# A Physics-based Approach to Hypersonic Nonequilibrium Chemistry

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# Overview

- This talk describes the recent efforts within NASA to develop detailed models for the physics and chemistry that takes place in the gaseous envelope that surrounds space vehicles when they enter the atmosphere of Earth, Mars and other planets.
  - The NASA Fundamental Aeronautics Program (2006) established hypersonics research as one of its priorities
  - One thread has been the development of a **physics and chemistry based model** to describe hypersonic flows
  - This research program is now supported by NASA's Space Technology Mission Directorate

- In this talk I will discuss some of the methods we are using to meet this objective

# Outline

- A short introduction to entry physics and observation-based models for nonequilibrium chemistry

- Theoretical and computational methods used for constructing physics-based models

- Example #1 (Earth entry): $N_2$ dissociation

- Example #2 (Mars entry): CO dissociation and exchange reactions

- Plans for a complete physics-based model

- Summary

- Atmospheric entry for Earth occurs at ~100 km altitude
  - 4-6 km/s for suborbital trajectories
  - 8 km/s for trajectories from LEO and GEO
  - 10-11 km/s for lunar return
  - 12+ km/s for return from beyond the Earth-Moon system
  - ≥ 20 km/s for meteors

*The flowfield around a spacecraft is characterized by complex physical and chemical phenomena*
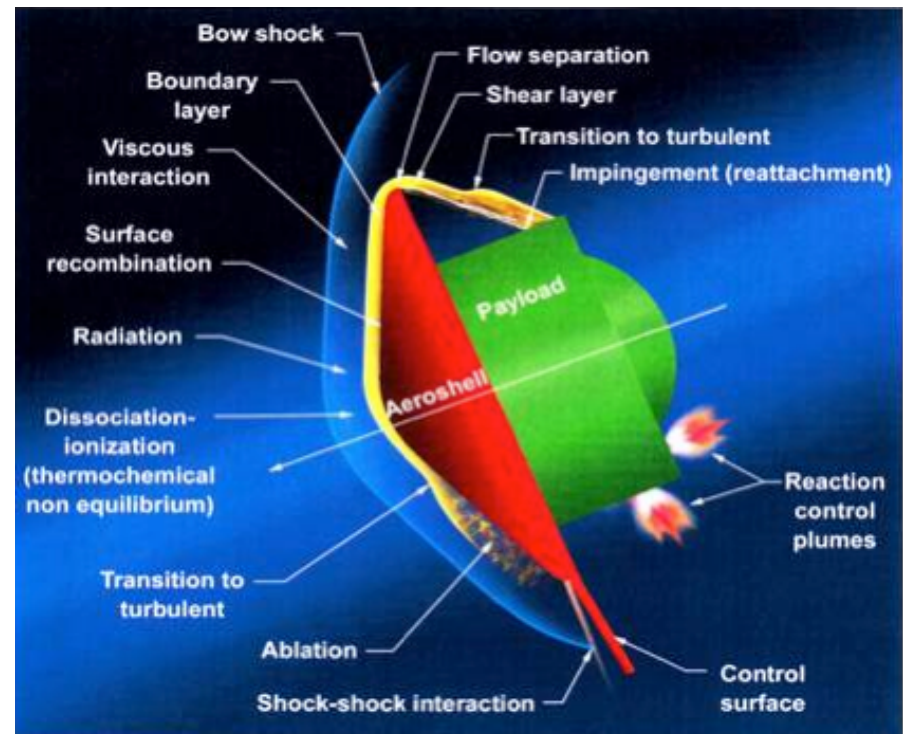
- Mars Science Laboratory (2012), payload weight 900 kg
  - Guided entry: 4.5 m diameter, 2400 kg aeroshell, velocity 5.8 km/s at entry (80 km) slowed to 0.5 km/s at 10 km altitude when parachute deployed (4 min after entry)
  - Landed 2.4 km from center of target ellipse (7x20km)
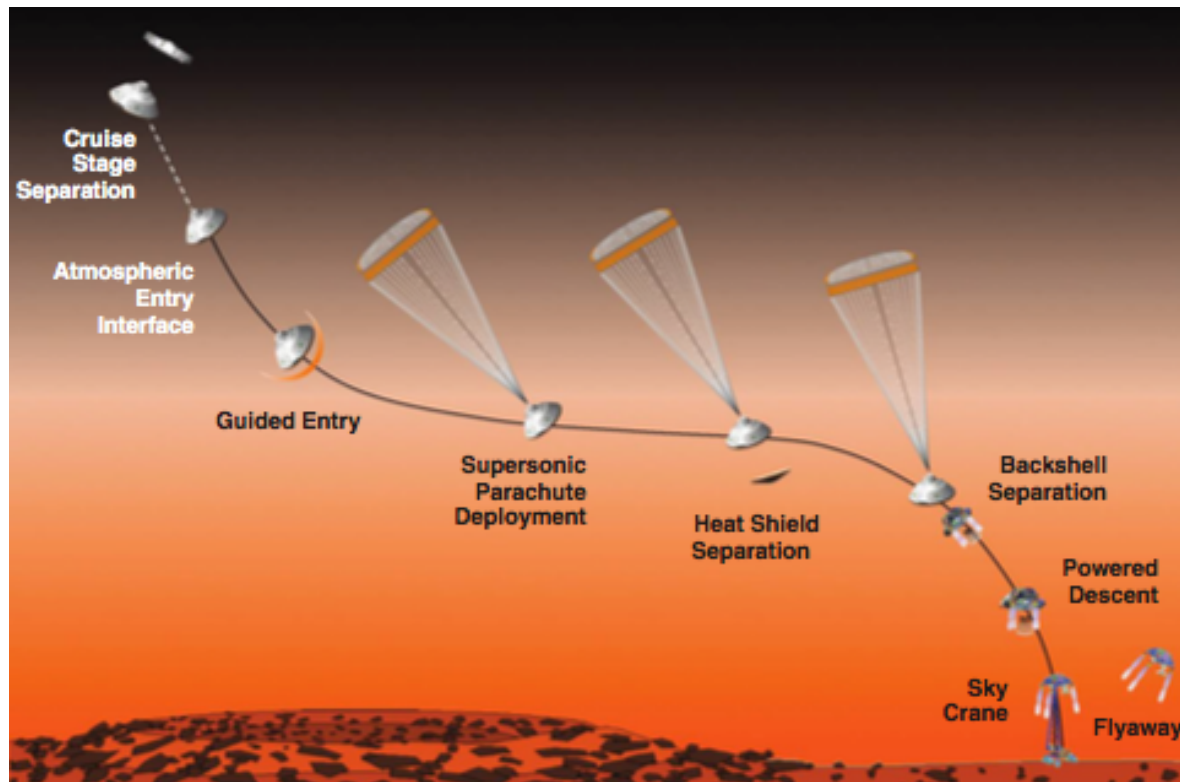
- Earth free stream condition: $N_2$ and $O_2$ at low $(T_0, P_0)$

- After the shock, flow relaxes toward new equilibrium condition $(T_{eq}, P_{eq})$ with $\tau_T < \tau_R < \tau_V$
  - Inelastic collisions result in transfer of translational energy to rotation, vibration and electronic modes
  - Dissociation of molecules occurs only from high-lying rotational and vibrational levels ($O_2$ dissociates first due to its smaller bond energy)
  - Once atomic species are present, exchange reactions (e.g., $O + N_2 \rightarrow N + NO$) also help convert $N_2$ to NO which also has a lower bond energy
  - Ions and electrons form initially by associative detachment (e.g., $N + O \rightarrow NO^+ + e^-$)
  - Inelastic electron-heavy particle collisions lead to further vibrational and electronic excitation and ionization of atoms and molecules

- If specific internal energy modes are described by separate temperatures $(T_R, T_V, T_E)$, these temperatures start at $T_0$ at the shock and increase until they are equal T

- As the flow is shock-heated, some atoms and molecules are highly excited and undergo intense radiative emission that is often observed just behind the shock

*This is called nonequilibrium radiation*

# Nonequilibrium Heating

## Example: 2-T model of Stardust sample return trajectory
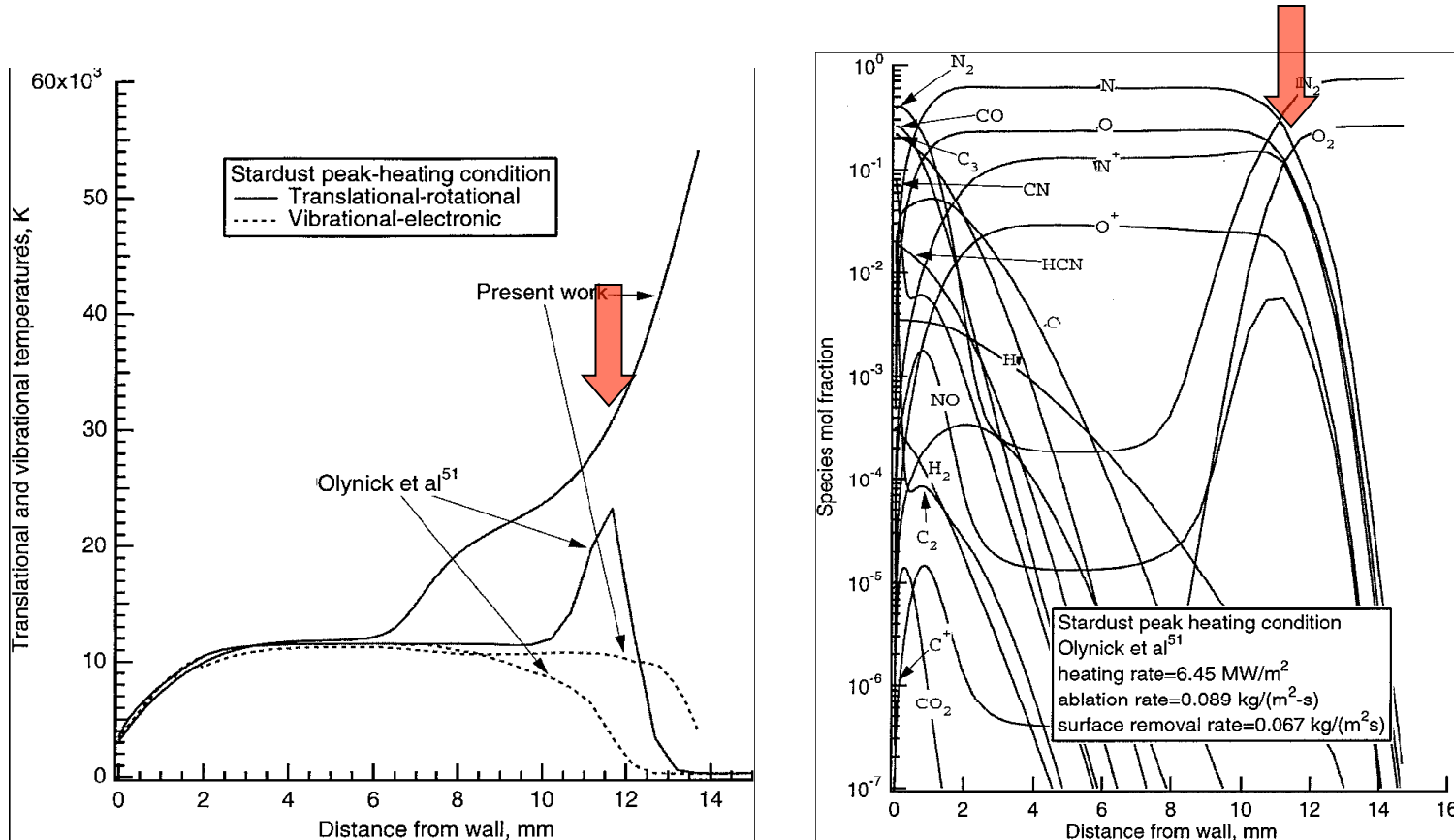


Red arrow locates nonequilibrium region
(from Park, Jaffe & Partridge, JTHT **15**, 76 (2001))

# Breakdown of LTE Models

- Shock tube experiments carried out for the Apollo program demonstrated that nonequilibrium radiation is produced during the induction period before dissociation

- Early CFD simulations of re-entry flowfields that assumed local thermodynamic equilibrium (LTE) did not adequately observed features of the flow
  - Incorrect shock stand-off distance
  - Pitching moment too small
  - Poor predictions of the radiative heat flux, especially in low-density regime

- It was evident that a one-temperature model could not accurately describe the chemical non-equilibrium in the shock layer

- Early efforts to describe these phenomena using multi-temperature models were not successful

- Chul Park created a successful two-temperature model to describe the important effects of nonequilibrium chemistry in the 1980's. The key development was the use of the geometric mean of T and $T_v$ to control the dissociation rate.

- The second temperature $T_v$ is determined from the Landau-Teller relaxation model

$$\frac{\partial E_V(T_V)}{\partial t} = \frac{(E_V(T_V) - E_V(T))}{\tau_V}$$

  - Geometric mean temperature used for dissociation is $T_{av} = [T \bullet T_V]^{1/2}$
  - Arrhenius rate coefficient expression ($k_{rxn} = AT^{-c} \exp(-T_{rxn}/T_{av})$ is used for collisional dissociation reactions
  - Rotation level populations are given by Boltzmann distribution at T ($T_R = T$)
  - Electronic state populations are given by Boltzmann distribution at $T_v$ ($T_{el} = T_v$)

- This empirical formulation enabled CFD simulations to reproduce certain nonequilibrium features observed in shock tube experiments and is <u>still widely used throughout the hypersonics community</u>

Ref. Park JTHT <u>2</u>, 8 (1988), JTHT <u>3</u>, 233 (1989), JTHT <u>7</u>, 385 (1993) & JTHT <u>8</u>, 9 (1994)

- Park's two-temperature (or T-T$_v$) model has been widely applied to forebody flows (compressing) and works _fairly_ well
  - Necessary parameters are easy to determine from experiment
  - The Arrhenius parameters are generally adjusted to match experimental data from shock tubes and from ground-based hypersonic test facilities (EAST,  LENS-XX, etc.)
  - Selected parameters are then adjusted so CFD-radiative transport simulations match benchmark radiative emission profiles or line-by-line emission spectra

- It can be readily incorporated into CFD codes without significant increase in the computational cost

# Shortcomings of the T-$T_v$ model

- For highly excited ro-vibrational levels, separation of the internal energy into vibrational and rotational components is problematic
  - Effective dissociation energy of a diatomic molecule varies with rotational level
  - For high degrees of rotational excitation, quasi-bound vibrational levels exist and can make important contributions to the reaction rates

- The tuning of reaction rate coefficients does not yield a unique set of parameters
  - Using benchmark data for different shock speeds results in different parameter sets (and the accuracy of a parameter set deteriorates as one strays farther from its benchmark condition)
  - Parameter set is also sensitive to the values of energy relaxation times used for the tuning
  - Comparison between tuned and experimental or computed rate coefficients is difficult

- Difficult to define $T_v$ (and $T_{el}$) when dissociation is nearly complete

- Sometimes different formulations of $T_v$ are needed for each diatomic species

# Biggest shortcoming of the T-T$_v$ model

- Parameter sets tuned for compressing flows generally work poorly for expanding flows
  - Expanding flows (e.g., afterbody) generally have $T_v > T$ and molecules formed by recombination
  - Physical models of the relaxation processes are quite different
  - T-T$_v$ model results (without retuning) seem to give anomalously high radiative heat flux

- Problematic to use this model for "whole vehicle" CFD

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# New Paradigm

- Accurate quantum mechanics calculations to quantify interaction energy between atoms and molecules

  - Potential Energy Surface (PES) obtained from solution of Schrödinger equation

  - Repeated for many geometric arrangements of the atoms (for *n* atoms there are *3n-6* degrees of freedom)

  - Goal is ≤ ± 5 kJ/mol relative accuracy (~0.05 eV, ~400 $cm^{-1}$)

- Classical mechanics simulations of heavy particle collisions to compute collision cross sections and rate coefficients

  - Compute trajectories of many individual collisions (Quasi-Classical Trajectory or QCT calculation)

  - Monte Carlo sampling used reproduce random or thermal collisional distributions

  *(Ref: Jaffe et al. "First principles calculation of heavy particle rate coefficients" in "Hypersonic onequilibrium Flows: Fundamentals and Recent Advances", E. Josyula, ed, AIAA 2015)*

# Quantum Mechanics Refresher

- Time independent electronic Schrödinger equation (clamped nuclei)

$$\left[ -\frac{\hbar^2}{2m} \nabla^2 + V(\mathbf{r}) \right] u(\mathbf{r}) = E u(\mathbf{r})$$

- $V = +1/r_{i,j}$ (for electron-electron interactions)
  - Coulomb interaction <u>for potential energy</u> calculations (indices i,j run over all the electrons)
  - Non-local term making exact solution difficult
- Wavefunctions (u) and energy (E) determined by double basis set expansion method

# Methods – PES for $N_2$ + N & $N_2$ + $N_2$

- With all the advances in computer hardware and quantum chemistry algorithms, achieving chemical accuracy (~4 kJ/mol) is still difficult for 4-atom cases (e.g., $N_2+N_2$)
  - Hybrid approach used for the $N_4$ PES
    1. Large atomic-orbital basis set expansion
    2. Multi-Reference Configuration Interaction (MRCI) for geometries where the N-N bonds are greatly stretched
    3. Single-reference method (CCSD(T)) for other geometries
    4. Accurate empirical diatomic potential function for $N_2$

- Energy computed for ~4000 geometries and fit to an analytic function for use in QCT calculations

Analytic representation of ab initio quantum chemical energies over an unstructured grid of $N_3$ geometries
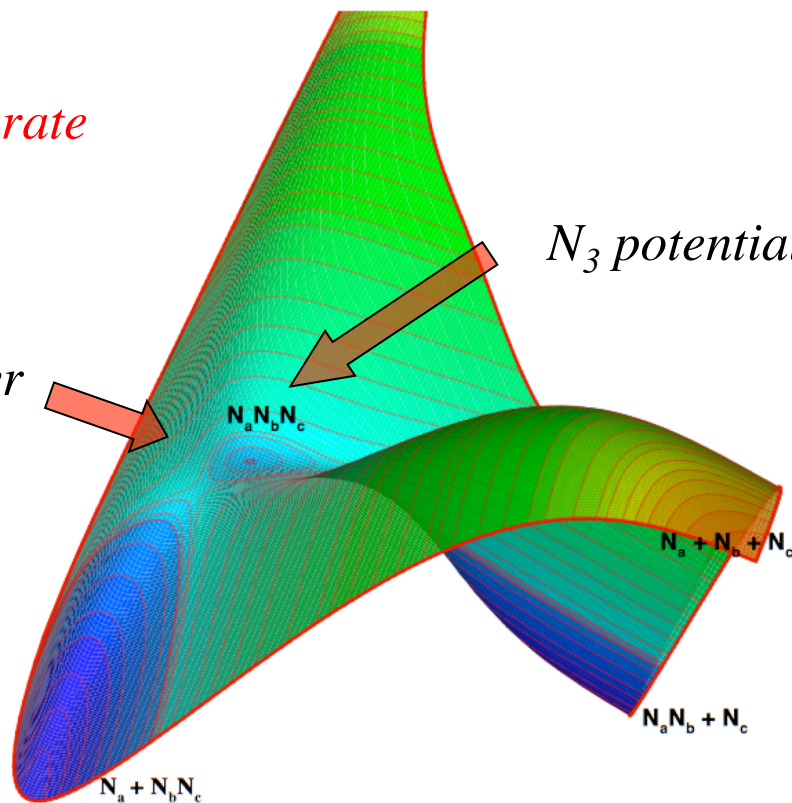
*Relative energies accurate to ±5 kJ/mol*

*$N_3$ potential well*

*Exchange reaction barrier*

$N_a N_b N_c$

$N_a + N_b + N_c$

$N_a + N_b N_c$

$N_a N_b + N_c$

color key
**blue - low energy**
**red - high energy**

- Some research groups choose to compute ab initio energies at 20,000-40,000 geometries and use multidimensional splines as a fitting function, **we choose to compute the energies at far fewer geometries (~5000) and use more complicated fitting functions**

- Many body decomposition of the potential energy for the $N_2$ + N PES:

$$V = V^{NN}(R_{ab}) + V^{NN}(R_{bc}) + V^{NN}(R_{ac}) + V^{3-body}(R_{ab}, R_{bc}, R_{ac})$$

  - $V^{NN}$ (2-body term) is sum of repulsive, short-range and long-range terms:

$$49\exp(-\alpha_r R)/R \quad + \quad R^6\exp(-\alpha_s R)\sum_j C_j(R - R_e)^j \quad + \quad -\frac{C_6}{R^6 + \delta^6} - \frac{C_8}{(R^4 + \delta^4)^2} - \frac{C_{10}}{(R^2 + \delta^2)^5}$$

  - $V^{3-body}$ term is the sum of short-range and long-range terms:

$$V^{3-body} = V^{LR}(R_c, r_c, \theta_c) + V^{LR}(R_a, r_a, \theta_a) + V^{LR}(R_b, r_b, \theta_b) + V^{SR}(R_{ab}, R_{ac}, R_{bc})$$

$$V^{SR} = \sum_\alpha \exp(-b_1 r_\alpha^2 - b_2 R_\alpha^2)\sum_{ijk} C_{ijk} r_\alpha^i R_\alpha^j P_k(\cos\theta_\alpha) \qquad V^{LR} = -C_6 P_2(\cos\theta)\exp\left[-a^{LR}(R - R^{LR})\right]/(r^6 + r_d^6)$$
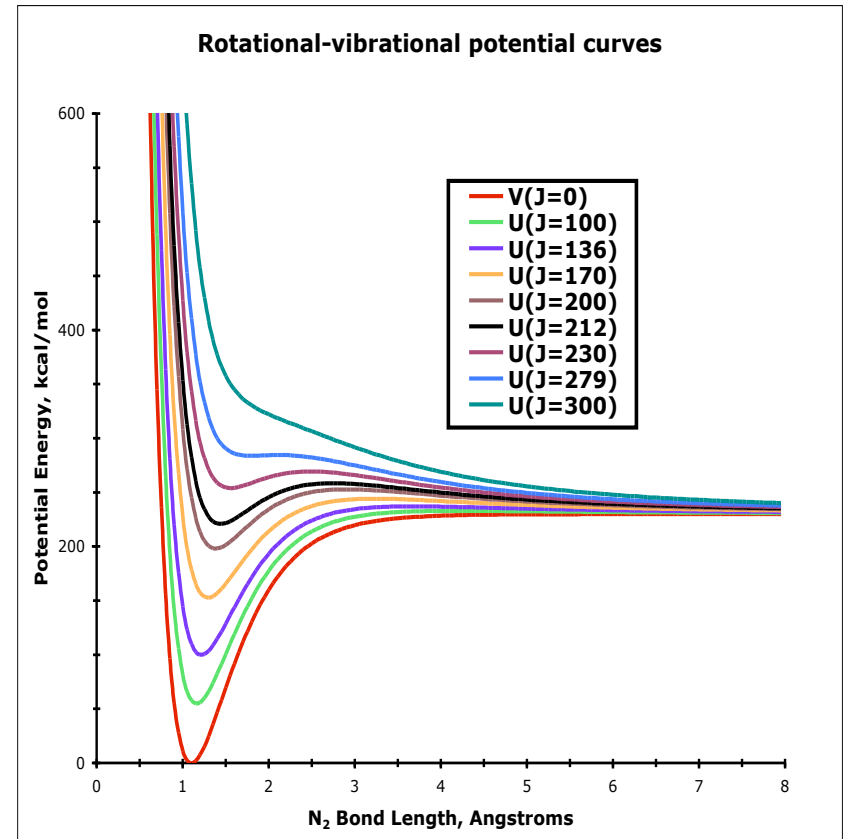
  - Non-linear least squares fit carried out using the following weight function:

$$w = 1 + a_w \sum_\alpha \exp\left[-a_R\left(R_\alpha - R_{ts}\right)^2 - a_\theta(\cos\chi_\alpha - \cos\chi_{ts})^2 - a\left(\tilde{R}_\alpha - \tilde{R}_{ts}\right)^2\right]$$

• ***Rotation and vibration are not separable!*** For high rotation quantum numbers (J), dissociation energy is reduced, rotational and vibrational spacings become comparable
• Many long-lived vibration levels above dissociation limit (quasibound)
• New $N_2$ potential (Leroy, J Chem Phys 2006) from experimental measurements; $v_{max}$ = 60 and $J_{max}$ = 279
• 9390 ro-vibrational levels for $N_2$ ($X^1\Sigma_g^+$)

**Rotational-vibrational potential curves**



Legend:
- V(J=0)
- U(J=100)
- U(J=136)
- U(J=170)
- U(J=200)
- U(J=212)
- U(J=230)
- U(J=279)
- U(J=300)

Y-axis: Potential Energy, kcal/mol
X-axis: $N_2$ Bond Length, Angstroms

$$V_J(r) = V_{J=0}(r) + J(J+1)B_{rot}(r_e/r)^2$$

- Hamilton's equations of motion ($q_i$, $p_i$ are coordinates and momenta; $H$ is total energy):

$$\frac{dq_i}{dt} = \frac{\partial H}{\partial p_i} \qquad \frac{dp_i}{dt} = -\frac{\partial H}{\partial q_i} = -\frac{\partial V_{PES}}{\partial q_i}$$

- Simulate collisions like $N + N_2$ or $N_2 + N_2$
  - Initial conditions correspond to $N_2(v,J)$ with random orientation, rotation and vibration phase, impact parameter ($b$) and relative collision velocity ($V_R$)
  - State-to-state cross section obtained from a large sample of trajectories
  $$S_R\left(v_i, J_i, v_f, J_f, V_R\right) = 2\pi \int_0^{bmax} P_R\left(V_R, b\right) b\,db$$

  - $P_R = N_f/N_{tot}$ ($N_f$ is the number of trajectories with final state $f$)

- State-to-state rate coefficient obtained by integration of Boltzmann-weighted cross section over collision energy $(E_R)$:

$$k_{if}(T) = \frac{1}{3}(2k_BT)^{\frac{3}{2}}(\mu\pi)^{-\frac{1}{2}} \int_0^\infty \sigma_{if}(E_i)\, E_i \exp\left(-\frac{E_i}{k_BT}\right) dE_i$$

- The thermal rate coefficient $k_{rxn}(T,T')$ is obtained as the Boltzmann weighted sum of the state-to-state values (with $T' = T$ or $T_{int}$ )

- QCT uses Monte Carlo sampling procedures with relative statistical sampling error $(1-\sigma)$ equal to $N_f^{-\frac{1}{2}}$

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# Example #1 N$_2$ Dissociation

- Energy Transfer:
  - $N_2(v_i,J_i) + N_2(v_i',J_i') \rightarrow N_2(v_f,J_f) + N_2(v_f',J_f')$     ~9390$^4$/4  reactions
  - $N_2(v_i,J_i) + N \rightarrow N_2(v_f,J_f) + N$                          ~9390$^2$     reactions

- Dissociation:
  - $N_2(v_i,J_i) + N_2(v_i',J_i') \rightarrow N + N + N_2(v_f',J_f')$     ~9390$^3$/2  reactions
  - $N_2(v_i,J_i) + N_2(v_i',J_i') \rightarrow N + N + N + N$            ~9390$^2$/2  reactions
  - $N_2(v_i,J_i) + N \rightarrow N + N + N$                           9390     reactions

- For N$_2$ + N we computed all state-to-state rate coefficients (AIAA-2008-1208,1209 & AIAA-2009-1569) but for N$_2$ + N$_2$ that is impractical

# Coarse Grained Models

- Use of complete sets of state-to-state rate coefficients is problematic for reactions involving $N_2$
  - We have been developing coarse-grain models as an alternative
  - These will be discussed by Professor Panesi in the next talk

- An alternative to coarse-grain models is a $T$-$T_{int}$ model which uses a separate temperature to rotation-vibration energy
  - However, this model does not capture the details of nonequilibrium chemistry, because the high-energy rovibrational level populations deviate from a Boltzmann distribution
  - Dissociation is initiated from these energy levels

- Dissociation of the atmospheric molecules is a key chemical process taking place in hypersonic flows
  - $O_2$ (with dissociation energy of 5eV) and $N_2$ (9.6 eV) in air

- Heterogeneous exchange reactions can provide lower energy (i.e. faster) routes for $N_2$ removal
  - $N_2 + O \rightarrow NO + O$
  - $O_2 + N \rightarrow NO + N$

- Homogeneous exchange and inelastic processes control ro-vibrational relaxation times

*For Earth entry at speeds of 10 km/s or greater, the rate of $N_2$ dissociation is one of the important processes for determining the non-equilibrium temperature overshoot, the magnitude of N-atom VUV radiation and the establishment of thermochemical equilibrium in the shock layer*

- Experimental data
  - Appleton (1968)      $N_2$ + Ar, $N_2$, N     6000 – 15,000 K
  - Hanson (1972)       $N_2$ + $N_2$, N        6000 – 12,000 K
  - Hornung (1972)      $N_2$ + $N_2$, N        5000 – 15,000 K
  - Roth (1985)          $N_2$ + $N_2$           3400 – 6500 K
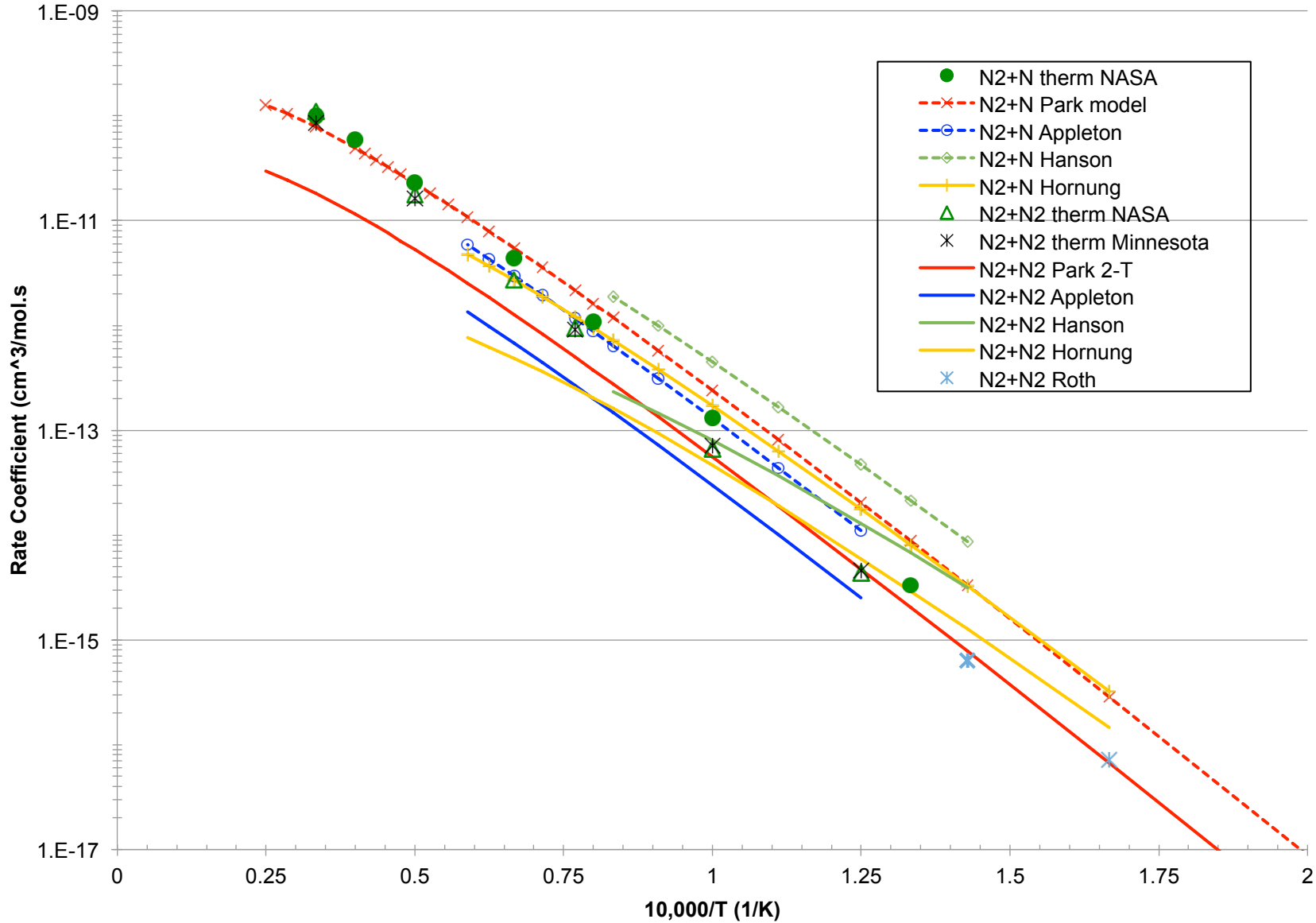
- Park (1993) T-$T_v$ model

- Computation
  - NASA Ames (2008-2013)    QCT
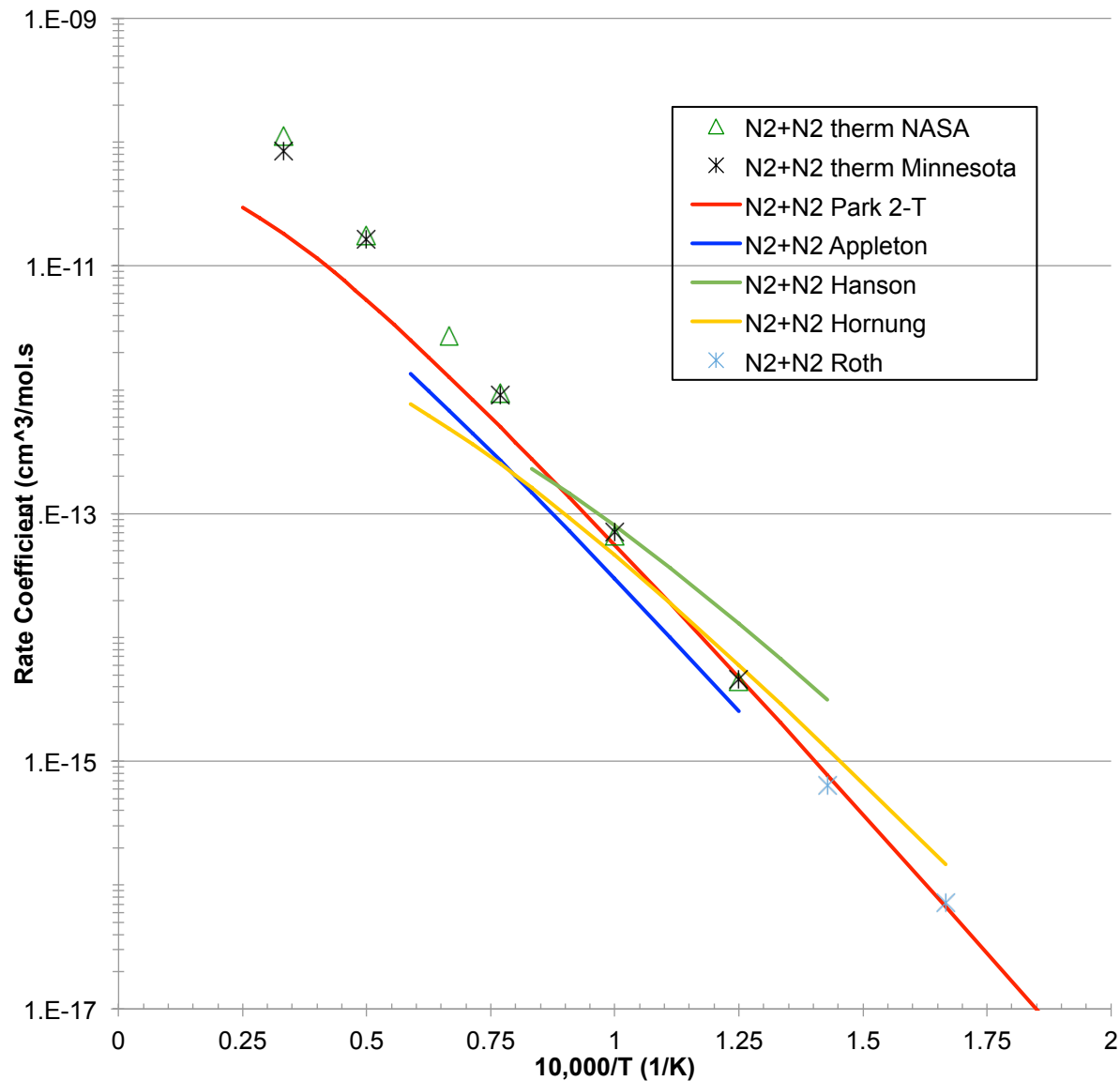  - U. Minn (2014)              QCT

Ref: Jaffe et al. AIAA 2016-0503
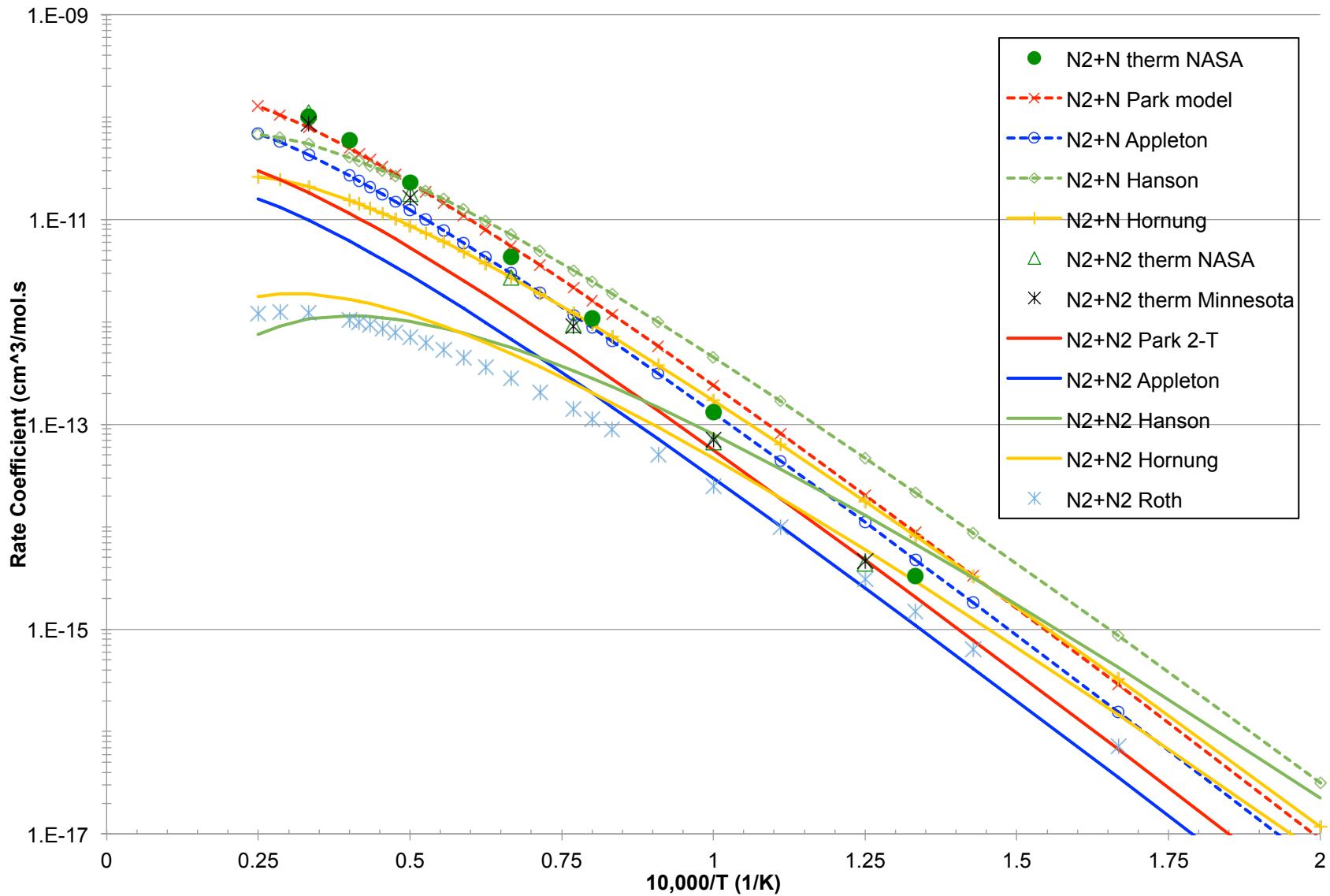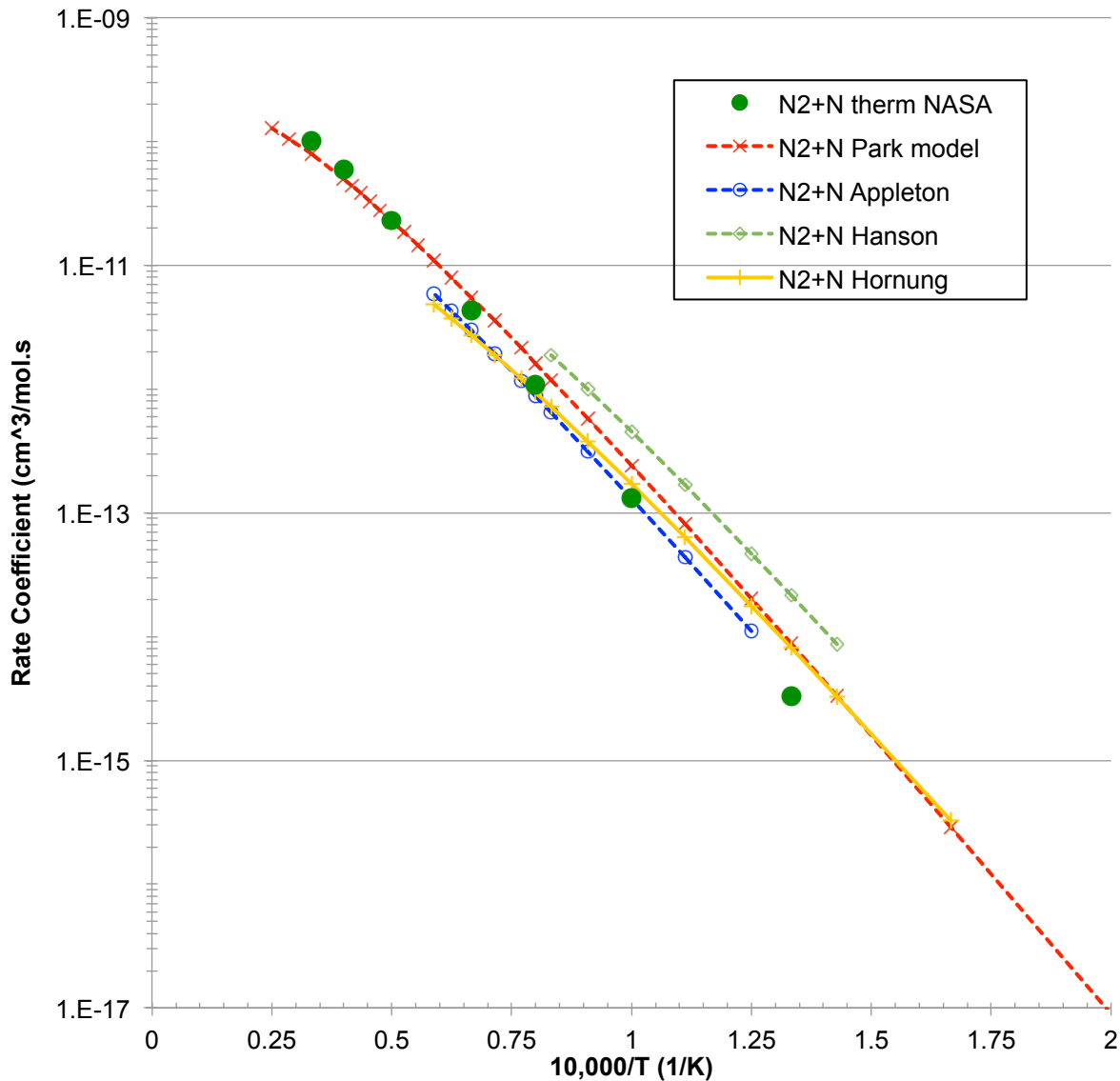
# N₂ Dissociation Rate Coefficients

# N$_2$ Dissociation Rate Coefficients

- Excellent agreement between NASA and U. Minn. QCT results
- Good agreement between Park model and QCT
- Fair agreement between QCT and expt.

- Good agreement between QCT and Appleton and Hornung expt data
- Fair agreement between Park model and QCT

# Conclusions for $N_2$ Dissociation

- Excellent agreement between QCT rate coefficients from two different research groups
  - PES calculations were planned and executed independently

- Inconsistencies in the experimental data make it difficult to use for validation of the QCT results
  - Individual experiments do not span the relevant temperature range
  - Use of multiplicative scale factors for different collision partners only justified because of data limitations

- Extrapolation of experimental Arrhenius expressions beyond the temperature range of the data is risky!
  - Large temperature exponents cause spurious curvature in the high temperature data

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- Mars atmosphere is ~96% $CO_2$, ~2% $N_2$ ~2% Ar

- Dissociation
  - $CO_2$ (5eV) very fast and nearly complete
  - CO (11 eV) rate determining process

- Heterogeneous exchange reactions can provide lower energy (i.e. faster) routes for CO removal
  - $CO + O \rightarrow C + O_2$
  - $CO + N \rightarrow O + CN$
  - $CO + C \rightarrow O + C_2$

*For Mars entry at 6-8 km/s, the rate of CO dissociation is critical because CO 4th positive radiation is a dominant source of heating*

# Example #2 CO Dissociation

- The radiative heat flux experienced by spacecraft entering the Martian atmosphere at 5-8 km/s is mostly due to the CO 4th positive band system ($\lambda < 200$ nm)
  - Radiative heating is therefore proportional to the mole fraction of CO in the bow shock layer
  - CFD-Radiative transport calculations using the T-$T_v$ Mars chemistry model (Park et al. JTHT, 1994) predict greater radiance than observed in tests using the Electric Arc Shock Tube at Ames
  - Johnston and Brandis (JQSRT, 2014) scaled some of the rate coefficient parameters to force agreement between CFD and EAST

- Ab initio PESs have been computed for CO + Ar and CO + O and used in QCT calculations of CO dissociation rate coefficients (Schwenke et al., J Chem. Phys. submitted )
  - Experimental data for CO + Ar seem more reliable than for CO + O

After nearly all of the $CO_2$ is dissociated, CO and O are the major species (with equal mole fraction)
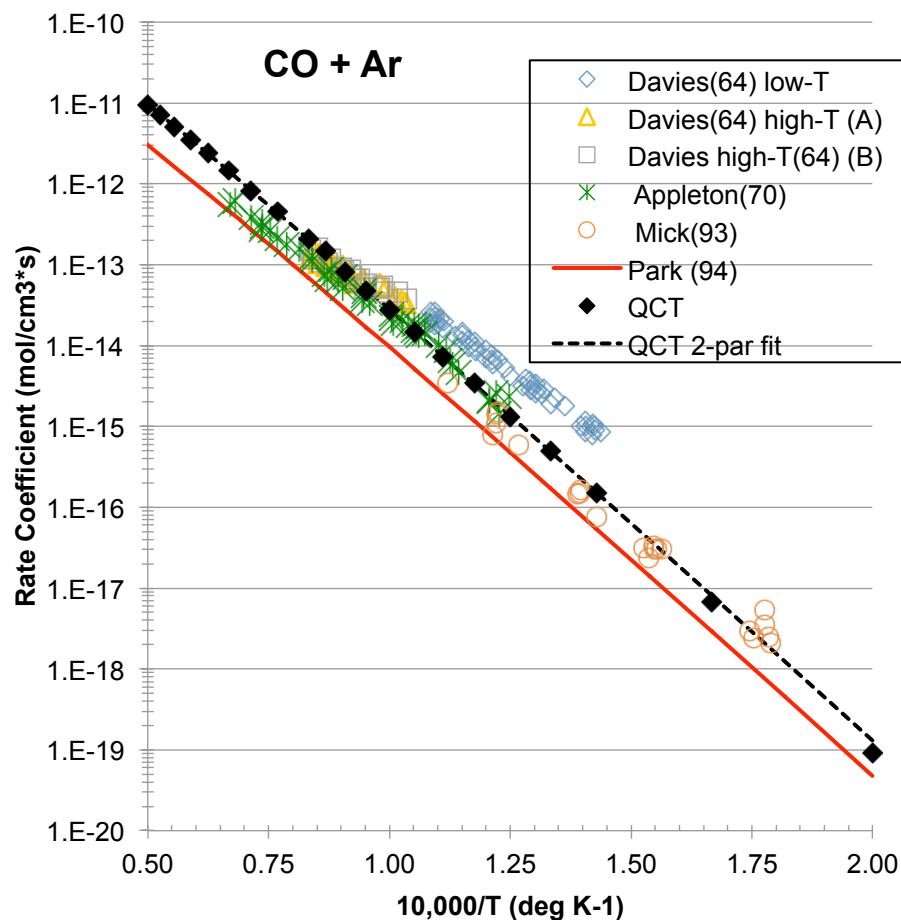
Experimental data

| | |
|---|---|
| Davies (1964) | CO + Ar, CO,O |
| Appleton (1970) | CO + Ar, CO, O |
| Hanson (1974) | CO + CO, O |
| Mick & Roth (1993) | CO + Ar, CO |

Park (1994) T-T$_v$ model

NASA Ames (2014-2015)   QCT

- Very good agreement between QCT and Appleton and Mick and Roth expt.
- Fair to good agreement between QCT and Davies and Park (94)

# Rate Coefficient Ratios for CO Dissociation

- Early experimental values (pre-1972)
  - $k_{CO+CO}/k_{CO+Ar} = 1\text{-}2$   $k_{CO+O}/k_{CO+Ar} = 15$
- Park (1994) T-$T_V$ model
  - $k_{CO+CO}/k_{CO+Ar} = 10$    $k_{CO+O}/k_{CO+Ar} = 15$
- Johnston and Brandis (2014)
  - $k_{CO+CO}/k_{CO+Ar} = 10$    $k_{CO+O}/k_{CO+Ar} = 15$
  - $k_{CO+Ar(JB)}/k_{CO+Ar(Park)} = 5$
- Our objective is to determine these rate coefficient ratios using QCT calculations and determine how they vary with temperature
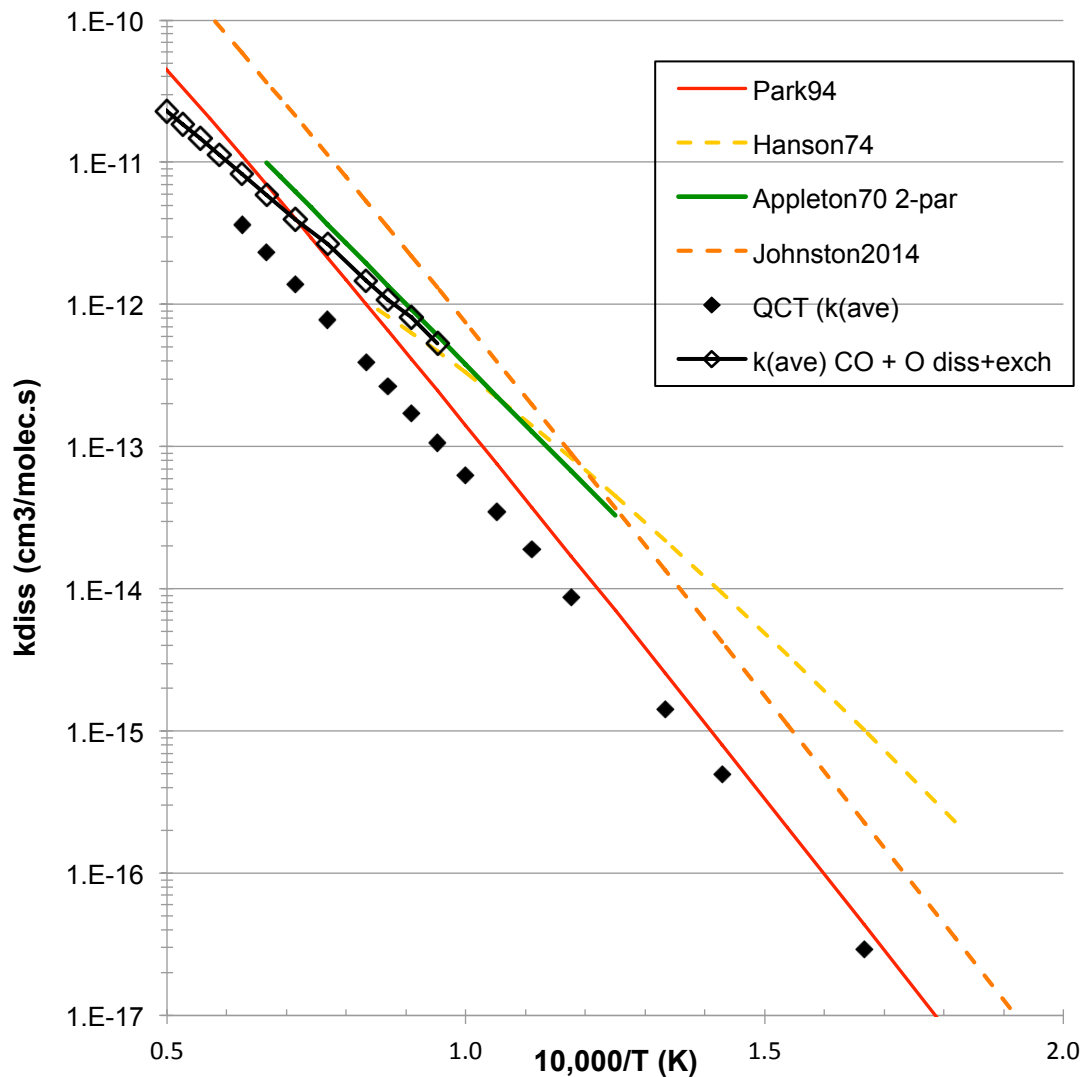
- Three triplet PESs with equal statistical weight needed for CO + O ($^3$P) collisions
  - Lead to different $CO_2$ triplet electronic states (1 $^3$A', 1 $^3$A" and 2 $^3$A")
  - These $CO_2$ states are bent (O-C-O angle ~120$^o$)
  - 1 $^3$A' has well depth ~45 kJ/mol (~0.6 eV); other states are more weakly bound

- Heterogeneous exchange reaction forming $O_2$ + C also possible on these PESs
  - Reaction is endothermic by 6.1 eV (low in comparison to the 11.2 eV dissociation energy of CO)

- All three PESs used for the QCT calculations of CO dissociation and exchange reactions

# CO + O Rate Coefficients
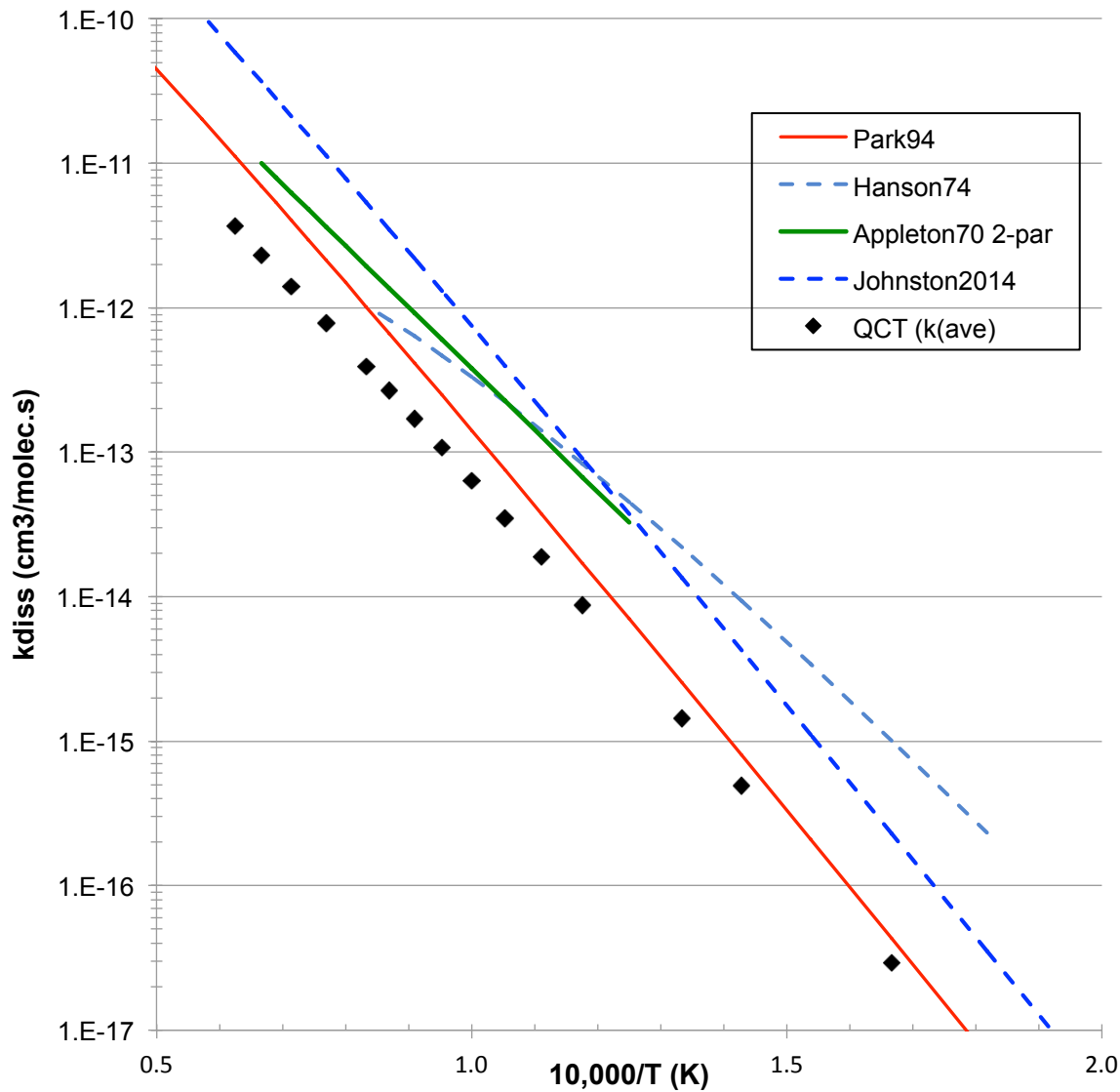


- Fair agreement between QCT and Park (94) model.
- Agreement between QCT and expt is poor
- $k_{CO+O}/k_{CO+Ar}$ ~3-5 for QCT, not 15

Legend:
- Park94
- Hanson74
- Appleton70 2-par
- Johnston2014
- QCT (k(ave))

Y-axis: kdiss (cm3/molec.s)
X-axis: 10,000/T (K)

# CO + O Rate Coefficients



Addition of hetero exchange rate coeff to dissociation greatly improves agreement with expt (especially at lower temperatures

- Heterogeneous exchange reaction provides a second pathway for CO removal, especially for T < 10,000 K

  - Converts CO to $O_2$, which is readily dissociated at these temperatures

  - Park (94) model has rate coefficient for this reaction that is 10x smaller than QCT value

  - Experimental CO dissociation rate coefficients implicitly include this reaction by having spuriously large dissociation rates and larger temperature exponents

- For CO + Ar, QCT and experimental dissociation rate coefficients show good agreement

- QCT rate coefficients needed for CO + CO to complete work on this set of reactions

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- CO dissociation by CO + CO collisions (PES and QCT)
  - Crrent models use a value that seems to be 5-10x too high

- For the other Earth and Mars reactions, high quality PESs exist and we will compute thermal rate coefficients for $O_2$ dissociation, Zeldovich exchange reactions (O + $N_2$ and N + $O_2$) and CO +N

- Coarse-grain methods will be used in conjunction with QCT calculations to provide non-equilibrium kinetics data
  - Will be used to compute relaxation parameters
  - Can provide an alternative to multi-temperature models

- This will yield a consistent set of reaction rate data for Earth and Mars entry

- The effort to create a new physics-based model for describing nonequilibrium phenomena in re-entry flowfields is bearing fruit

- Potential energy surfaces and thermal rate coefficients for dissociation and exchange reactions have been computed for collisions involving $N_2$ and CO
  - Independent calculations for $N_2$ dissociation from two research groups show remarkable agreement
  - Rate coefficients for different dissociation reactions uncovered inconsistencies in the older data from shock tube experiments

- Work is progressing toward creation of a data base of QCT rate coefficients for the complete set of heavy particle reactions for modeling Earth and Mars entry

- Support from the Entry Systems Modeling Program (NASA Space Technology Directorate)

- NASA Collaborators
  - David Schwenke
  - Galina Chaban
  - Winifred Huo
  - Lu Xu

- U. Illinois Collaborators
  - (Prof) Marco Panesi
  - Simone Venturi
  - Robyn Mcdonald

- U. Minnesota Collaborators
  - (Prof) Tom Schwartzentruber
  - Maninder Grover



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MARS

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NASA's Mars Exploration Program seeks to understand whether Mars was, is, or can be a habitable world. Missions like Mars Pathfinder, Mars Exploration Rovers, Mars Science Laboratory and Mars Reconnaissance Orbiter, among many others, have provided important information in understanding of the habitability of Mars. This poster imagines a future day when we've achieved our vision of human exploration of Mars and takes a nostalgic look back at the great imagined milestones of Mars exploration that will someday be celebrated as "historic sites."

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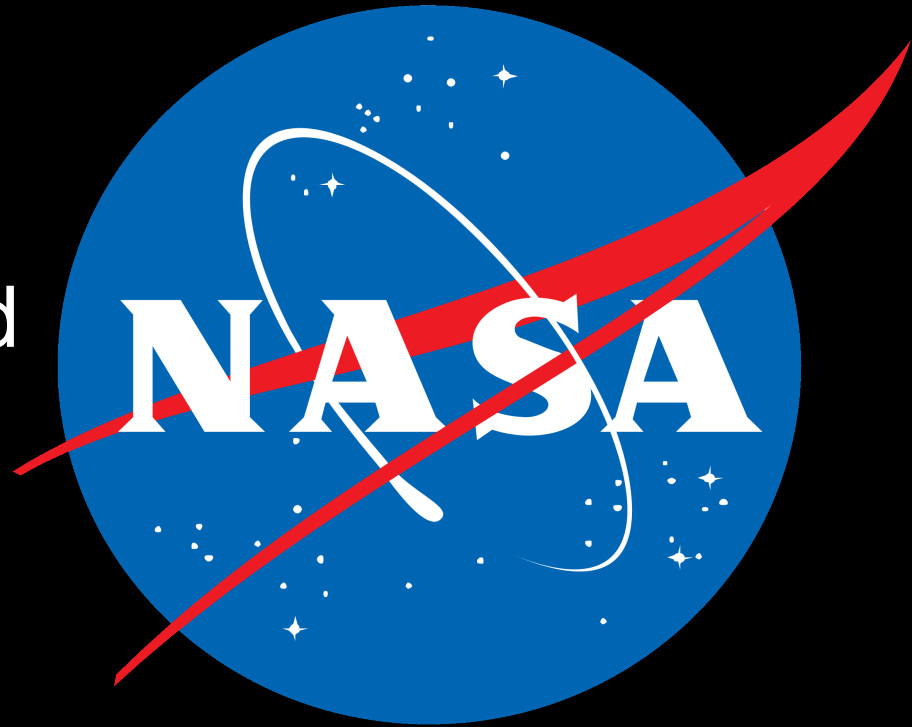
NASA Jet Propulsion Laboratory
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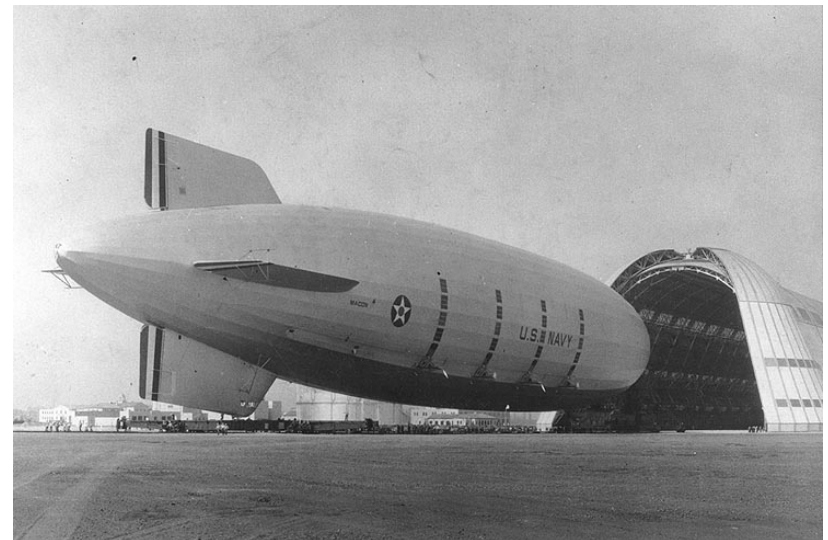- Back up material

1933 Moffett Field (US Navy Airfield) dirigible hangar completed

1938 NACA Ames Lab for aeronautics research

1951 Ames involved in missile R&D (pioneering work in CFD & thermophysics)

1958 NACA became part of NASA Today NASA Ames Research Center has more than 2300 research personnel and contributes to many NASA programs

- Theoretical/computational study of the formation of bow shocks for vehicles traveling at hypersonic speeds
- Shock stand off results in reduced heat flux

- Work at Ames in early 50's
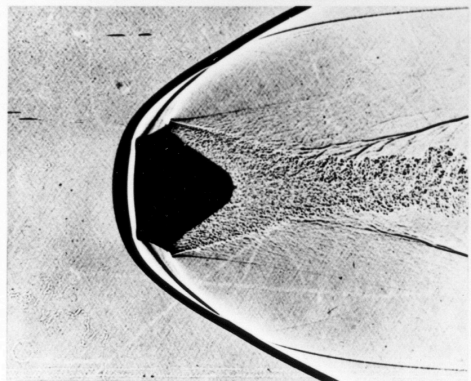- Confirmed by ballistic range tests (Schlieren photographs)

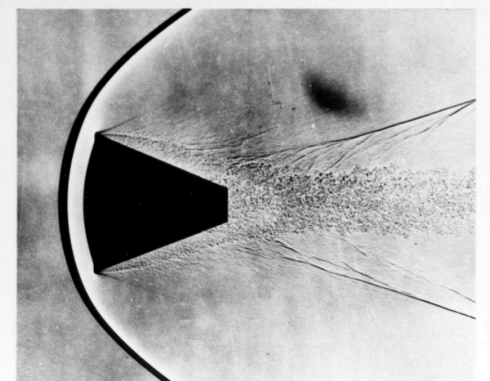Sphere-cone has become standard shape for atmospheric entry
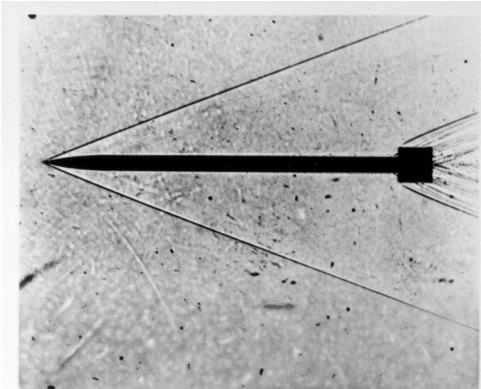


INITIAL CONCEPT
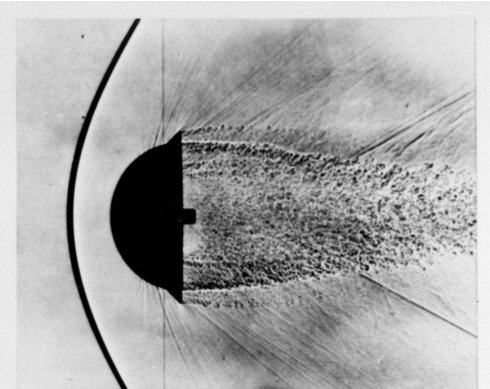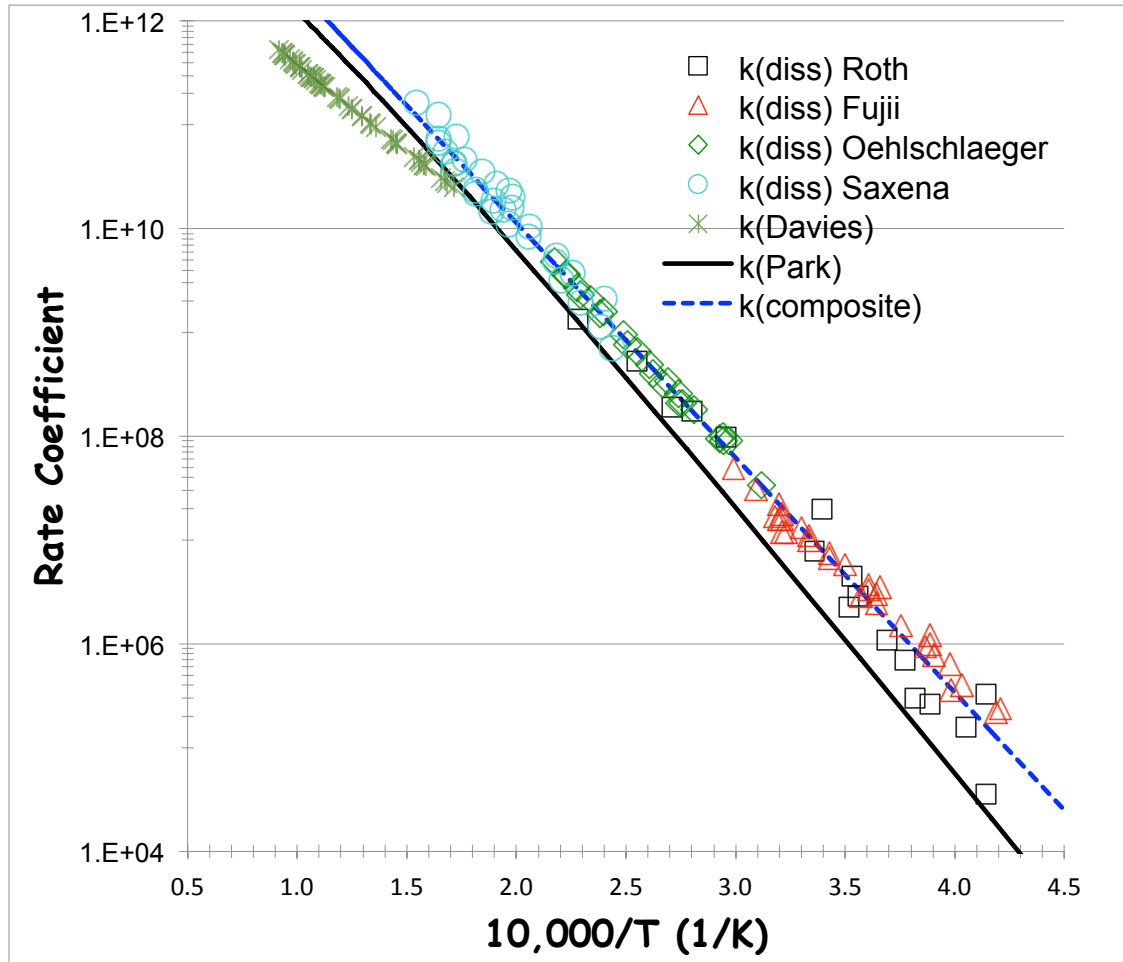
BLUNT BODY CONCEPT 1953

MISSILE NOSE CONES 1953-1957

MANNED CAPSULE CONCEPT 1957

# Meta-Analysis of New CO$_2$ Dissociation Rate Coefficient Data



Data for $CO_2$ + Ar/Kr

k(composite) = 3.83E-14 *exp(-52116/T)

$E_A$ = 103.56 kcal/mol

T = 2300-6500 K
P = 50-150 kPa

Need data for $CO_2$ + $CO_2$ and P < 10 kPa