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Correlations for the NO $A^2\Sigma^+$ ($v'=0$) Electronic Quenching Cross-section

P. H. Paul, C. D. Carter, J. A. Gray, J. L. Durant Jr.,
J. W. Thoman and M. R. Furlanetto

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Correlations for the NO $A^2\Sigma^+$ Electronic Quenching Cross-section

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Abstract

This report summarizes recent progress in the development of a model for collisional electronic quenching of NO $A^2\Sigma^+$ by a number of molecules commonly found in combustion and aerodynamic systems. The model provides a means to reliably extend the measurement data base as well as suggesting simple analytic forms which can be used to provide physically plausible fits to the measurements. This report also contains a compilation of the available experimental data for collisional electronic quenching of NO $A^2\Sigma^+$.

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Contents

	<u>page</u>
I. Overview _____	6
II. The Harpoon Model _____	6
III. Limitations of the Model _____	9
IV. Recent Experimental Results _____	10
V. Future Updates to the Model and the Correlation Data Base _____	10
Appendix A. Tabulation of Fit Coefficients _____	11
Appendix B. Publications and Reports Related to this Work _____	12
Appendix C. Comparison of model predictions with measurements _____	13
Distribution List _____	22

I Overview

This report summarizes recent progress in the development of a model for collisional electronic quenching of NO $A^2\Sigma^+$ by a number of molecules commonly found in combustion and aerodynamic systems. The model is based on a simple physical picture of the process and is found to be quite successful at predicting both the magnitude and the temperature dependence in the quenching cross-section. The model provides a means to reliably extend the measurement data base as well as suggesting simple analytic forms which can be used to provide physically plausible fits to the measurements. This report also contains a compilation of the available experimental data for collisional electronic quenching of NO $A^2\Sigma^+$.

II The Harpoon Model

The collisional quenching rate, Q , of the NO $A^2\Sigma^+$ state by the perturber M is given by

$$Q = \frac{P_M}{k_B T} \langle u_M \rangle \langle \langle \sigma_M \rangle \rangle \quad (1)$$

Here P_M and T are the partial pressure of the perturber and temperature, respectively, k_B is Boltzmann's constant, and $\langle u_M \rangle \equiv \sqrt{8k_B T / \pi \mu_M}$ where μ_M is the reduced mass. The thermally averaged cross-section for the process is defined by

$$\langle \langle \sigma_M \rangle \rangle \equiv \langle u_M \sigma_M \rangle / \langle u_M \rangle \quad (2)$$

Here σ_M is the relative-velocity dependent cross-section for the process with the single brackets denoting a formal integration over the collision impact parameter, b , and the relative velocity, u . Invoking the conventional transformation, $\eta \equiv u_M^2 / (2k_B T / \mu_M)$, the velocity weighted thermal average is given by

$$\langle \langle \sigma_M \rangle \rangle = Z_{XM} \pi \int_0^\infty \eta \exp(-\eta) \int_0^\infty Z_M(\eta, b) db^2 d\eta \quad (3)$$

Here $Z_M(\eta, b)$ is the probability of the process occurring (i.e. collisional quenching) and Z_{XM} is the probability of subsequent separation of the molecules with the NO in a vibrational level of the ground state $X^2\Pi$.

We postulate that upon a collision, quenching of NO $A^2\Sigma^+$ occurs by transfer of an electron to the collision partner, M, at a crossing between the NO_A^- -M and the $NO^+ - M^+$ surfaces (here for notational simplicity we use the subscripts and superscripts to denote electronic states, i.e. NO_A or NO^+ are a shorthand for NO $A^2\Sigma^+$ or the ground electronic state ion $NO^+ X^1\Sigma^+$, respectively). The degree to which the electron transfer process is allowed is modeled using a Landau-Zener formalism modified to include the effect of vibrational overlaps in the electron donors and

electron acceptors. The potential energy surfaces are described using a classical multipole expansion with the molecular electrostatic properties obtained from the literature. Owing to the strong charge-charge attractive nature of the ionic surface, this surface can also make a crossing with the many surfaces corresponding to having the NO in the ground electronic state. Thus crossing from the $\text{NO}_A^- - \text{M}$ to the $\text{NO}^+ - \text{M}^-$ surface on approach acts as an entrance channel, the subsequent crossing from the $\text{NO}^+ - \text{M}^-$ to the $\text{NO}_X - \text{M}$ surface on separation acts as the exit channel for electronic quenching of $\text{NO } A^2\Sigma^+$ via the harpoon mechanism. Full evaluation of this model has been performed numerically (A complete description of the model and the details of the calculation are given by Paul *et al.* in the publications listed in Appendix B).

Using asymptotic solutions to equation 3 we identify six classes of collision partners:

- i Species having negative ions that are unstable towards auto detachment on a collisional time scale. For example, the adiabatic electron affinity of H_2 is nearly -3 eV, which is comparable to the auto detachment width of the negative ion. The large negative value for the affinity (here a negative affinity denotes an ion with a ground state energy above that for the corresponding neutral-molecule ground state) as well as the very short lifetime of the ion results in a prediction of a negligibly small cross-section for quenching by H_2 . The same behavior is predicted for quenching by He, Ne, Ar, Kr, Xe, CF_4 , SiF_4 and many of the saturated hydrocarbons. Species in this class are predicted to have a negligible quenching cross-section.
- ii Species having negative ions that are stable on a collisional time scale but having relatively negative affinities, e.g. N_2 and CO with measured affinities of -1.925 and -1.375 eV, respectively. These affinities are sufficiently negative to present an energy barrier to the harpoon process since the surface crossing is located on the inner repulsive wall of the covalent surface. At low temperatures, the cross-section will display classical Arrhenius behavior for a reaction subject to an energy barrier, i.e. $\langle\langle\sigma\rangle\rangle \propto \exp(-\epsilon/k_B T)$. At high temperature the slope of $\ln(\sigma(1/T))$ will increase as if the barrier height were decreasing with temperature owing to the contribution of vibrationally excited collision partners.

When the harpoon process is poorly allowed, energetically accessible secondary quenching mechanisms can be important. For example; quenching by CO is predicted to have a near-zero harpoon cross-section at room temperature, however the measured cross-section is of order $6 \text{ \AA}^2$ at 300 K. This can be accounted for by including the action of electronic to vibrational energy transfer between NO_A and $\text{CO } X^1\Sigma^+(v'' \approx 22)$. This process will operate for CO (owing to a dipole moment of 0.1 D) but not for H_2 or N_2 . Whereas at high temperatures the harpoon process becomes dominant. For such cases the probability in equation 3 is taken as a conditional linear sum of the probability for the harpoon process with that for the secondary process.

- iii Species with moderately negative affinities and strongly attractive covalent potentials, e.g. H_2O and CO_2 . A strongly attractive covalent surface can lead to the formation of a centripetal barrier. Collisions with particular combinations of impact parameter and relative velocity can cross this barrier leading to the formation of a long-lived 'orbiting' collision or 'collision-complex'. At low temperatures the quenching cross-section is dominated by such capture leading to a power-law decrease in the cross-section with

increasing temperature. Note that collisional capture in itself is not a quenching process, these mechanics only serve to bring the collision partners close enough to facilitate the harpoon process. At high temperatures the harpoon process becomes dominant and being sufficiently allowed yields a near constant cross-section with temperature.

- iv Species with moderately positive affinities and reasonably strong electron acceptor vibrational overlaps, e.g. NO, O₂, OH, NH, CH, H and O. For temperatures in excess of 300 K the strongly allowed surface crossing is at a sufficiently large radius that the cross-section is functionally constant with temperature. For species in this class the model predicts a very weak dependence of the cross-section on the NO_A vibrational state. Note that at very low temperatures, below 250 K (say), species in this class are expected to display a behavior similar to those found in class *iii*, i.e. an increase in the cross-section with decreasing temperature owing to the action of a collision-complex.
- v Species with large positive affinities and/or weakly allowed vibrational ground state neutral-negative ion vibrational overlaps, e.g. NO₂ and a number of the halocarbons. Here the surface crossing occurs at such a large radius that there is a poor overlap between the electronic wavefunctions. Electron transfer at large radii crossing, corresponding to the low vibrational states in the collision partner, is only probable for small relative radial velocities. Crossings that correspond to the higher vibrational states in the collision partners which are at smaller separations, provide a strongly allowed channel for the process at any relative velocity. Thus the low temperature cross-section is expected to fall almost inversely with temperature whereas the high temperature cross-section is expected to be nearly constant.
- vi Species in this class are strongly harpoon disallowed (i.e. class *i*) but have electronic energy levels near that of NO_A, e.g. NH₃, C₂H₂, C₂H₄, benzene, toluene, and CH₃I. We postulate that quenching by species in this class proceeds by electronic-to-electronic energy transfer. To date no satisfactory model for the temperature dependence of quenching by species in this class has been developed. However we expect collision-complex behavior at low temperatures and observe from the experimental data a relatively constant cross-section at high temperature.

The results of the full calculation can be provided in tabular form. However it is far more convenient to provide them in an easily evaluated functional form. To this end we have investigated analytic asymptotic solutions to the problem to suggest a set of functions for use as representations of the full numerical results. For the various classes delimited above, these are:

$$i) \quad \langle\langle\sigma_M\rangle\rangle \approx 0.0 \quad (4a)$$

$$ii) \quad \langle\langle\sigma_M\rangle\rangle \approx c_0 + c_1 \exp(-c_2 T_r/T) + c_3 \exp(-c_4 T_r/T) \quad (4b)$$

$$iii) \quad \langle\langle\sigma_M\rangle\rangle \approx c_0 \{ (1+\eta) \exp(-\eta) + c_1 \eta^{1/3} \gamma(5/3, \eta) \} \quad (4c)$$

$$\eta \equiv c_2 (T_r/T) + c_3 (T_r/T)^2$$

$$iv) \quad \langle\langle\sigma_M\rangle\rangle \approx c_0 \quad (4d)$$

$$v) \quad \langle\langle\sigma_M\rangle\rangle \approx c_0 + c_1 (T_r/T)^{c_2} \quad (4e)$$

$$vi) \quad \langle\langle\sigma_M\rangle\rangle \approx c_0 \{ (1+\eta) \exp(-\eta) + c_1 \eta^{1/3} \gamma(5/3, \eta) \} \quad (4f)$$

In equations 4c and 4f, $\gamma(a,x)$ is the incomplete gamma function. The free coefficients, c_i , in equations 4a through 4e were obtained by fitting to the result of the full harpoon calculation of equation 3. The only adjustable parameter in the calculation is the value of Z_{XM} which is set to match the experimental data and is bounded on the range of zero to unity. That is the reported values of c_0 through c_4 are those obtained from the theoretical calculation and the function is simply scaled to match the measurements at a single temperature. The coefficients in equation 4f were obtained by fitting to the results of a pure collisional-capture calculation with a gas-dynamic diameter hard-sphere core.

The fit coefficients for a number of collision partners of interest in combustion and aerothermodynamic research are given in Appendix A. A comparison of the model calculations to measured cross-sections is summarized in Appendix C.

III Limitations of the Model

The results of the model as given here are specific to quenching of NO $A^2\Sigma^+(v'=0)$. The harpoon model is inappropriate to describe quenching of the NO $B^2\Pi$ state. The model is appropriate to quenching of the $C^2\Pi$ and $D^2\Sigma^+$ states of NO however there will be a substantial change in the magnitudes of the cross-sections and in the temperature dependence from that given here for the $A^2\Sigma^+$ state.

The results given here are for a thermal rotational distribution in NO $A^2\Sigma^+$. Further the results are also for a thermal vibrational distribution in the collision partners. A discussion of the issues associated with rotational level-dependent quenching is given in Paul *et al.* (Appl. Phys. B, 1993). The issue of the effect of vibrational non-equilibrium is briefly commented upon in Paul *et al.* (AIAA J., 1994).

The results given here are specific to the temperature range of 275 to 3000 K with several exceptions (as noted) to higher temperatures. Care should be taken in any extrapolation of the results outside of the specified temperature range: At lower temperatures the model used for collision complex formation will break down leading to a substantial overprediction in the cross-section. At very high temperatures the actual cross-section is expected to be greater than that given by the model due to the contribution of free positive ions and to vibrationally excited neutral collision partners.

IV Recent experimental results

A number of recent measurements in this laboratory have provided additional input to calibrate and enlarge the model. Specifically:

Data on quenching of $\text{NO } A^2\Sigma^+(v'=0)$ by OH have been obtained in our shock tube. The measurements were performed by reacting lean $\text{H}_2 - \text{N}_2\text{O}$ mixtures to provide a source of OH. As of this writing, measurements have been performed at 1900 and 2300 K suggesting that the high temperature cross-section is independent of temperature with a value near $72 \text{ \AA}^2$. A full description of the measurements, the error analysis, and an a refined value is the subject of a forthcoming publication. The indicated value gives a value of $P_X \approx 0.53$ for quenching by OH. All other collision partners tested displayed values in the range $0.52 < P_X < 0.96$, with the specific exceptions of quenching by O and H which had much lower values. Two explanations for this have been considered: 1) the free radicals (e.g. OH, H and O) and $\text{NO } A^2\Sigma^+$ have magnetic moments due to unpaired electrons, magnetic coupling could introduce additional orientation effects not accounted for in the present model; and 2) the model assumes an isotropic dependence in the electronic matrix element that couples the neutral and ionic surface, this is obviously an oversimplification when the collision partner is a simple atomic species. The present result for OH would appear to support the second conjecture suggesting that the model predictions for quenching by CH and NH are high but by no more than a factor of two.

High temperature data on quenching of $\text{NO } A^2\Sigma^+(v'=0)$ by CH_4 and C_2H_6 confirms the prediction that these species are harpoon-disallowed collision partners, extending the previous data base covering the 300 to 1000 K range to 2000 K. A single measurement on quenching of $\text{NO } A^2\Sigma^+(v'=0)$ by C_3H_8 made at 300 K confirms the prediction that this species is also a harpoon-disallowed collision partner.

Formation of the negative ions of NH_3 , C_2H_4 , and C_2H_2 is poorly allowed. Thus, these species are unlikely to quench $\text{NO } A^2\Sigma^+$ via the harpoon mechanism. However all of these species have electronic states with energies at or near that of $\text{NO } A^2\Sigma^+$ and thus can quench via near resonant energy exchange. For these species measured cross-sections at 300 K are of order but do not exceed that predicted by collisional capture. However the high temperature cross-sections appear to level-out with some indication of a slow increase in cross-section with temperature for temperatures in excess of 1400 K. A full description of the measurements including those on CH_4 , C_2H_6 , and C_3H_8 , the error analysis, and a proposed model for near-resonant electronic quenching is the subject of the paper by Furlanetto et al. (J. Chem. Phys., 1994).

V Future updates to the model and the correlation data base

In future, refinements to the model and the incorporation of new experimental data will be reported in the literature. A FORTRAN program for evaluation of equations 4 combined with a data base containing all of the fitting coefficients has been developed. This code can be obtained by written request to Sandia National Laboratories or by electronic mail request to PHPAUL@CA.SANDIA.GOV.

Appendix A: Tabulation of Fit Coefficients

Fitting coefficients for quenching of NO A²Σ⁺ ^a

M	class	c0 (Å ²)	c1	c2	c3	c4	T _x ^b (K)	note
nobles	<i>i</i>	0.0					300-1900	c
CH ₄	<i>i</i>	0.0					300-1450	c
C ₂ H ₆	<i>i</i>	0.0					300-1900	c
C ₃ H ₈	<i>i</i>	0.0					300	c
CF ₄	<i>i</i>	0.0					300	c
SiF ₄	<i>i</i>	0.0					300	c
H ₂	<i>i</i>	0.0					300-1900	c
N ₂	<i>ii</i>	0.0	0.88	4.8	3.1	16.0	300-4500	v'=0,d
	<i>ii</i>	0.0	0.88	4.9	48.0	32.0	300-4500	v'=0,e
	<i>ii</i>	0.0	1.3	4.4	54.0	28.0	300,1900	v'=1,e
CO	<i>ii</i>	5.9	5.3	7.0	22.1	14.0	300-2300	v'=0,d
H ₂ O	<i>iii</i>	28.2	3.39	0.15	2.95		300-2300	
CO ₂	<i>iii</i>	54.2	0.95	3.24	0.18		300-2300	
N ₂ O	<i>iii</i>	59.0	0.99	3.98	0.16		300,1450,2300	
SO ₂	<i>iii</i>	52.0	1.68	0.11	3.01		300	
NO	<i>iv</i>	43.0					300-4500	c
O ₂	<i>iv</i>	25.1					300-2300	c
O	<i>iv</i>	26.8					2300	c
H	<i>iv</i>	12.8					2307	c
OH	<i>iv</i>	71.5					2320	c
NH	<i>iv</i>	58.0					na	c,d
CH	<i>iv</i>	101.0					na	c,d
NO ₂	<i>v</i>	82.0	9.0	0.54			300,1450,1900	
NH ₃	<i>vi</i>	50.7	0.91	10.74	6.5		300-2300	e,f
C ₂ H ₄	<i>vi</i>	5.0	7.1	1.07	0.72		300-1800	e,f
C ₂ H ₂	<i>vi</i>	28.0	0.81	13.1	14.85		300-1800	e,f

- a. Fits for v'=0 and 300-5000 K except as noted, T_r ≡ 300 K.
b. Temperature range for verification to experimental data.
c. Predicted to be independent of v' in NO_A for v' ≤ 3.
d. Fit to full model prediction only.
e. Best fit to experimental data.
f. not harpoon allowed.
na Not available.

Appendix B: Publications and reports related to this work

J. A. Gray, Rev. Sci. Inst. **61**, 1825 (1990)

J. A. Gray, P. H. Paul and J. L. Durant, Chem. Phys. Lett. **190**, 266 (1992).

J. W. Thoman Jr., J. A. Gray, J. L. Durant Jr. and P. H. Paul, J. Chem. Phys. **97**, 266 (1992).

P. H. Paul, J. A. Gray, J. L. Durant Jr. and J. W. Thoman Jr., Appl. Phys. B **57**, 249, (1993).

P. H. Paul, J. A. Gray, J. L. Durant Jr. and J. W. Thoman Jr., AIAA J. **32**, 1670 (1994).

P. H. Paul, J. Quant. Spectrosc. Radiat. Transfer, **51**, 511 (1994).

J. A. Gray, P. H. Paul, J. L. Durant Jr. and J. W. Thoman Jr. 'Collisional electronic quenching of NO $A^2\Sigma^+$ measured at high temperatures,' presented at: AIAA 31st Aerospace Sciences Conf., (AIAA, Washington, DC, 1993) paper AIAA-93-0924

P. H. Paul, J. A. Gray, J. L. Durant Jr. and J. W. Thoman Jr. 'Collisional electronic quenching rates for NO $A^2\Sigma^+$ (submitted to: Chem. Phys. Lett., Dec. 1994).

J. L. Durant Jr., P. H. Paul, M. R. Furlanetto, C. Flesner, J. W. Thoman Jr. and J. A. Gray, 'Collisional electronic quenching of NO $A^2\Sigma^+$ by H, O, and OH,' (in preparation for: Chem. Phys. Lett., 1995).

M. R. Furlanetto, J. W. Thoman Jr., J. A. Gray, P. H. Paul and J. L. Durant Jr., 'Near-resonant electronic energy transfer in the electronic quenching of NO $A^2\Sigma^+$ by hydrocarbons and ammonia,' (to appear as: J. Chem. Phys **101**, Dec. 1994).

M. R. Furlanetto, *Mechanisms for the High temperature collisional electronic quenching of nitric oxide ($A^2\Sigma^+$)*, Bachelors Dissertation (Williams College, Williamstown, MA, 1993).

P. H. Paul, J. A. Gray, J. L. Durant Jr. and M. R. Furlanetto, 'Collisional electronic quenching of OH $A^2\Sigma^+$ ($v'=0$) measured at high temperatures in a shock tube,' (submitted to J. Chem. Phys., Nov., 1994).

P. H. Paul, 'Vibrational energy transfer and collisional quenching of OH $A^2\Sigma^+$ ($v'=0$) measured at high temperatures in a shock tube,' (submitted to J. Phys. Chem., Dec., 1994).

Appendix C: Comparison of model predictions with measurements

The figures presented in this section show a comparison of the full model predictions to the available experimental data. The correlation coefficients listed in Appendix B and provided as a computer data base were obtained as best fits to the full model results. For all of the species except class *vi* the only freely adjustable parameter was the value of Z_{XM} all other coefficients were taken as those provided directly from the model. When experimental data were not available the value of P_X was set to unity. For class *vi* collision partners, the values of Z_{XM} and c_3 were adjusted to fit the measured cross-sections, all other parameters were taken as those directly provided by the model.

The experimental data plotted here were obtained from the following references.

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- ◆ M. R. Furlanetto, J. W. Thoman Jr., J. A. Gray, P. H. Paul and J. L. Durant Jr., 'Near-resonant electronic energy transfer in the electronic quenching of NO $A^2\Sigma^+$ by hydrocarbons and ammonia,' (to appear as: J. Chem. Phys. **101**, Dec. 1994).
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