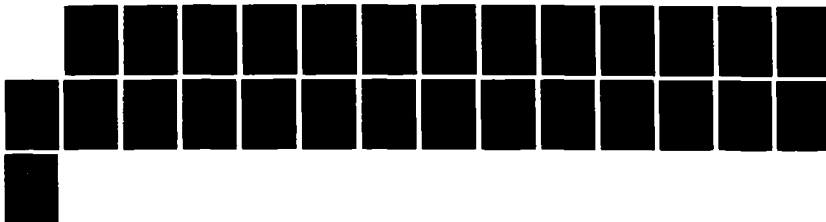
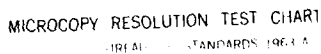


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Nascent Vibrational/Rotational Distribution
Produced by Hydrogen Atom Recombination

A. O. WOODIN
Aerophysics Laboratory
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The Aerospace Corporation
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1 February 1988

Prepared for
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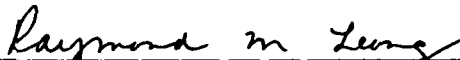
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The nascent vibrational/rotational distribution produced by the reaction $H + H + H + H_2(v, J) + H$ is calculated using resonance complex theory. The calculations are performed using the accurate Siegbahn-Liu-Truhlar-Horowitz surface. Results are presented for the dependence of the total recombination rate coefficient on temperature, and specific rate coefficients into individual vibrational/rotational states. Key					
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PREFACE

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I. INTRODUCTION

For almost 50 years there has been great experimental¹⁻⁸ and theoretical⁹⁻¹⁸ interest in the gas-phase recombination of hydrogen, the process



Reaction (1), where $\text{M} = \text{H}$, represents one of the simplest chemical reactions and is of fundamental interest to chemical dynamics and astrophysics. Indeed, this reaction tests the outer limits of the $(\text{H})_3$ potential energy surface. This reaction is of particular interest because of the recent calculations and experiments on the similar $\text{H} + \text{D}_2$ system.¹⁹⁻²² The comparison of theory and experiment in these simple three-electron systems tests our ability to understand chemical reactions.

Recombination of hydrogen is of practical importance in both plasma physics and rocket propulsion, especially for hybrid and electrothermal propulsion systems.^{23,24} This reaction becomes important in chemical rockets if hydrogen is a major atomic component of the fuel. In these systems, the total recombination rate coefficient and the distribution of energy into the product vibrational/rotational states are important. For efficient propulsion, the translational energy of the exhaust must be maximized. Frozen flow efficiency losses occur when the molecules do not sustain enough collisions in the nozzle to equilibrate the translational and vibrational/rotational degrees of freedom. The energy that is trapped in the vibrational/rotational modes of the system is not available for thrust.

Experimental reaction rate data were critically reviewed by Baulch et al. in 1972²⁵ and by Cohen and Westberg in 1982.²⁶ For many third bodies, including $\text{M} = \text{H}_2$, He, and Ar, accurate measurements^{4,5} and accurate calculations¹⁷ are available and are in reasonable agreement. For $\text{M} = \text{H}$, there is a lack of reliable experimental data. Shock tube measurements at high temperatures have been performed but have not been very reproducible.^{25,26} At lower temperatures, only an upper limit to the rate coefficient at 300 K is available, which was obtained by Bennett and Blackmore.² This upper limit is not in

agreement with the theoretical result,^{12,17} and the experimental method has been questioned by Baulch et al.²⁵

The nascent vibrational/rotational product state distribution of recombined H_2 has never been measured. In this report, we present a calculation of this distribution using resonance complex theory. The theory and computational details are described in Section II. The total recombination rate coefficient is presented and compared to previous calculations in Section III. The nascent vibrational/rotational distribution is discussed, and rate coefficients into individual vibrational/rotational states as functions of temperature are presented.

II. COMPUTATIONAL DETAILS

A three-body recombination process can take place primarily via two separate channels: the "energy-transfer" mechanism



and the "exchange" or "chaperone" mechanism



where H_2^* and HM^* are intermediate (unbound) complexes that are in equilibrium. In the present case, where $\text{M} = \text{H}$, the two processes can be distinguished in a classical calculation; quantum mechanically these two channels are indistinguishable.

The resonance complex theory of Roberts, Bernstein, and Curtiss¹¹ is used to study the recombination process. Because this theory has been discussed in detail elsewhere, only a brief description follows. In this theory, the complex $[\text{H}_2^*]$ in Eq. (2)] is identified with orbiting resonance states of the H_2 system. These states are bound classically but have a finite lifetime quantum mechanically, because of tunneling. We assume that some of these states reach a steady-state population distribution determined by their equilibrium constant. Hence the reaction rate can be written as a sum over individual rate constants

$$k_r(v, J, T) = \sum_i k_r^i(v, J, T) \quad (4)$$

The individual rate coefficient for deactivation of an orbiting resonance i can be expressed as

$$k_r^i(v, J, T) = \bar{v}_r K_{eq}^i \bar{\sigma}_i^-(v, J, T) \quad (5)$$

where

$$\bar{v}_r = (8kT/\pi\mu)^{1/2} \quad (6)$$

K_{eq}^i is the equilibrium constant for the formation of the i^{th} resonance complex state, μ is the reduced mass for the (H_2M) system, and

$$\bar{\sigma}_i(v, J, T) = (kT)^{-2} \int_0^\infty E \sigma_i(v, J, E) \exp(-E/kT) dE \quad (7)$$

is the thermal average of the energy dependent, state-selective, cross section.

To calculate vibrational/rotational rate coefficients, the resonance states contributing to the hydrogen ($H + H + H$) recombination process must be determined. There are approximately 50 orbiting resonance states of H_2 .^{27,28} The procedure for assessing the contribution of each resonance state was discussed in the first calculation using resonance complex theory.¹¹ Most of these states can be eliminated by simple energy considerations because the equilibrium constant contains a factor of $\exp(-E/kT)$. The majority of the remaining states (about 15) have lifetimes much too long (i.e., too narrow in resonance width) to contribute to the recombination. The result is 6 possible contributing orbiting resonance states. The properties of these states are summarized in Table 1. The literature includes discussions about which of these 6 resonance states contributes to recombination under various experimental conditions.^{6,8} We decided to calculate contributions from all 6 states. If it is determined that, under certain experimental conditions, some of the states do not contribute or are not in complete thermal equilibrium, they could then easily be excluded from the final calculation of the rate coefficient.

Table 1. Properties of Six Resonance States in Recombination Calculation

v	J	$E_i (\text{cm}^{-1})$	$\langle R \rangle (\text{au})$	$\tau_i (\text{sec})$
14	5	45.7	6.51	2.7×10^{-13}
14	4	3.6	6.05	4.0×10^{-10}
13	8	89.8	5.31	2.0×10^{-12}
12	11	216.0	4.86	1.8×10^{-12}
12	12	387.0	5.30	7.0×10^{-14}
11	13	199.5	4.27	2.0×10^{-9}

Two of the resonance states require an additional approximation. The $v = 14$, $J = 5$ and the $v = 12$, $J = 12$ states are classically unbound. In order to carry out classical trajectories with these initial states, it is therefore necessary to select them to be slightly classically bound, that is, with a slightly lower internal energy. The vibrational quantum number defined by²⁹

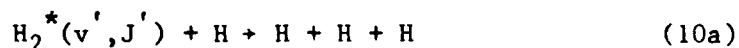
$$v = -1/2 + (1/\pi\hbar) \int_{R_-}^{R_+} \{2m[E_{\text{int}} - V(R) - (J_r J_r / 2mR^2)]\}^{1/2} dR \quad (8)$$

where $R_{\pm}$ are the diatomic turning points, E_{int} is the internal energy, $V(R)$ is the diatomic potential energy, m is the diatomic reduced mass, and J_r is related to the rotational quantum number by

$$J_r^2 = j(j+1)\hbar^2 \quad (9)$$

then takes on noninteger values. The "vibrational quantum number" for the $v = 14$ resonance state is $v = 13.85$ and for the $v = 12$ state is $v = 11.72$. To assess the effects of this change, calculations are run for not only these two states but also for other nearby (E_{int} slightly different) states. No effect is found either on the total cross section or on the final vibrational/rotational state distribution.

Once the resonance state is selected, quasiclassical trajectories are calculated for the process



to converge $\sigma_1(v, J, E)$. A similar calculation with $M = \text{H}, \text{He}, \text{Ar}$, and H_2 was performed by Whitlock et al.¹⁷ for the total recombination rate coefficient. The present calculation follows the same basic procedure. However, several changes were needed to determine the vibrational/rotational distribution of products. Classical trajectories on the $(\text{H})_3$ potential energy surface used by Whitlock et al.¹⁷ were calculated using CLASTR.³⁰ We discovered that the surface supports many bound vibrational/rotational states that are unbound for the true H_2 system. A highly accurate surface for H_3 is available, the

SLTH^{31,32} surface. Therefore, we modified CLASTR to accept this surface and to determine the final vibrational and rotational state quantum number for the SLTH surface. We constructed the correct initial conditions for the trajectories based on the orbiting resonance state and calculated the final vibrational and rotational quantum numbers. The procedure is similar to the one described by Truhlar and Muckerman²⁹ and will not be reviewed. Because of the highly excited states encountered for both the initial and final conditions, we made one modification to their described procedure. The Newton-Raphson iteration technique, to locate the turning points of the effective potential, was replaced with the method of Pade approximates.

After the individual vibrational/rotational cross section has been calculated as a function of E, it is then fitted to the form

$$\sigma_i(E, v, J) = b(E/k)^{-1/2} \exp[-a(E/k)^{-1}] \quad (11)$$

as suggested by Whitlock et al. After thermal averaging to determine $\bar{\sigma}_i(v, J, T)$, the cross sections are combined to produce the rate coefficient for the desired vibrational/rotational state.

For each of the six resonance states, five energies are calculated ($E/k = 50, 100, 300, 1000, \text{ and } 2000 \text{ K}$). This is found to be adequate to calculate the thermally averaged cross section. For each energy and initial resonance state, approximately 1000 trajectories were run, producing a Monte Carlo error of less than 5% for the total cross section, of approximately 10% for the summed vibrational cross sections, and of approximately 20% for the individual vibrational/rotational cross sections.

III. RESULTS

The individual final state vibrational/rotational probabilities for one initial resonance state, $v = 13$, $J = 8$, are shown for $E/k = 50$ K in Fig. 1 and for $E/k = 1000$ K in Fig. 2. The qualitative features are the same for all the resonance states. The dissociation level for the hydrogen molecule is shown by the heavy line in the figures. The figures indicate that the distribution is peaked near this line, that is, in the highest bound states of the H_2 manifold. The distribution broadens and extends to lower vibrational states as the collision energy increases. The total reaction probability decreases with increasing energy. Figure 3 exhibits the individual vibrational cross sections for the same intermediate state as functions of collision energy. Again, the qualitative features are the same for all resonance states. In this figure, the widening of the vibrational distribution and the falloff of the total cross section with increasing collision energy can be seen clearly. (Space considerations preclude the display of all the individual vibrational/rotational cross sections as functions of energy. The complete results are available on request.) The nascent vibrational/rotational distribution is the correctly weighted sum over the individual orbiting resonance states.

In Fig. 4, the results for the total rate coefficient as a function of temperature are plotted and compared with the calculation of Whitlock et al.¹⁷ The two data sets reveal the same qualitative behavior with temperature. However, the present results are approximately a factor of 2 smaller than the previous results. This difference occurs because the potential energy surface used by Whitlock et al.¹⁷ binds too many vibrational/rotational states, as discussed previously. In Fig. 5, the rate coefficients into individual vibrational states are plotted and compared with the total rate coefficient and with the sum over the plotted states. This figure indicates that $v = 14$, $v = 13$, $v = 12$, and $v = 11$ make up the largest contribution to the rate coefficient. Figure 5 also exhibits the broadening of the distribution with energy, with $v = 14$ dominating the rate constant at lower temperatures but becoming overshadowed by $v = 13$ at higher temperatures. Figure 6 displays the rotational rate coefficients for $v = 14$. The higher rotational levels

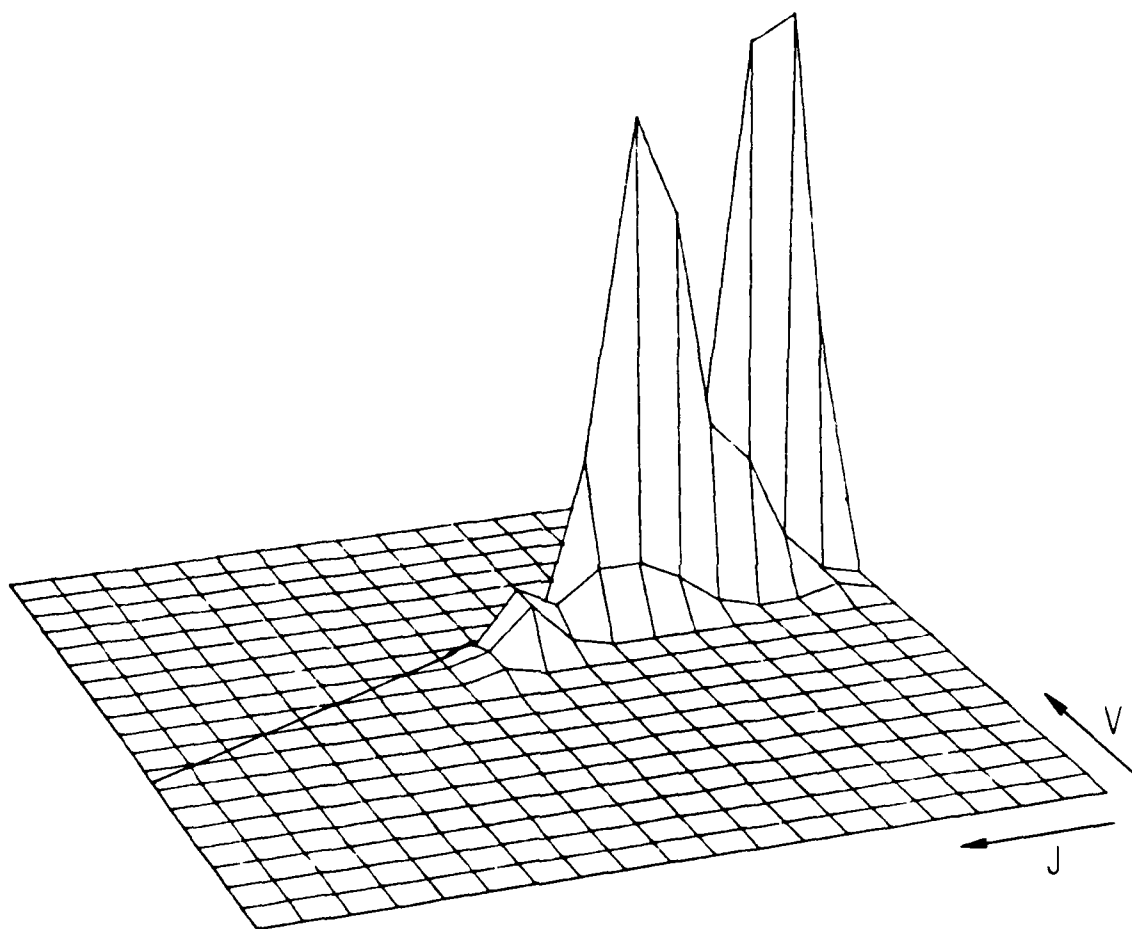


Fig. 1. Probabilities for Final Vibrational/Rotational State Formed from Initial Resonance State $v = 13, J = 8$ for Initial $E/k = 50$ K

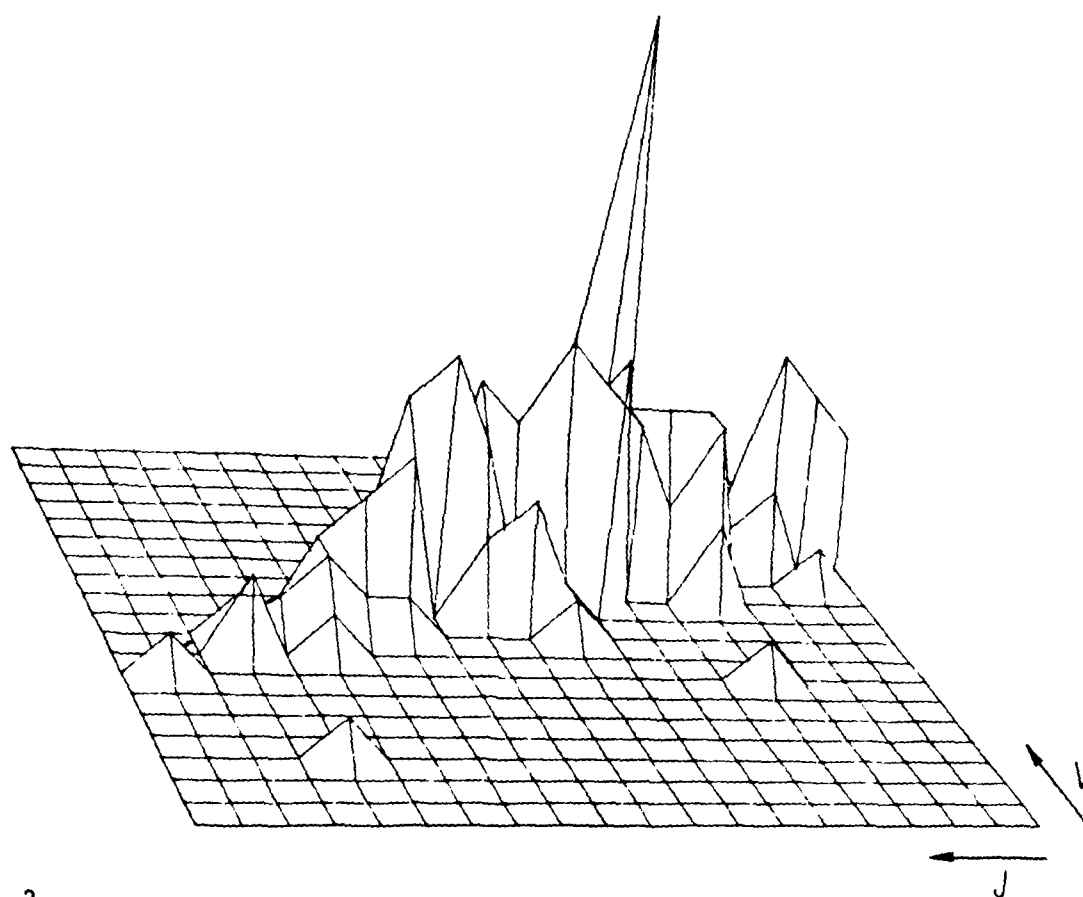


Fig. 2. Probabilities for Final Vibrational/Rotational State Formed from Initial Resonance State $v = 13$, $J = 8$ for Initial $E/k = 1000$ K

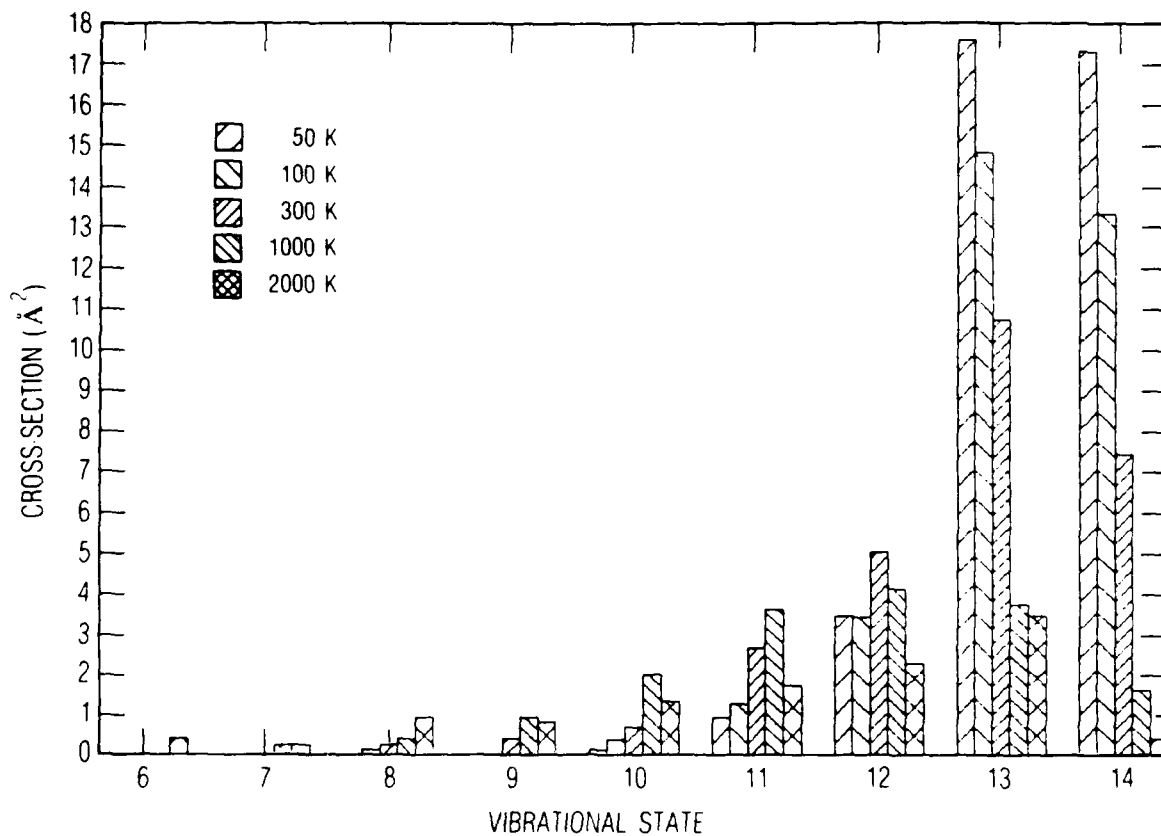


Fig. 3. Vibrational Cross Sections Formed from Initial Resonance State $v = 13$, $J = 8$

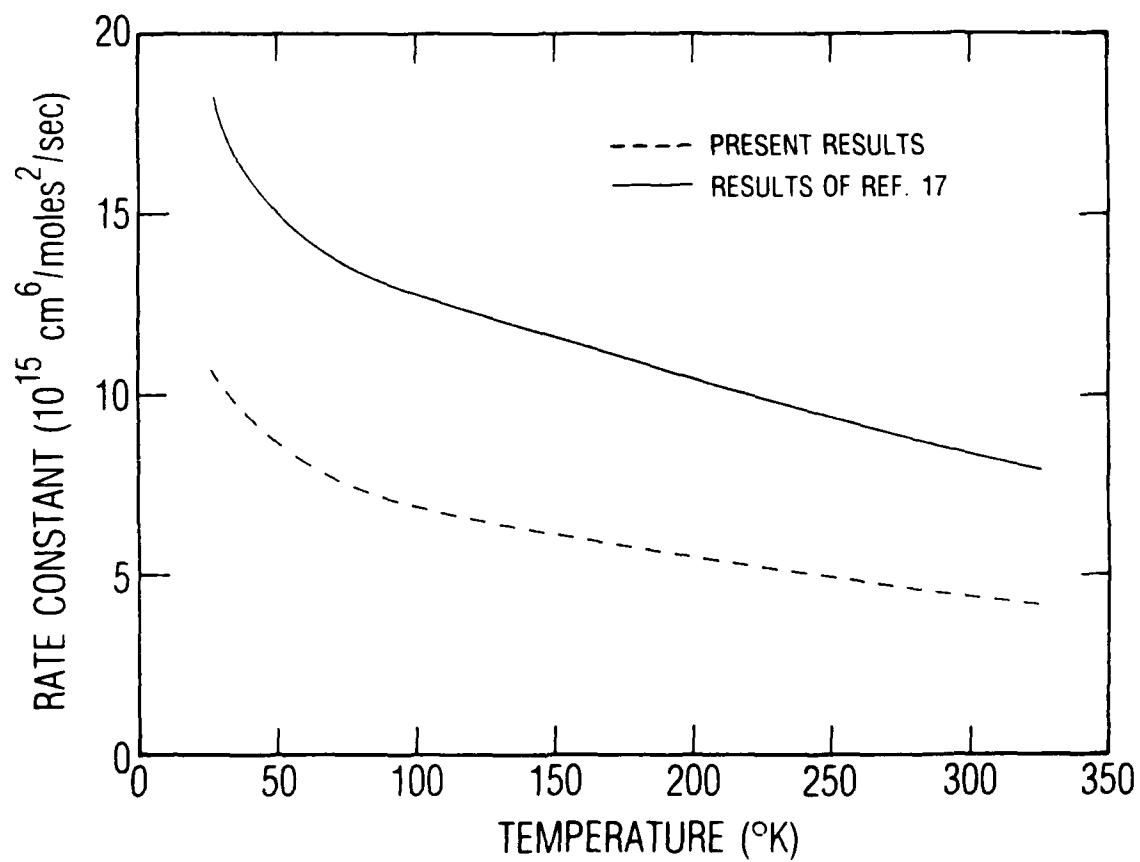


Fig. 4. Total Recombination Rate Constant as a Function of Temperature

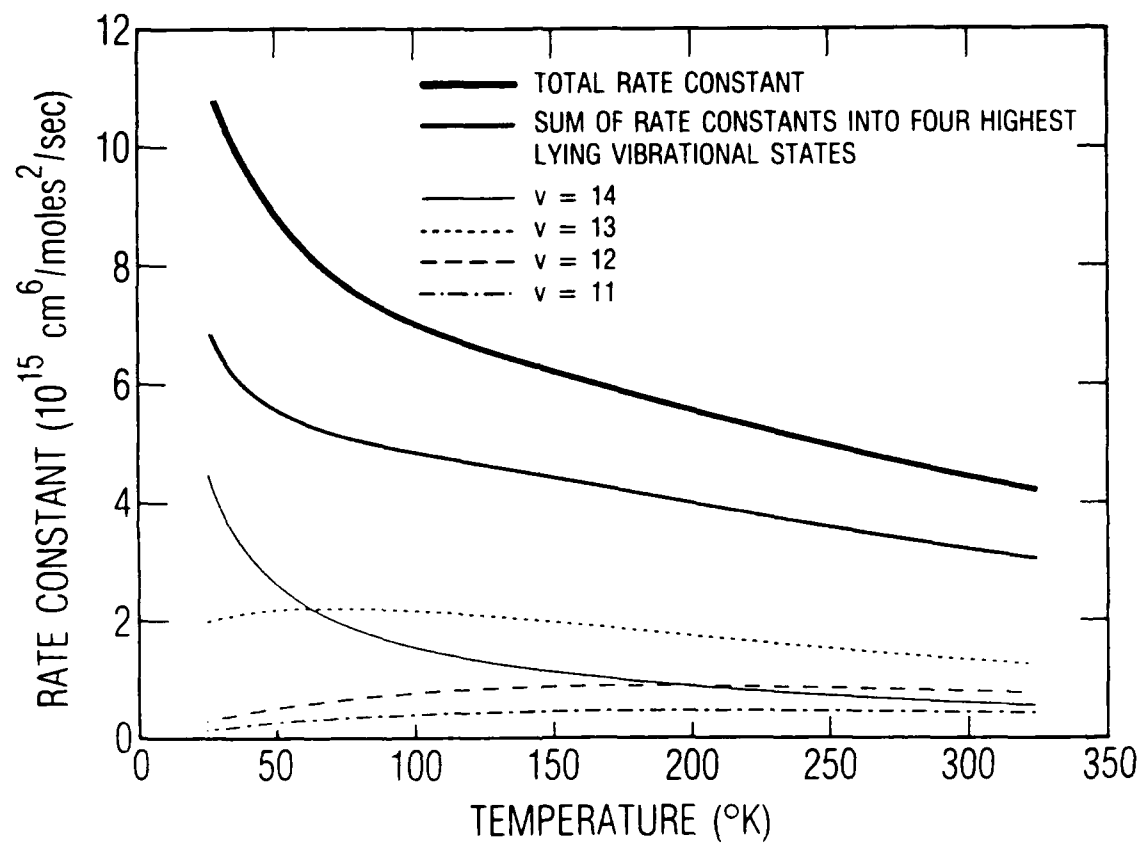


Fig. 5. Rate Constants into Given Final Vibrational State as a Function of Temperature

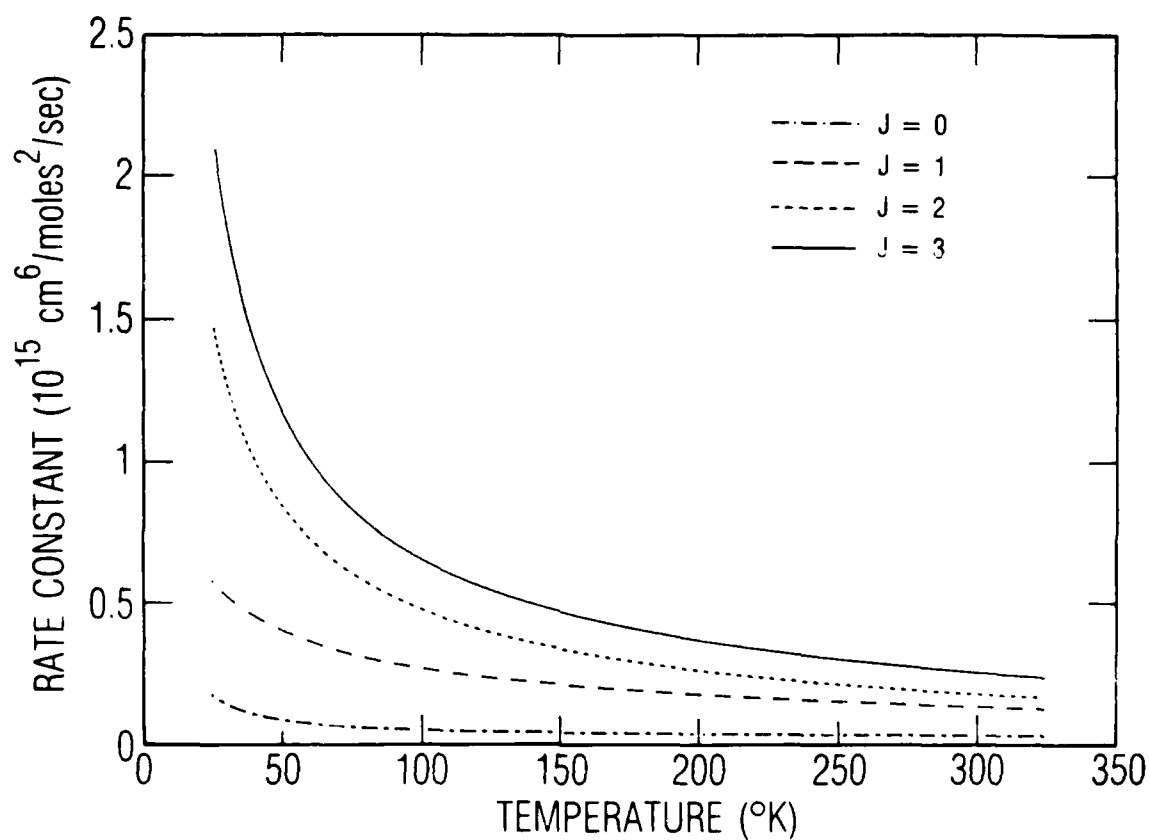


Fig. 6. Rate Constants into Given Final Rotational State for Final $v = 14$ as a Function of Temperature

dominate; $J = 3$ is the highest bound state for $v = 14$. Figure 7 exhibits the rotational rate coefficients for $v = 13$. Again, the higher rotational states dominate, with the highest four states constituting approximately 80% of the total rate coefficients. ($J = 7$ is the highest bound state for $v = 13$.)

These results indicate that the nascent vibrational/rotational distribution peaks in the highest bound states of the hydrogen molecule. The average vibrational level is high. Because such high vibrational states can only support a small number of bound rotational states, the average rotational level is small.

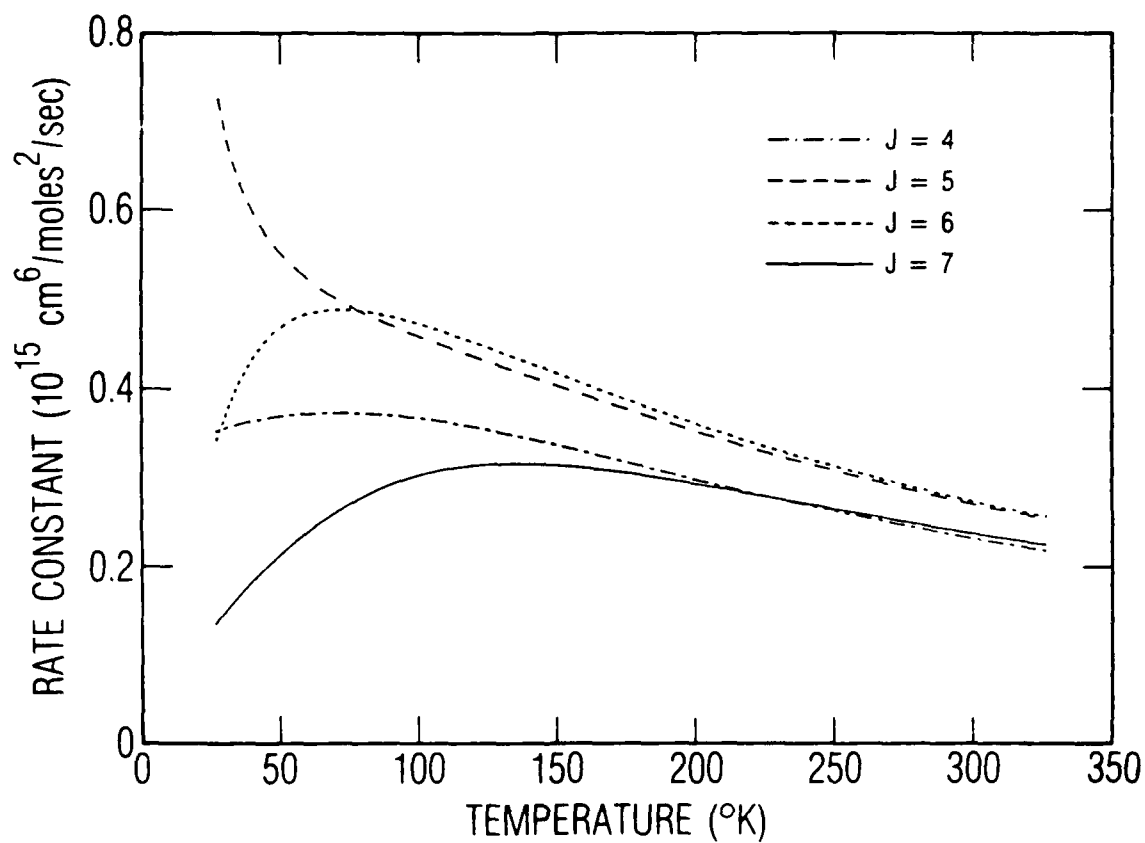


Fig. 7. Rate Constants into Given Final Rotational State for Final $v = 13$ as a Function of Temperature

IV. CONCLUSIONS

We have used resonance complex theory to investigate the nascent vibrational/rotational state distribution produced by hydrogen atom recombination. We have shown that this distribution peaks in the highest bound states of the hydrogen molecule. Because of the highly excited nature of the products, the nascent distribution is greatly changed by subsequent collisions. To assess the effect of a collision on the distribution, calculations must be performed to determine the rate constant for dissociation and relaxation from these states. This is particularly important when inert third bodies such as helium are considered. The measured hydrogen ortho/para ratio in such systems may not be the nascent distribution because of the effect of vibrationally enhanced exchange reactions.

This state distribution was calculated using the approximations of quasi-classical theory. Though a correct quantum-mechanical state was used for initial conditions, the subsequent collision was treated using classical mechanics. As has been previously discussed, classical results are more accurate the greater the degree of averaging in the calculation.²⁹ Therefore we would expect the total rate coefficient to have the greatest accuracy, the vibrational rate constants to be less accurate, and the individual vibrational/rotational rate constants to be the least accurate. The qualitative features of the distribution should be correct, that is, the distribution is almost certainly peaked in the highest states of the bound hydrogen molecule.

Future calculations will address the effects of a collision on the nascent distributions and will investigate the nascent vibrational/rotational distribution produced with an inert third body such as helium. We hope that the results will be more readily subject to experimental test.

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