Title

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October 1971

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POTENTIAL ENERGY SURFACE INCLUDING ELECTRON CORRELATION FOR THE CHEMICAL REACTION $F + H_2 \rightarrow FH + H$.

I. PRELIMINARY SURFACE*

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ABSTRACT

Rigorous quantum mechanical calculations have been carried out for about 150 linear and 200 nonlinear geometries for the FH_2 system. The contracted gaussian basis set used consisted of four s and two p functions on fluorine and two s functions on hydrogen. The barrier height and exothermicity are poorly predicted by single configuration self-consistent-field calculations. However, the 214 configuration correlated results are in qualitative agreement with experiment (low barrier height and substantial exothermicity). The reaction coordinate is discussed and pictures of the potential surface are presented. A second series of calculations is being carried out with a larger basis set. These latter calculations yield nearly quantitative agreement with experiment for both the barrier height and exothermicity.

INTRODUCTION

Recent ab initio electronic structure calculations on diatomic molecules have shown that by the inclusion of electron correlation, potential energy curves of chemical accuracy can now be obtained.[1,2] However, a recent review[3] of quantum mechanical potential energy surfaces indicates that such accurate calculations for polyatomic systems have only been completed for relatively simple systems such as H_3[4,5] and H_3^+.[6] In this report we present the results of a series of calculations, including electron correlation, on the chemical reaction $F + H_2 \rightarrow FH + H$. This reaction is of particular interest since it has been studied experimentally by laser spectroscopy,[7] infrared chemiluminescence,[8] and crossed molecular beams.[9]

BASIS SET

The basis set used in this work was of the double-zeta variety, namely two 1s, 2s and 2p functions on fluorine and two 1s functions on each hydrogen atom. These functions were obtained by Dunning[10] through optimum contraction of Huzinaga's fluorine 9s 5p and hydrogen 4s set of primitive gaussian functions.[11] It is well known from extensive calculations[1,2] on diatomic molecules that additional functions, polarization functions (d functions on F and p functions on H), are required in order to obtain accurate dissociation energies. However, the basis set described here is probably the largest that can be currently used to describe chemical reactions involving larger molecules. Thus it is of interest to ascertain whether this relatively modest basis set is of practical value. In a later section we will briefly discuss results obtained with a larger basis.

TYPE OF CONFIGURATION INTERACTION (CI)

As a first step, self-consistent-field (SCF) calculations[12] were carried out at each geometry. The SCF configuration for linear geometries is

$$1\sigma^2\ 2\sigma^2\ 3\sigma^2\ 4\sigma\ 1\pi^4 \quad ,$$

while for non-linear geometries (C_s symmetry) we have

$$1a'^2\ 2a'^2\ 3a'^2\ 4a'^2\ 5a'\ 1a''^2 \quad .$$

Electron correlation was explicitly considered using multiconfiguration first-order wave functions.[2,13] When adequate basis sets are used, first-order wave functions have been shown[2,14] to yield reliable dissociation energies for a variety of diatomic molecules. This type of wave function[2] includes only that part of the correlation energy due to low-lying valence orbitals not occupied in the single-configuration SCF wave function. For the FH_2 system these orbitals can be thought of as the sixth (unoccupied) F atom 2p spinorbital and the $1\sigma_u$ orbital of H_2. More precisely, for C_s geometry, the valence orbitals not fully occupied in the SCF wave function are 5a' and 6a'. In all configurations (except single excitations) the lowest orbital (essentially the F 1s orbital) was held doubly occupied. With this restriction, for general geometry (C_s symmetry) our first-order wave functions included 214 configurations. These configurations are indicated in Table I.

The first-order wave function will provide a reasonable description of the FH_2 potential surface only if the orbitals as well as the 214 CI expansion coefficients are optimized. The difficult problem of obtaining a nearly optimum set of molecular orbitals was solved, beginning with the SCF wave functions, using the iterative natural orbital procedure.[15] Typically 4 or 5 natural orbital iterations were required to obtain convergence in the CI energy.

GEOMETRIES CONSIDERED

The first ab initio calculations on the $F + H_2$ system were those of Newton.[16] He carried out SCF calculations (with a small basis set) which seemed to indicate clearly that the least energy path or reaction coordinate occurs for the linear arrangement F-H-H.

Since a primary purpose of our research was to assess the accuracy of our theoretical surface by comparison with the experimental activation energy and exothermicity, special emphasis was placed on linear FH_2. Figure 1 shows the FH_2 coordinate system adopted. Calculations were carried out for about 150 linear ($\theta = 0$ in Fig. 1) geometries. For each of the angles $\theta = 10, 30, 50, 70$, and 90°, about 40 additional calculations were carried out. Thus about 350 calculations were carried out in all.

TOTAL ENERGIES

The total energies obtained in our ~350 calculations are not reported here. This decision was made to discourage the use of the present potential surface for quantum dynamical calculations, e.g., classical trajectories. The present "first generation" potential surface is in fact not sufficiently realistic for such purposes. However, our "second generation" FH_2 surface (to be described briefly in a later section) does appear to approach the accuracy required for dynamical calculations.

It may be useful to quote a few total energies for reference. For H-H separation 1.4 bohrs and F atom at infinite separation, our calculated SCF and CI energies are -100.5208 and -100.5425 hartrees. Similarly for F-H separation 1.7 bohrs and the second H atom at infinite separation, the SCF and CI energies are -100.5191 and -100.5726 hartrees.

LINEAR RESULTS--THE MINIMUM ENERGY PATH

The essential results of our linear calculations are seen in Table II. The SCF barrier height and exothermicity are ∿34.3 and -0.6 kcal/mole, in poor agreement with the experimental activation energy, 1.7 kcal/mole,[17] and exothermicity +31.2 kcal/mole.[18] The CI results ∿5.7 and +20.4 are in qualitative agreement with experiment; that is, the barrier is predicted to be small and the reaction significantly exothermic. As will be seen later, most of remaining discrepancy with experiment is due to the lack of polarization functions (d functions on F and p functions of H) in our basis set.

It should of course be remembered that the barrier height (highest point on the minimum energy path) and activation energy E_a (obtained from kinetic data via $k = Ze^{-E_a/RT}$) are not the same quantity.[19] A simple analysis by Muckerman[20] suggests that the true barrier height for the F + H_2 reaction is about 1 kcal/mole.

Perhaps the most valuable information obtained from our calculations regards the position of the saddle point and general shape of the surface. The SCF saddle point occurs near an F-H distance of 2.01 bohrs (1.06 Å) for H-H distance of 1.54 bohrs (0.81 Å), while the 214 configuration calculation predicts 2.58 and 1.54 bohrs (1.37 and 0.81 Å). For comparison the isolated diatomic bond distances are 0.92 and 0.74 Å. Thus both calculations predict the saddle point to occur rather early (significantly lengthened F-H distance) for a slightly expanded H-H distance.

The position of the saddle point is not a quantity that can be measured by current experimental methods. However, experimental data for the F + H_2 reaction seems most consistent with a potential surface with saddle point occurring early in the entrance valley.[8,21] This experimental "prediction" is for the most part consistent with our theoretical saddle point position. In addition, it should be noted that two semi-empirical potential surfaces recently used for classical trajectory studies[20,22] also display an early saddle point.

The minimum energy path or reaction coordinate is usually defined with respect to the saddle point. For linear FH_2, if x is the F-H distance and y the H-H distance, the saddle point has the property

$$\frac{\partial V}{\partial x} = \frac{\partial V}{\partial y} = 0 \quad ,$$

where V is the potential energy. Given the saddle point, the minimum energy path may be found by following the negative of the gradient

$$-\vec{\nabla} V = \frac{-\partial V}{\partial x} \vec{i} - \frac{\partial V}{\partial y} \vec{j} \quad .$$

Table III shows the linear minimum energy path for the SCF and CI potential surfaces.

The minimum energy path (Table III) divides the surface into two regions. In the first, a very gentle uphill grade is traversed as the F atom approaches the nearly unperturbed H_2 molecule. At R(F-H) = 2.7 bohrs the H-H separation is only 1.49 bohrs. The second region of the surface is the downhill part in which simultaneously a) the R(F-H) is decreased until it reaches r_e, b) the H_2 molecule separates, and c) the energy goes monotonically downward to that of the products, HF + H.

The <u>ab initio</u> surface is of the type often referred to[23] as "highly repulsive." Evans and Polanyi[24] long ago argued that this type of surface would convert the exothermicity to translational, rather than vibrational, product energy. However, classical trajectory studies by Polanyi and coworkers[23] have shown that a highly repulsive surface usually leads to "mixed energy release," resulting in considerable vibrational excitation of the products (FH in the present case). In this light, our highly repulsive surface is consistent with experimental findings[7-9] for the F + H_2 reaction, namely that the HF molecule is frequently formed in an excited vibrational state.

Table III shows the prediction by both calculations of a small long-range attraction (∿0.2 kcal/mole) between FH and H. For FH separation 1.7 bohrs, the minimum for this attraction occurs at H-H distance 5.08 bohrs. A similar attraction was found in the SCF calculations of Lester and Krauss[25] for the interaction between Li and HF. This type of attraction is also seen in the recent semi-empirical calculations of Raff *et al.*[26] for the IH_2 surface.

The linear *ab initio* energies have been fit to an analytic form involving exponentials and powers of r, yielding an rms error of slightly less than 1/2 kcal/mole. Figures 2 and 3 show three-dimensional plots of the electronic entry and exit channels, while Figures 4 and 5 show an overall view of the surface and a traditional contour map. It is hoped that these plots will provide a useful physical picture of the linear potential energy surface.

ANISOTROPY OF THE POTENTIAL SURFACE

Another interesting feature of a potential surface is its anisotropy, or behavior as a function of angle. The 200 nonlinear FH_2 calculations discussed above were carried out for the purpose of characterizing the anisotropy.

Table IV gives the barrier heights and saddle point positions for approaches of $\theta = 10°$, $30°$, $50°$, $70°$, and $90°$. This table verifies Newton's earlier conclusion[16] that the true minimum energy path is linear. However, note that the $\theta = 10°$ approach has a barrier height only 0.02 kcal above the linear approach. Note also that the $\theta = 90°$ approach is quite unfavorable energetically. Another interesting feature of Table IV is that the FH distance at the saddle point decreases significantly as θ goes from 0 to 90°. Simultaneously the HH distance becomes longer as the angle of approach goes to 90°.

More information on the anisotropy of the calculated surface is given in Table V. There the calculated energies as a function of angle are given for R(FH) = 3.0. The HH distance in these calculations is 1.46 bohrs, that specified by the minimum energy path from the linear CI calculation.

Table V shows that the 214 configuration calculations predict a small force constant for bending. In fact the CI energy for the 30° approach is only 0.04 kcal above the linear. The SCF energy rises much more rapidly as a function of bending angle. This is consistent with our general observation that the CI surface is much smoother than the SCF surface.

Finally the CI miminum energy path for the 90° approach is shown in Table VI. Comparison with the 0° approach (Table III) emphasizes that the downhill part of the surface occurs much "later" for $\theta = 90°$.

DISCUSSION

It should be emphasized that configuration interaction calculations of the type reported here are not excessively time consuming using our computational methods. For each point on the potential surface, a complete calculation requires about three minutes of CDC 6600 computer time. Analogous calculations are currently practical for any A + H_2 chemical reaction, if A is no larger than chlorine or argon. In addition potential surfaces including electron correlation may be obtained for H + AB reactions, if A and B are restricted to atoms smaller than sodium. Our recent work[27] on the HO_2 radical illustrates this point.

As mentioned above, the present potential surface does not predict the barrier height and exothermicity accurately enough to be useful for scattering calculations. For this reason, a series of calculations using a larger basis set (including d functions on F and p functions on H) are underway. These calculations predict a barrier height of 1.69 kcal and exothermicity of 34.4 kcal, both in good agreement with experiment.

It is important to note that the minimum energy path predicted by these extended calculations is qualitatively similar to that seen in Table III. Thus it seems likely that calculations such as those reported here (using modest basis sets) will be useful in evaluating qualitative features of chemical reactions, i.e. reaction coordinates.

This study shows clearly that the energetic aspects of a chemical reaction are the most difficult to describe *ab initio*. The Hartree-Fock approximation does not appear to yield reasonable results in this regard, since the correlation energies for the reactants, transition state, and products may be quite different.

In addition, an extended basis set including polarization functions is needed to reliably predict barrier heights and exothermicities.

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FOOTNOTES AND REFERENCES

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Table I. Configurations included in the first-order wave functions for FH$_2$, C$_s$ geometry. In some cases, only those configurations i having nonvanishing matrix element H$_{1i}$ with the SCF configuration were included (see C. F. Bender and H. F. Schaefer, J. Chem. Phys. 55, 0000 (1971)). na' refers to the 7a', 8a', ... orbitals, while na'' refers to 2a'', 3a'', ...

Excitations	$^2A'$ Number of Configurations Included Per Orbital Occupancy
$1a'^2\ 2a'^2\ 3a'^2\ 4a'^2\ 5a'\ 1a''^2$	1
$1a', 2a', 3a', 4a' \rightarrow 5a'$	1
$1a', 2a', 3a', 4a' \rightarrow 6a'$	2
$1a', 2a', 3a', 4a' \rightarrow na'$	2
$5a' \rightarrow 6a'$	1
$5a' \rightarrow na'$	1
$1a'' \rightarrow na''$	2
$2a'^2, 3a'^2, 4a'^2 \rightarrow 5a'\ 6a'$	1
$2a'^2, 3a'^2, 4a'^2 \rightarrow 6a'^2$	1
$2a'\ 3a', 2a'\ 4a', 3a'\ 4a' \rightarrow 5a'\ 6a'$	2
$2a'\ 3a', 2a'\ 4a', 3a'\ 4a' \rightarrow 6a'^2$	2
$2a'\ 5a', 3a'\ 5a', 4a'\ 5a' \rightarrow 6a'^2$	1
$1a''^2 \rightarrow 5a'\ 6a'$	1
$1a''^2 \rightarrow 6a'^2$	1

(Continued)

Table I. Continued.

Excitations	$^2A'$ Number of Configurations Included Per Orbital Occupancy
$3a'^2,\ 4a'^2 \rightarrow 5a'\ na'$	1
$3a'^2,\ 4a'^2 \rightarrow 6a'\ na'$	2
$3a'\ 4a' \rightarrow 5a'\ na'$	2
$3a'\ 4a' \rightarrow 6a'\ na'$	2
$3a'\ 5a',\ 4a'\ 5a' \rightarrow 6a'\ na'$	2
$1a''^2 \rightarrow 5a'\ na'$	1
$1a''^2 \rightarrow 6a'\ na'$	2
$3a'\ 1a'',\ 4a'\ 1a'' \rightarrow 5a'\ na''$	2
$3a'\ 1a'',\ 4a'\ 1a'' \rightarrow 6a'\ na''$	2
$5a'\ 1a'' \rightarrow 6a'\ na''$	2
$2a'^2\ 3a' \rightarrow 5a'\ 6a'^2$	1
$2a'^2\ 4a' \rightarrow 5a'\ 6a'^2$	1
$2a'\ 3a'^2 \rightarrow 5a'\ 6a'^2$	1
$2a'\ 4a'^2 \rightarrow 5a'\ 6a'^2$	1
$2a'\ 1a''^2 \rightarrow 5a'\ 6a'^2$	1
$3a'^2\ 4a' \rightarrow 5a'\ 6a'^2$	1
$3a'\ 4a'^2 \rightarrow 5a'\ 6a'^2$	1
$3a'\ 1a''^2 \rightarrow 5a'\ 6a'^2$	1
$4a'\ 1a''^2 \rightarrow 5a'\ 6a'^2$	1

Table II. Summary of self-consistent-field (SCF) and configuration interaction (CI) results for the F + H_2 reaction.

	SCF	CI	Experiment
Barrier Height (kcal/mole)	34.3	5.7	1.7[a]
Exothermicity (kcal/mole)	-0.58	20.4	31.2[b]
Saddle Point Geometry (Å)			
F-H	1.06	1.37	--
H-H	0.81	0.81	--

[a] Experimental activation energy, reference 17.

[b] Reference 18.

Table III. Minimum energy paths for the $F + H_2 \rightarrow FH + H$ reaction. Internuclear separations are given in bohrs and energies in kcal/mole relative to the reactants. Saddle points are indicated by asterisks.

Self-Consistent-Field			214 Configurations		
R(FH)	R(HH)	Energy	R(FH)	R(HH)	Energy
6.0	1.380	0.03	6.0	1.425	0.00
4.0	1.381	1.48	4.0	1.429	0.82
3.5	1.401	3.90	3.5	1.439	2.09
3.0	1.397	10.07	3.0	1.459	4.22
2.8	1.403	14.33	2.9	1.470	4.73
2.6	1.428	20.04	2.8	1.484	5.20
2.4	1.466	27.41	2.7	1.490	5.48
2.2	1.502	32.40			
			2.58	1.54	5.72*
2.01	1.54	34.33*			
			2.412	1.6	5.41
1.930	1.7	29.11	2.229	1.7	3.36
1.890	1.8	25.99	2.095	1.8	0.22
1.845	1.9	21.91	2.005	1.9	-2.87
1.811	2.0	18.40	1.931	2.0	-5.67
1.792	2.1	15.41	1.874	2.1	-8.14
1.782	2.2	12.93	1.840	2.2	-10.17
1.776	2.3	10.88	1.826	2.3	-11.83
1.771	2.4	9.16	1.817	2.4	-13.24
1.768	2.5	7.72	1.811	2.5	-14.46
1.750	3.0	3.12	1.809	3.0	-18.12
1.743	4.0	0.53	1.799	4.0	-20.43
1.742	5.0	0.37	1.798	5.0	-20.61
1.742	10.0	0.58	1.798	10.0	-20.40

Table IV. Saddle point geometries and energies (relative to $F + H_2$) for different angles θ of approach.

θ	R(F-H)	R(H-H)	Energy
0°	2.58	1.54	5.72
10°	2.55	1.54	5.74
30°	2.50	1.56	6.08
50°	2.44	1.60	7.31
70°	2.39	1.68	10.43
90°	2.35	1.87	17.52

Table V. Angular dependence of the F + H_2 potential surfaces for R(FH) = 3.0 bohrs and R(HH) = 1.46 bohrs. Energies are given in kcal/mole relative to F and H_2 at infinite separation.

θ (degrees)	E(SCF)	E(CI)
0	10.22	4.08
10	10.25	4.06
30	10.57	4.12
50	11.38	4.55
70	13.21	5.58
90	17.18	7.69

Table VI. Minimum energy path for the 90° approach of F to H_2. All points refer to the 214 configuration first-order calculations.

R(FH)	R(HH)	Energy
6.0	1.43	0.00
4.0	1.43	1.41
3.5	1.44	3.56
3.0	1.47	8.00
2.8	1.53	10.78
2.6	1.64	14.54
2.4	1.78	17.23
2.35	1.87	17.52*
2.20	2.1	14.58
2.06	2.2	10.76
1.99	2.3	6.97
1.94	2.4	3.62
1.86	2.6	-2.27
1.82	3.0	-10.36
1.80	3.5	-15.94
1.80	4.0	-18.70
1.80	6.0	-20.45
1.80	10.0	-20.40

FIGURE CAPTIONS

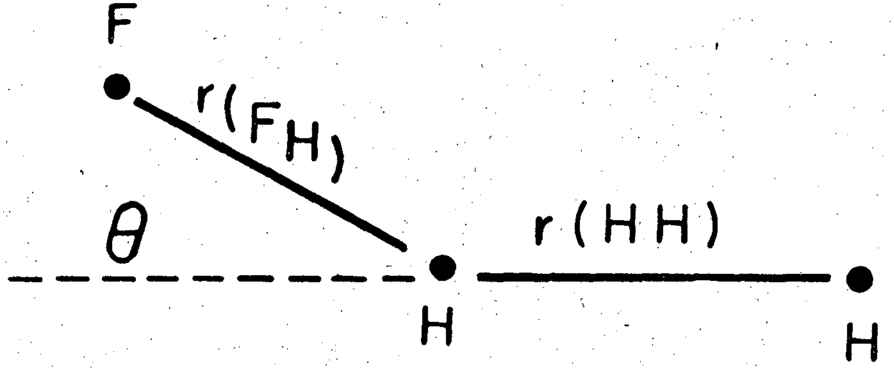
Fig. 1. Coordinate system describing F + H_2.

Fig. 2. Plot of the electronic energy of linear FH_2 as a function of F-H and H-H distances. Each small section bounded by four sides represents a square region in space 0.075 bohrs on a side. Thus the F-H distances ranges from 1.4 to 8.0 bohrs and the H-H distance from 1.0 to 7.6 bohrs.

Fig. 3. Same as 2 but viewed from the exit channel FH + H.

Fig. 4. A view of the FH_2 surface from the three atom region.

Fig. 5. Traditional contour map of the FH_2 surface. As in the three-dimensional plots, the F-H distance goes from 1.4 to 8.0 bohrs and the H-H distance from 1.0 to 7.6 bohrs.



XBL7110-4543

Fig. 1

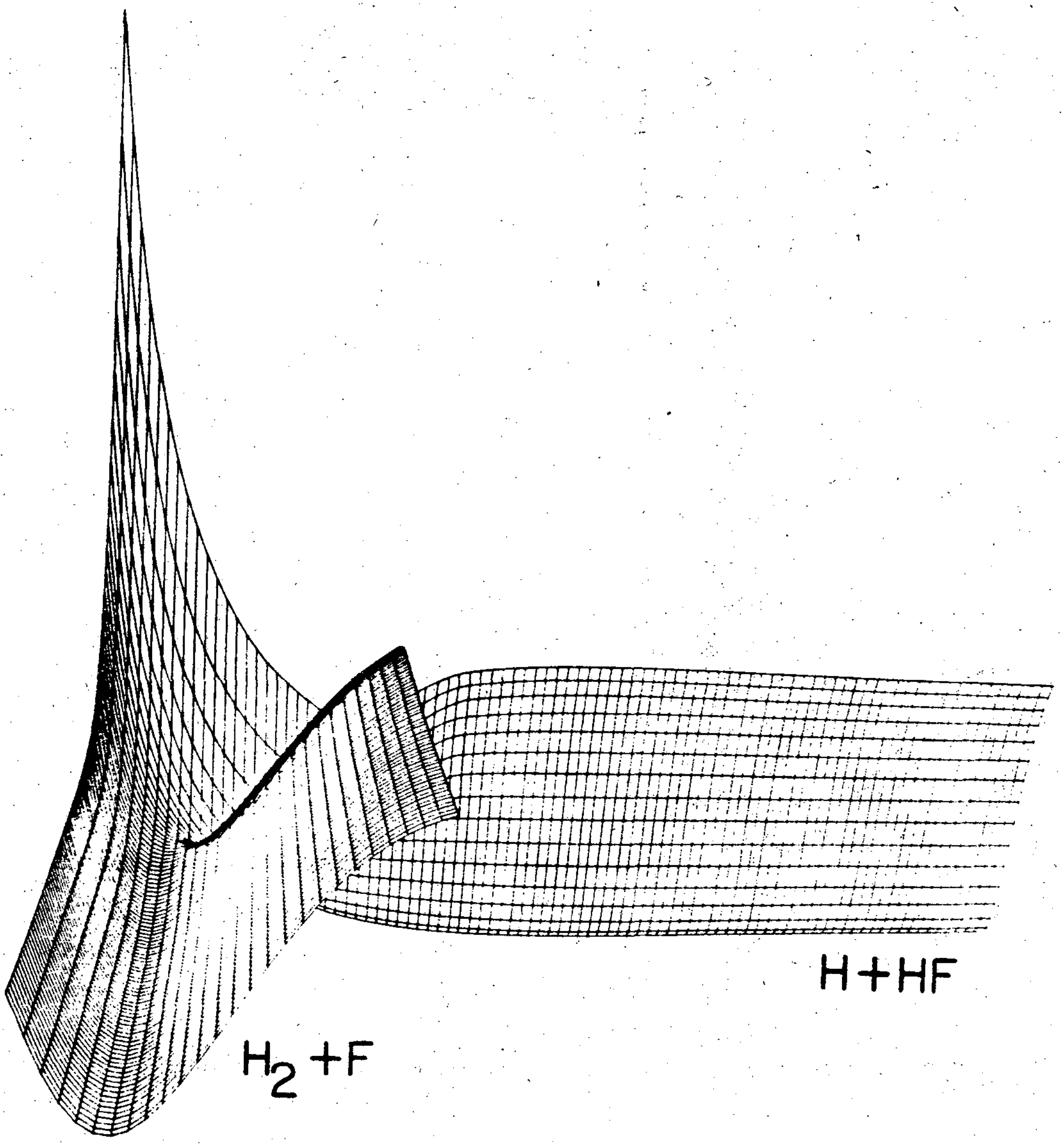


Fig. 2

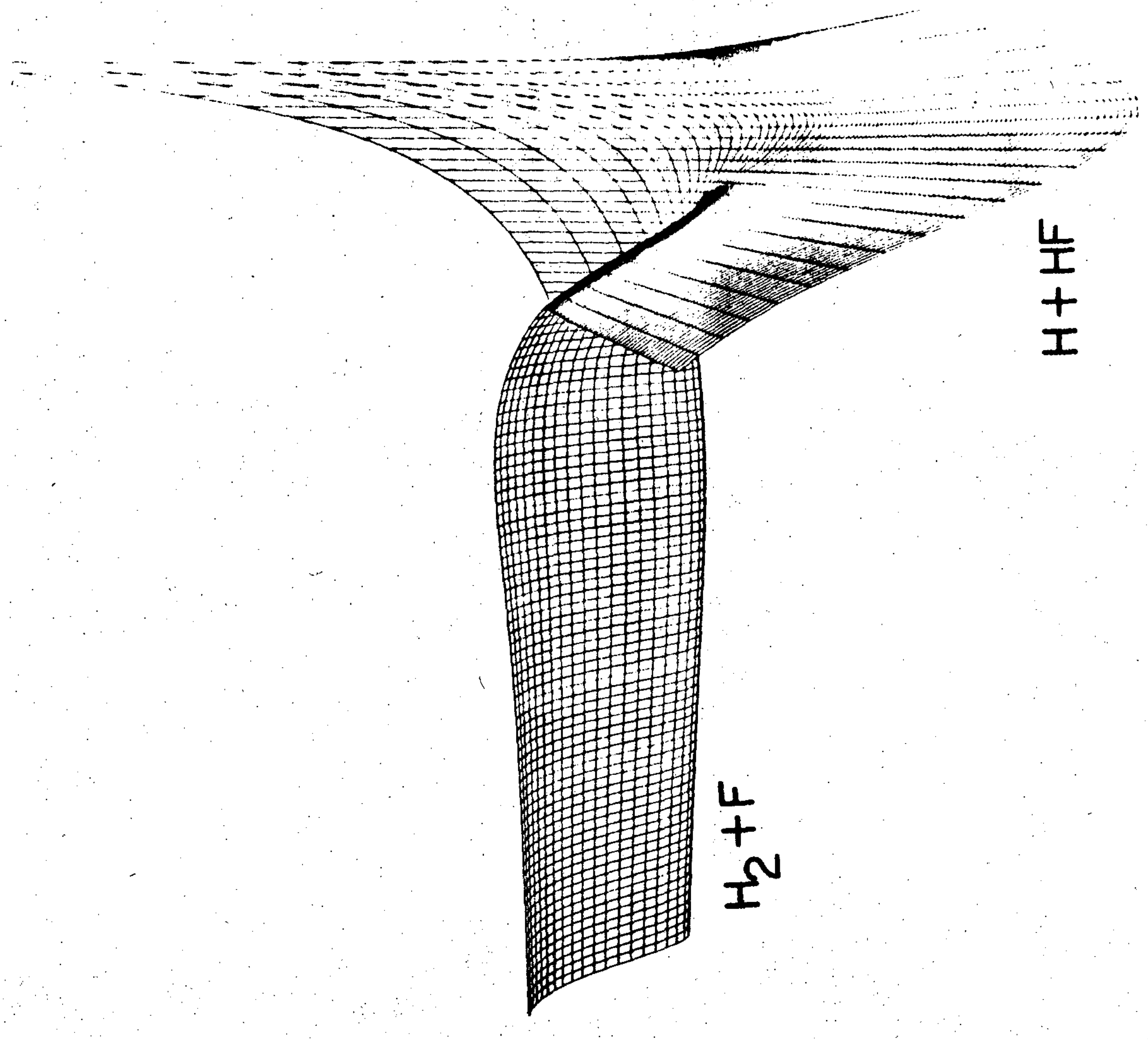


Fig. 3

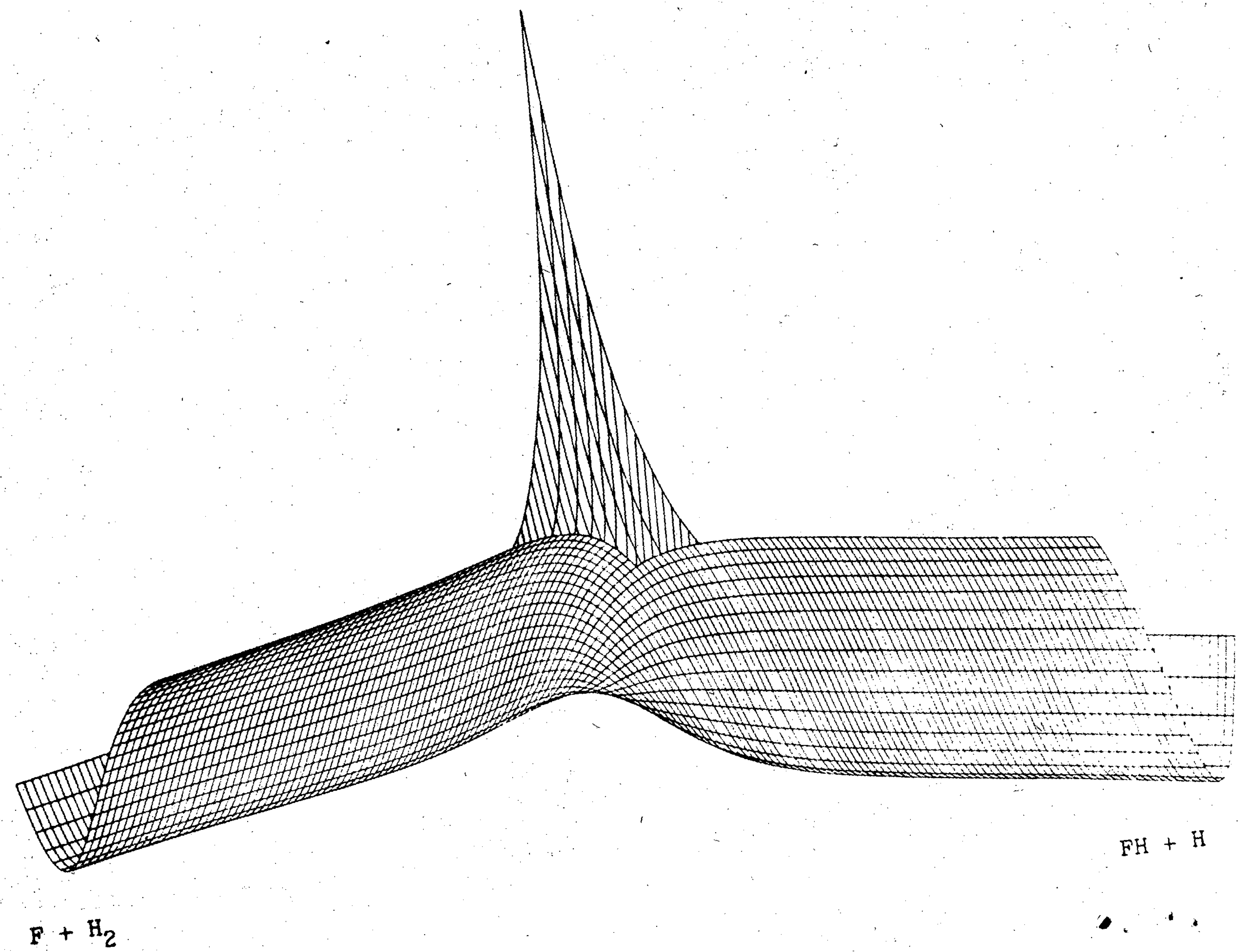


Fig. 4

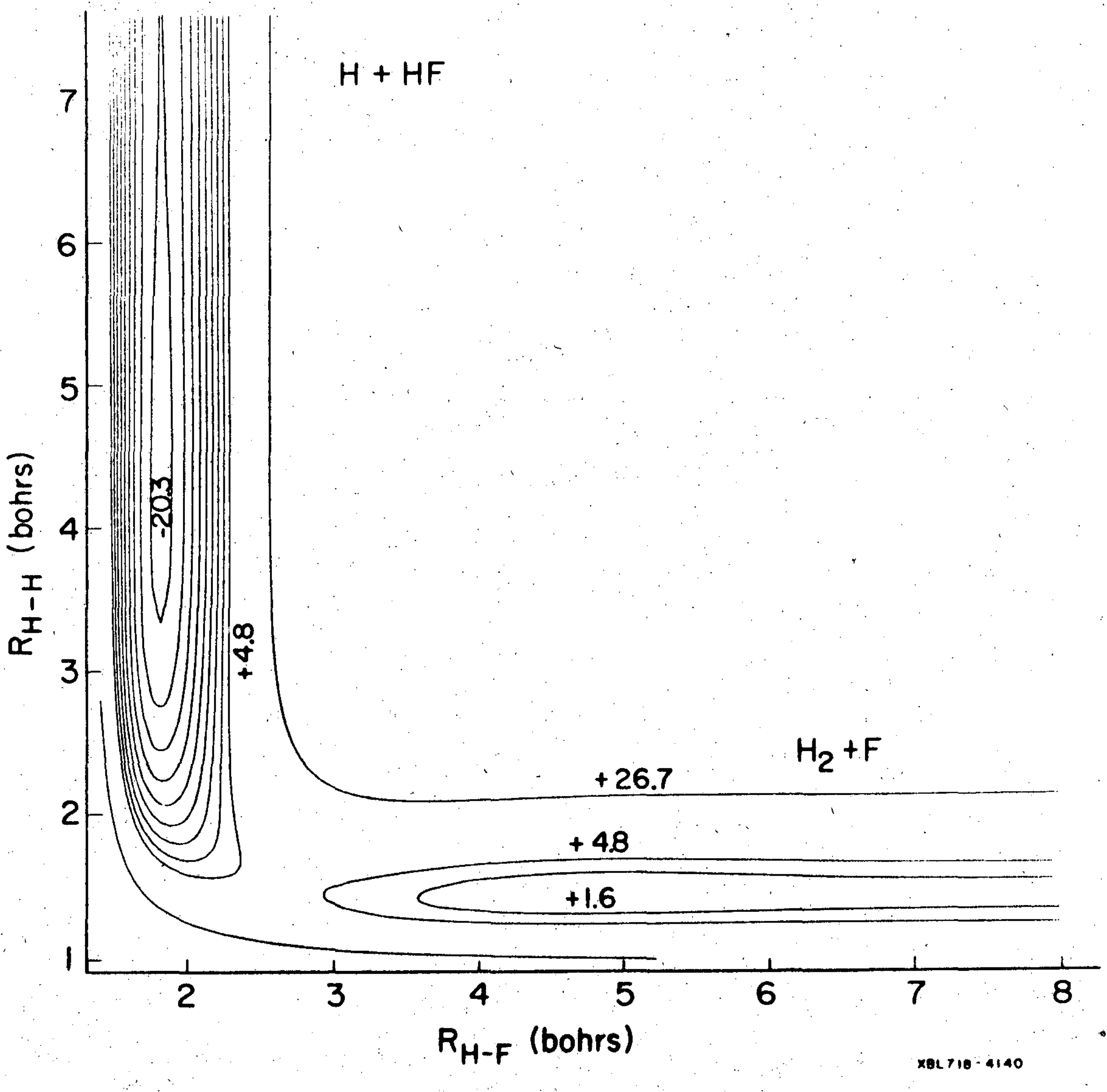

H + HF
H_2 + F
-20.3
+4.8
+26.7
+4.8
+1.6
R_{H-H} (bohrs)
R_{H-F} (bohrs)
1
2
3
4
5
6
7
2
3
4
5
6
7
8
XBL 718-4140

Fig. 5

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