_d is the Hamiltonian for the nuclear motion in the diabatic potential, $|v_i J\rangle$ is the initial rovibrational state of the neutral molecule, \mathbf{k}_i is the momentum of the incoming electron, F is the nonlocal complex potential and $V_{b\mathbf{k}_i}$ is the coupling amplitude between the diabatic negative-ion state and the electron continuum.

If we expand $|\Psi_{\mathbf{k}_i E}\rangle$ in the eigenstates $|v J\rangle$ of the Hamiltonian for the neutral molecule,

$$|\Psi_{\mathbf{k}_i E}\rangle = \sum_{vJ} b_{vJ\mathbf{k}_i}|v J\rangle, \quad (2)$$

and make a substitution

$$b_{vJ\mathbf{k}_i} = V_{b\mathbf{k}_i}(R_{v_i})a_{vJ}, \quad (3)$$

the quasiclassical approach for nuclear dynamics gives the following equation for a_{vJ} [20]:

$$a_{vJ} - \sum_{v'} \langle vJ | G_d^{(+)}(E) | v' J \rangle F_{v'} a_{v'J} = \langle vJ | G_d^{(+)}(E) | v_i J \rangle. \quad (4)$$

Here, F_v is related to the partial resonance widths Γ_v and shifts Δ_v at the Franck-Condon point R_v as

$$F_v = \int k dk d\hat{\mathbf{k}} \frac{|V_{b\mathbf{k}}(R_v)|^2}{E + i0 - E_v - \frac{1}{2}k^2} = \Delta_v - \frac{i}{2}\Gamma_v, \quad (5)$$

where E_v is the energy of molecular vibrational state v .

The Green's-function matrix element in Eq. (4) can be represented in a separable form [24,26,27]. For inclusion of the rotational motion, we extend the representation to be the following:

$$\langle vJ | G_d^{(+)}(E) | v' J \rangle = \frac{A(vJ)A(v'J)}{W} B_{>}(v_{>}J) B_{<}(v_{<}J), \quad (6)$$

where $v_{>}$ and $v_{<}$ are $\max(v, v')$ and $\min(v, v')$, respectively, W is a constant Wronskian, and $A(vJ)$, $B_{<}(vJ)$, and $B_{>}(vJ)$ are coefficients depending on the case under consideration. In what follows, we drop the rotational quantum number J as an index in order to simplify notation.

If the transition point is closer to the left molecular turning point $a_0(v)$, then

$$B_{<}(v) = \left(\frac{\partial S(v)}{\partial E_v} \right)^{-1/2} \left(\frac{3}{2} S(v) \right)^{1/6} \text{Ai} \left[- \left(\frac{3}{2} S(v) \right)^{2/3} \right], \quad (7)$$

$$B_{>}(v) = \left(\frac{\partial S(v)}{\partial E_v} \right)^{-1/2} \left(\frac{3}{2} S(v) \right)^{1/6} \frac{i-1}{\sqrt{2}} W^{-1} \left(- \left(\frac{3}{2} S(v) \right)^{2/3} \right), \quad (8)$$

$$A(v) = \left[\omega(v) \frac{dR_v}{dE_v} \frac{\tau_v(a_0(v), R_v)}{2} \times \left(E_v - U_0(R_v) - \frac{J(J+1)}{2MR_v^2} \right)^{-1/2} \right]^{1/2}, \quad (9)$$

where U_0 is the potential of the neutral molecule, $\omega(v)$ is the vibrational frequency of molecular vibrational state v , and

$$\tau_v(a_0(v), R_v) = \int_{a_0(v)}^{R_v} dR \left(E_v - U_0(R) - \frac{J(J+1)}{2MR^2} \right)^{-1/2} \quad (10)$$

is the reduced time of nuclear vibration from the left turning point to the transition point. The action function $S(v)$ is defined as

$$S(v) = \int_{a_0(v)}^{R_v} \left[2M[E_v - U_0(R)] - \frac{J(J+1)}{R^2} \right]^{1/2} dR - \int_{a_1(E)}^{R_v} \left[2M[E - U_1(R)] - \frac{J(J+1)}{R^2} \right]^{1/2} dR, \quad (11)$$

where $U_1(R)$ is the diabatic molecular energy curve, $a_1(R)$ is the left turning point for the negative ion with energy E .

When the transition point R_v is closer to the right molecular turning point $b_0(v)$, the following representations are used instead:

$$B_{<}(v) = \left(\frac{\partial S(v)}{\partial E_v} \right)^{-1/2} \left(\frac{3}{2} S(v) \right)^{1/6} \text{Ai} \left[- \left(\frac{3}{2} S(v) \right)^{2/3} \right], \quad (12)$$

$$B_{>}(v) = \left(\frac{\partial S(v)}{\partial E_v} \right)^{-1/2} \left(\frac{3}{2} S(v) \right)^{1/6} \frac{1-i}{\sqrt{2}} W^+ \left(- \left(\frac{3}{2} S(v) \right)^{2/3} \right), \quad (13)$$

$$A(v) = (-1)^v \left[\omega(v) \frac{dR_v}{dE_v} \frac{\tau_v(R_v, b_0(v))}{2} \right. \\ \left. \times \left(E_v - U_0(R_v) - \frac{J(J+1)}{2MR_v^2} \right)^{-1/2} \right]^{1/2}, \quad (14)$$

$$S(v) = \int_{R_v}^{b_0(v)} \left[2M[E_v - U_0(R)] - \frac{J(J+1)}{R^2} \right]^{1/2} dR \\ + \int_{a_1(E)}^{R_v} \left[2M[E - U_1(R)] - \frac{J(J+1)}{R^2} \right]^{1/2} dR. \quad (15)$$

When the Franck-Condon point R_v lies between the two molecular turning points, the two representations for the Green's-function matrix element match smoothly.

The action function $S(v)$ and its derivative in E , the reduced time of nuclear vibration τ_v , and the vibration frequency $\omega(v)$ are all calculated numerically. Equation (4) can then be solved recursively in v and the solution is determined by matching with the vibrational continuum function at the dissociation threshold [24]. We should emphasize that this matching process takes into account the effect of the vibrational continuum in a suitable way and thus allows for a reliable description of the dissociative attachment to highly excited states [14].

Finally, the total DA cross section $\sigma_{DA}(v_i)$ is given by [4]

$$\sigma_{DA} = \frac{2\pi^2}{k_i^2} \nu \frac{K}{M} \left| \lim_{R \rightarrow \infty} \Psi_{k_i E}(R) \right|^2, \quad (16)$$

where K is the relative momentum of the dissociating fragments, and ν accounts for the spatial degeneracy of the electronic resonance state.

III. RESULTS FOR THE H₂ MOLECULE

Figure 1 gives the DA cross section for different vibrational states of H₂ up to $v=9$. As was shown in Ref. [20], our cross section for the ground state is in good agreement with the nonlocal results based on the Lanczos-basis treatment of the nuclear dynamics [8]. Since the electronic parts of the two calculations are almost identical, this agreement implies that the quasiclassical treatment of the nuclear dynamics is a good approximation for DA to the ground state of the H₂ molecule. In view of the comparison made for HCl in Ref. [24], it is reasonable to expect the quasiclassical approach to work better for excited states of H₂. Our calculated cross sections for vibrationally excited states ($v=1,2$), however, are slightly different from those of Mündel *et al.* [8]. For highly excited vibrational states ($v>2$), our cross sections are quite different from those obtained using local cal-

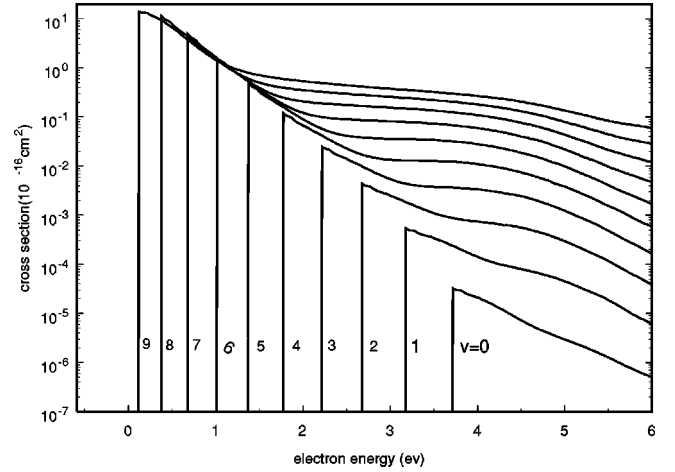


FIG. 1. Dissociative attachment cross sections for various vibrational states of H₂.

culations [3]. The difference is mainly in the energy dependence of the cross section. In the energy range above the dissociation threshold, our cross section exhibits a plateau structure, and for higher v the cross section drops more slowly with increasing energy. For $v=9$, the DA cross section remains almost a constant from 1 to 4 eV.

This interesting feature of the cross section can be interpreted as a result of the nonlocal effect. Indeed, at higher energies the point of transition between the initial vibrational state and the resonance state (the Franck-Condon point) moves towards smaller internuclear distances where the resonance width is larger. In addition, the Franck-Condon point is moving away from the classically allowed region which makes the Franck-Condon overlap smaller. This causes a fast drop in the local cross section. In the nonlocal theory larger internuclear distances, where the width is smaller, are also involved in the process. Therefore, nonlocality stabilizes the DA process. At smaller electron energies, however, nonlocality might substantially suppress the DA rate.

This is illustrated in Fig. 2 where we compare our local

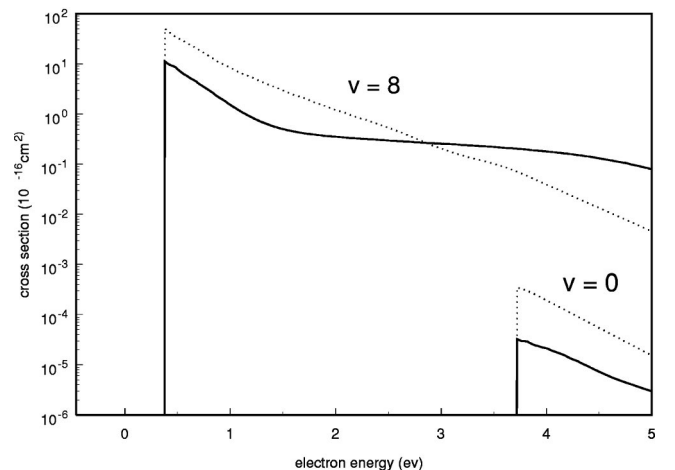


FIG. 2. Local and nonlocal DA cross sections for vibrational states of H₂ with $v=0$ and $v=8$. Solid curves, nonlocal result; dotted curves, local result.

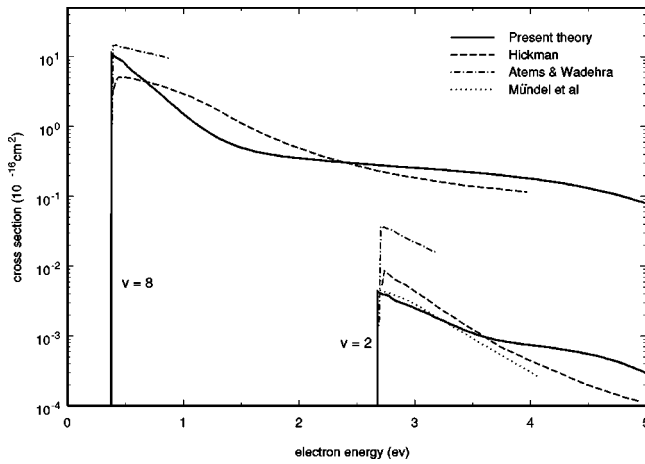


FIG. 3. Comparison of calculated DA cross sections for vibrationally excited states of H_2 with several previous nonlocal calculations.

and nonlocal DA cross sections for vibrationally ground state and excited state $v=8$. In accordance with the earlier observations [8,4], the local cross section for the vibrationally ground state is an order of magnitude higher than the nonlocal cross section. For $v=8$, although the local cross section is somewhat higher near the threshold, it drops much faster with increasing energy, and the nonlocal curve crosses the local one in the middle of the plateau region.

Atems and Wadehra [10] and Hickman [11] calculated nonlocal DA cross sections for vibrationally excited states up to $v=9$ and also made comparison between their version of local and nonlocal theories. The former calculations use a parametrization of the resonance width which is compatible with the Wigner threshold law and fit the parameters to reproduce the experimental cross section [15] at the DA threshold. Hickman [11] uses the same description of the resonance state as that of Mündel *et al.* [8] but includes only open channels in the expression for the nonlocal potential. Both papers, in contrast to what Fig. 2 shows, conclude that the local theory agrees reasonably well with the nonlocal results, especially for highly excited states. We should note

first that the semiempirical aspect of the Atems and Wadehra calculation does not allow for rigorous conclusions about the validity of the local approximation. Second, the local version of the approximation used by Hickman is based on special parametrization of the resonance width and is different from the standard version, therefore, it is not clear how general his conclusion is. Finally, perhaps most importantly, both calculations use the separable approximation for the nonlocal potential. The validity of this approximation is questionable [4,12], especially in the energy region close to and above the dissociation threshold [14]. This is suggested by Fig. 3, where we compare our results with previous nonlocal calculations involving the separable approximation and the calculations using the Lanczos-basis approach [8]. For $v=2$, our cross section is close to that of Mündel *et al.* [8] in the low-energy range, but drops slower with increasing energy above the dissociation threshold. For both $v=2$ and $v=8$, the energy dependence of our cross sections is somewhat different from that of Hickman's calculation [11], although Hickman's results do show a sign of the plateau behavior. A similar comparison with the results of Atems and Wadehra cannot be done since they present cross sections only in vicinities of DA thresholds.

In Fig. 4 we show the DA cross sections for different rotational states of the vibrational ground state of H_2 . The cross sections for highly excited states exhibit the same plateau structure above the dissociation threshold. We also observe weak irregularities between the DA threshold and the dissociation threshold. This type of structure appears also in DA to vibrationally excited states. We find that these irregularities emerge only at vibrational excitation thresholds and thus are caused by the opening of higher vibrational channels.

Figure 5 compares our DA cross section to H_2 at $T=1400$ K with experimental data and previous nonlocal calculation of Čížek *et al.* [22]. Our cross sections were obtained by averaging the DA cross section over the Maxwell-Boltzmann distribution. All rovibrational states below the DA threshold were included. The experimental data were normalized to the absolute cross-section value of Schultz and

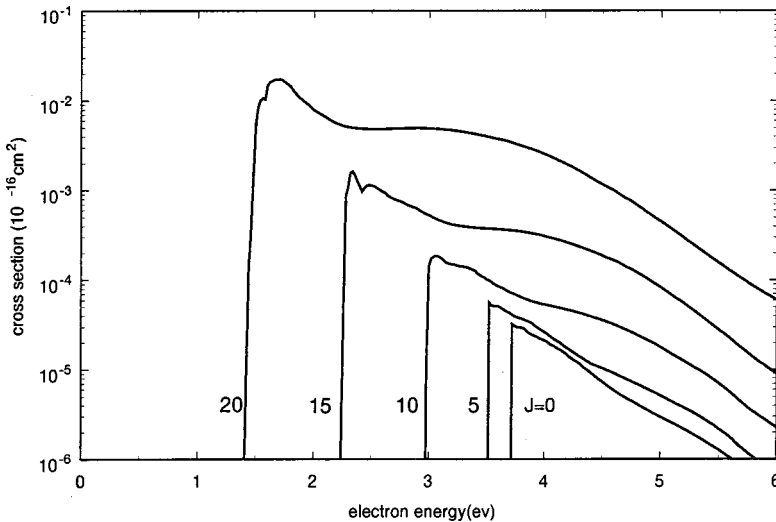


FIG. 4. Dissociative attachment cross sections for various rotational states of H_2 . From right to left the curves correspond to $v=0$ and $J=0,5,10,15,20$.

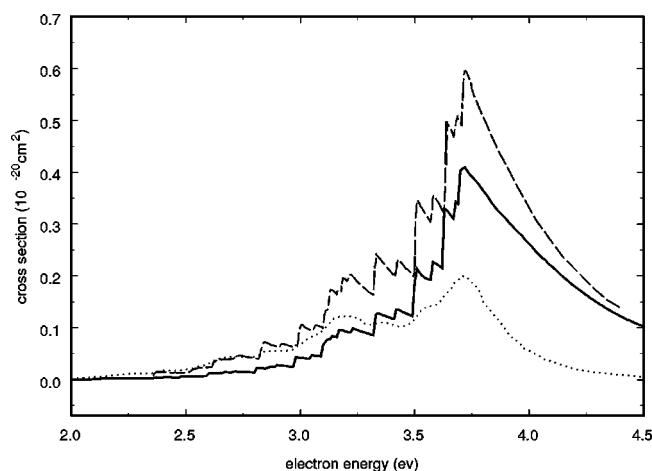


FIG. 5. Cross section for dissociative attachment to H_2 in thermal equilibrium at 1400 K. Solid curve, present result; dashed curve, nonlocal calculations [22]; dotted curve, experimental data [16].

Asundi [15]. Our results are in general closer to the experimental data in the high-energy range than that of the previous calculation, but smaller than both the experiment and the previous theory in the low-energy range, since our threshold cross section for the ground state is about 50% lower than the result of Čížek *et al.*, and the vibrational enhancement of these two calculations are about the same.

Our calculation does not resolve the major discrepancy between the experiment and theory. As was pointed out by Čížek *et al.* [22], the major problem with nonlocal calculations for DA to H_2 is that the calculated threshold peak for the ground state is usually too high, and the vibrational enhancement is usually too weak.

IV. RESULTS FOR THE HF MOLECULE

In Fig. 6 we present the DA cross section for vibrationally excited HF ($v=0-5$). As was observed for $v=0-2$ in Ref. [20], the cross section displays a step structure at each vibrational excitation threshold. This phenomenon was ob-

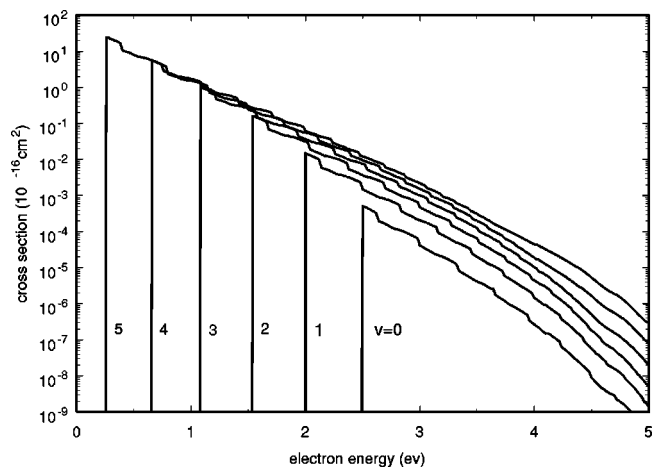


FIG. 6. Dissociative attachment cross sections for various vibrational states of HF.

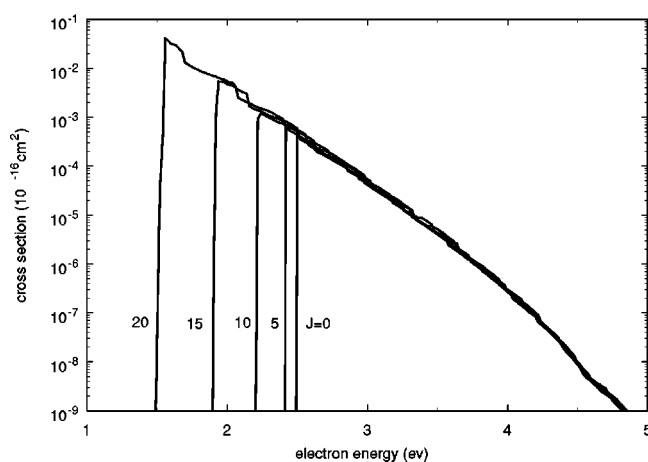


FIG. 7. Dissociative attachment cross sections for various rotational states of HF. From bottom to top the curves correspond to $v=0$ and $J=0,5,10,15,20$.

served experimentally for DA to the ground state of HF [28] and HCl [29], and can be explained by the sudden drop of the negative-ion survival probability at each vibrational excitation threshold. In our theory, this drop is represented by the sudden change of the partial resonance width at zero electron energy for a fixed internuclear distance, which is well known for molecules possessing permanent dipole moments [30,31].

Figure 7 gives our result for different rotational states of the vibrational ground state of HF. Except for threshold effects, the cross sections for different rotational states are about the same at the same electron energy. Although somewhat unexpected, this phenomenon can be qualitatively rationalized by considering the relatively weak influence of rotational motion on vibrational structure. For HF the spacing between vibrational levels is about the same for different rotational quantum numbers. A similar and even more drastic effect was obtained by Wadehra in his calculations of the rotational effect in dissociative attachment to Li_2 [32]. The DA cross section to Li_2 in the $J=22$ state is even slightly smaller than that for the ground rotational state. Apparently DA cross section becomes less dependent on J for heavier molecules.

In Fig. 8 we compare our cross section for the production of F^- at 1150 K with the experimental data of Allan and Wong [33]. The experimental cross sections were normalized to the peak value of our calculation. The results agree quite well. The rotational structure was not observed in the experiment because of its low-energy resolution.

The ratios of the peak cross sections for different vibrational and rotational states to that of the ground state of HF are given in Fig. 9. For two available experimental points our results are quite close to the experiment. The cross section enhancement by vibrational excitation was found to be much stronger than that by rotational excitation at the same internal energy. This confirms previous conclusions [3] based on results of local calculations.

V. CONCLUSIONS

We have performed an *ab initio* study of vibrational and rotational effects in dissociative electron attachment to the

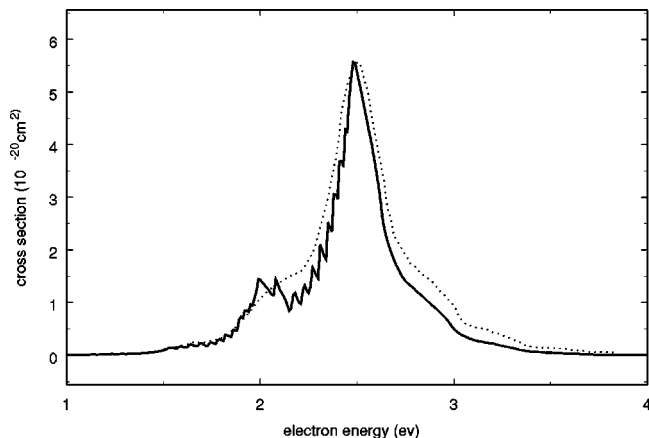


FIG. 8. Cross section for dissociative attachment to HF in thermal equilibrium at 1150 K. Solid curve, present result; dotted curve, experimental data [33] normalized to the peak value of the present theory.

H_2 and HF molecules using the projection operator formalism. The quasiclassical approach to the nuclear dynamics allows us to formulate the nonlocal resonance theory in such a way that it takes into account explicitly the vibrational continuum for the nuclear motion. This enables us to obtain reliable cross sections for DA to highly excited vibrational and rotational states.

Our calculated DA cross section for H_2 at $T=1400$ K basically agrees with recent nonlocal calculations [22], although our result is somewhat lower. Both theoretical sets of cross sections differ quite noticeably from experimental results [15,16]. The peak cross section for the ground state is too big, and the vibrational enhancement is too weak. We should emphasize that the near-threshold DA cross section for H_2 is very small, which creates substantial uncertainties both in theory and experiment. While more accurate theoretical calculations of the H_2 and H_2^- potential energy curves still need to be done, further experimental investigations of this fundamental collision process would be very desirable.

In the energy region above the dissociation threshold we find a plateau-type behavior of the cross sections which becomes more pronounced with higher v or J . This behavior should substantially affect the DA rates at higher tempera-

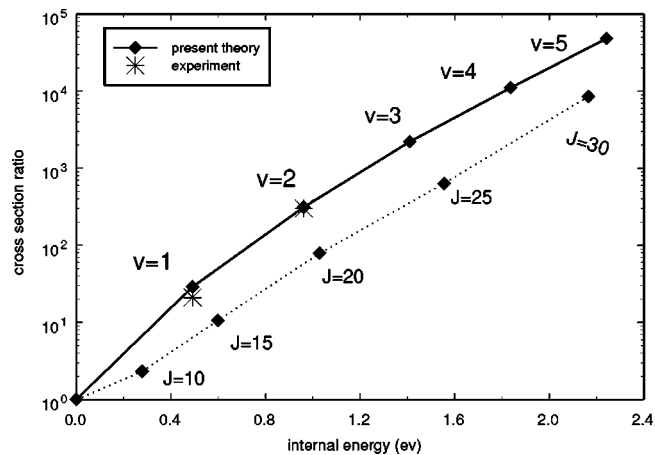


FIG. 9. Internal-state dependence of threshold dissociative attachment cross section in HF. Solid curve, $\sigma_v/\sigma_{v=0}$; dotted curve, $\sigma_J/\sigma_{J=0}$ for vibrational ground state. The experimental data are from Allan and Wong [33].

tures when the contribution of highly excited states becomes more important.

The DA cross section for vibrationally and rotationally excited HF agrees better with the available experimental data. The main features of the experimental DA cross section are represented by our theory very well. The enhancement of the cross section due to vibrational motion is found to be much stronger than that due to rotational motion, as was found previously for H_2 [3].

Note added in proof. Recently, the authors have learned about calculations of DA to H_2 by Kazansky [34] using the nonstationary nonlocal theory [35]. His results do not exhibit the plateau structure discussed in the present paper, and this disagreement might be caused by an inaccuracy of the quasiclassical theory. However, the major results of the present paper (dependence of cross sections on J and v and the temperature effects) would not be affected by this possible inaccuracy. We are grateful to A. K. Kazansky for communicating his unpublished results to us and for useful comments.

ACKNOWLEDGMENT

This work was supported by the National Science Foundation through Grant No. PHY-9801871.

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