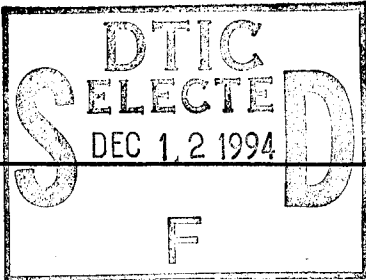


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6. AUTHOR(S) Dr Nicholas J. Turro					
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Final Technical Report for the period 1 September 1991 - 31 August 1994

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Final Technical Report

Stabilization and Control of High Energy Intermediates through Adsorption on Restricted Spaces

**Nicholas J. Turro
Columbia University
New York, NY 10027**

I. Abstract of technical progress

The research supported by this AFOSR grant has involved a multi-technique (NMR, ESR, laser flash photolysis, single photon counting, photoluminescence, tailor-made syntheses, computer simulation) approach to address problems concerning the structure and dynamics of transient high energy intermediates. These techniques have been employed to investigate the behavior of these intermediates when the latter are confined to the interfacial regions of restricted reaction spaces such as micelles, zeolites, cyclodextrins, water soluble polymers, and silica. Unique information has been obtained concerning the nature of interactions between the transients as substrates bound to receptors by non-covalent bonding. This information, in turn, has been exploited to investigate situations for which the transients display extraordinary dynamic properties such as reactivity which is controlled by the application of very weak applied magnetic fields.

II. Status of the research effort: Significant accomplishments

This AFOSR grant produced the following significant accomplishments through 53 published manuscripts during the funding period (numbers in parentheses refer to the list of publications below):

- A. The establishment of the mechanisms of a range of photophysical phenomena and photochemical reactions in restricted spaces including **cyclodextrins** (1, 20); **zeolites** (8, 11, 19, 26, 35); **micelles** (6, 10, 14); **polymers** (21, 32), **starburst dendrimers** (7, 42); and **DNA** (39, 50).
- B. The establishment of the mechanisms of reactions of electronically excited states in solution. The systems investigated include **acyclic ketones** (18, 27, 33, 34, 40, 41, 46, 49); and **cyclic ketones** (2, 36).
- C. The establishment of the mechanism of magnetic isotope and magnetic field effects on photochemical reactions in restricted spaces. The systems studied include **magnetic isotope effects** on radical pair reactions in homogeneous solution (13) and in micelles (4, 5, 12, 23, 24, 28, 44, 48); and the use of **chemically induced dynamic nuclear polarization** as a probe of magnetic effects (17, 43).
- D. The use of steady state and time resolved electron spin resonance to investigate photoreactions in solution and in restricted spaces. The systems investigated include the use of spin probes to investigate **restricted spaces** (15, 22); the use of electron spin polarization to examine **reaction mechanisms** (29, 31, 37, 38); and the use of electron spin resonance to investigate the **mechanism**

of **polymer stabilization** by hindered amines (45), and the structure of **triplet states** (52, 53).

E. The use of time resolved laser flash spectroscopy to investigate the mechanism of electron transfer reactions in **solution** (3, 9, 51) and in **restricted spaces** (47).

III. Cumulative list of publications

1. V.P. Rao, M.B. Zimmt, and N.J. Turro, "Photoproduction of Remarkably Stable Benzylic Radicals in Cyclodextrin Inclusion Complexes," *J. Photochem. Photobiol. A: Chem.* **1991**, 60, 355-360.
2. N. Han, K.C. Hwang, X.G. Lei, and N.J. Turro, "Photochemistry of 2,2,13-Trimethylcyclododecanone and 2,2,12,12-Tetraethylcyclododecanone: Product Distribution, Photo-CIDNP, and Magnetic Isotope Effect," *J. Photochem. Photobiol. A: Chem.*, **1991**, 61, 35-46.
3. W.-S. Cheng, N.J. Turro I.R. Gould, and S. Farid, "Effect of External Pressure on Photoinduced Electron-Transfer Reactions in the Marcus Inverted Region," *J. Phys. Chem.*, **1991**, 95, 7752-7757.
4. E.N. Step, A. Buchachenko, and N.J. Turro, "Magnetic Isotope Effect in the Reaction of Disproportionation of Radical Pairs," *Chem. Phys. Letts.*, **1991**, 186, 405-409.
5. V. Tarasov, N.D. Ghatlia, A. Buchachenko, and N.J. Turro, "Photostereoisomerization and the Magnetic Isotope Effect," *J. Phys. Chem.*, **1991**, 95, 10220-10229.
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11. M.A. Garcia-Garibay, X.G. Lei and N.J. Turro, "Radical Scavenging in Zeolite Media," *J. Am. Chem. Soc.*, **1992**, 114, 2749-2751.

12. E.N. Step, A.L. Buchachenko, and N.J. Turro, "Magnetic Effects in the Photolysis of Micellar Solutions of Phenacylphenylsulfone," *Chem. Phys.*, **1992**, *162*, 189-204.
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-Papers In Press

1. M.R. Arkin, Y. Jenkins, C.J. Murphy, N.J. Turro, and J.K. Barton, "Metallointercalators as Probes of the NDA π -way," *ACS Advances in Chemistry Series*, **1994**.
2. N.J. Turro, A. Evenzahav, and K.C. Nicolaou, "Photochemical Analogue of the Bergman Cycloaromatization Reaction," *Tetrahedron Letts.*, in press.
3. E.N. Step, N.J. Turro, P.P. Klemchuk, and M.E. Gande, "Model Studies on the Mechanism of HALS Stabilization," *Macromolecular Symposia*, in press.
4. E.N. Step and N.J. Turro, "Photolysis of Ketones in Oxygen-Saturated Micellar Solution: Oxygen Scavenging of C-Centered Radicals in Microheterogeneous Media," *J. Photochem. Photobiol. A: Chem.*, in press.
5. V.F. Tarasov, E.G. Bagranskaya, I.A. Shkrob, N.I. Avdievich, N.D. Ghatlia, N.N. Lukzen, N.J. Turro and R.Z. Sagdeev, "Examination of the Exchange Interaction Through Micelle Size. III. Stimulated Nuclear Polarization and Time Resolved Electron Spin Resonance Spectra from the Photolysis of Methyl Deoxybenzoin in Alkyl Sulfate Micelles of Different Sizes," *J. Am. Chem. Soc.*, in press.

6. N.J. Turro, "Supramolecular Organic and Inorganic Photochemistry: Radical Pair Recombination in Micelles, Electron Transfer on Starburst Dendrimers, and the Use of DNA as a Molecular Wire," *Pure & Appl. Chem.*, in press.

-Papers Submitted for Publication

1. A.L. Buchachenko, L.V. Ruban, E.N. Step and N.J. Turro, "Catalysis of the Radical Recombination Reaction," *Chem. Phys. Letts.*, submitted.
2. J. Casper, I.V. Khudyakov, N.J. Turro and G.C. Weed, "ESR Study of Lophyl Free Radicals in Dry Films," *Macromolecules*, submitted.
3. N.J. Turro and I.V. Khudyakov, "Time Resolved Electron Spin Resonance and Laser Flash Spectroscopy Investigation of the Photoreduction of Anthraquinone-2,6-disulfonic Acid, Disodium Salt by Sodium Sulfite in Aerosol OT Reverse Micelles," *J. Phys. Chem.*, submitted.
4. L.S. Schulman, S.H. Bossmann and N.J. Turro, "Analysis of Luminescence Quenching on Calf Thymus DNA," *J. Phys. Chem.*, submitted.
5. C. Turro, S.H. Bossmann, S. Niu, D.A. Tomalia and N.J. Turro, "Quenching of $^*Ru(phen)_3^{2+}$ by $Co(phen)_3^{3+}$ Bound to Starburst Dendrimer Surfaces: Evidence for Intra-Starburst Quenching at High Host Concentration," *J. Phys. Chem.*, submitted.
6. N.J. Turro, A.L. Buchachenko and V.F. Tarasov, "Modern Supramolecular Photochemistry: How Complicated Can It Be to Make a Carbon-Carbon Bond Between Two Reactive Carbon Centered Radicals in a Collision Complex?", *Acc. Chem. Res.*, submitted.

IV. List of professional personnel:

Dr. Gabriella Caminati
Dr. Naresh Ghatlia
Mr. Matthew Lipson
Mr. Zhi Liu

Ms. Shufang Niu
Mr. Chung-Hsi Wu

Ph. D. 1993 Ms. Shufang Niu Thesis title: "Photoinduced Electron Transfer Quenching of Excited Ruthenium(II) Complexes by Metal Complexes Bound to Anionic Micelles, Starburst Dendrimers, DNA, and Sodium Poly(styrenesulfonate)"

Ph.D. 1994 Mr. Chung-Hsi Wu Thesis title: "Photochemistry and Photophysics of Micellized Radical Pair Systems"

Ph.D. 1994 Mr. Matthew Lipson Thesis title: "Conformational Control of the Photochemistry and Photophysics of Benzophenone and Diphenylacetone"

V. Coupling Activities

Papers presented at meetings, conferences, seminars, etc.

Year 1: September 1, 1991 to August 31, 1992

Gramatakakis Lecturer, Swiss Photochemical Society, Lausanne, Sept. 1991
Dow Distinguished Lecturer, Michigan State Univ., Oct. 1-4, 1991
Appleton Lecturer, Brown University, Nov. 18-19, 1991
Yale University, Dec. 11, 1991
Castle Lecturer, Univ. South Florida, Tampa, FL Dec. 4, 1991
University of Utah, Feb. 25, 1992
Colorado State Univ., March 18, 1992
Linus Pauling Lecturer, Oregon State Univ., March 9-13, 1992
Stanley Cristol Lecturer, Univ. Colorado, March 16, 1992
R.T. Major Lecturer, Univ. Connecticut, April 16, 1992
Lemieux Lecturer, Univ. Ottawa, May 4-6, 1992
Plenary Lecturer, VIth Intern. Symp. on Magnetic Resonance in Colloid and Interface Sci., Florence, IT June 22-26, 1992.
Plenary Lecturer, "Radicals in Chemistry Symposium" ACS National Meeting, Washington, D.C. August 26, 1992.

Year 2: September 1, 1992 to August 31, 1993

Univ. No. Carolina, Chapel Hill, Sept. 25, 1992
Edward Arnett Symposium, Duke University, Sept. 26, 1992
Henry Kwivila Lecturer, SUNY Albany, October 13, 1992
Drexel University, December 2, 1992
Ramapo College, December 4, 1992
George Washington Univ., January 14, 1993
Powell Lecturer, University of Richmond, February 12, 1993
Broberg Lecturer, North Dakota State Univ., April 5-6, 1993
University of Virginia, April 23, 1993
Welch Foundation Lecturer May 5-7, 1993
Intern. Conf. Bio-Inorganic Chem. Lecturer in San Diego, CA, August 23-27, 1993

Year 3: September 1, 1993 to August 31, 1994

Dow Distinguished Lecturer, University of Western Ontario, Sept. 20-22, 1993
Dauben Lecturer, Univ. Calif. at Berkeley, November 2, 1993
G.L. Closs Memorial Lecturer, November 15, 1993
University of Amsterdam, NL May 30, 1994
University of Utrecht, NL May 31, 1994
University of Delft, NL June 1, 1994
Havinga Memorial Award Lecture, Leiden, NL, June 2, 1994
Porter Medal Award Lecture, IUPAC Photochemistry Conf., Prague, Czech Republic July 22, 1994
XVth Solar Energy Conference, Interlaken, Switzerland, July 24-29, 1994
Plenary Lecturer, IIIrd Inter. Symp. on Magnetic Field and Spin Effects in Chemistry & Related Phenomena, Chicago, Sept. 26, 1994

**Consultative & advisory functions to other laboratories, especially DoD labs:
Institutions, locations, dates and names of individuals involved**

Year 1. The PI spoke to AFOSR research personnel at Edwards Air Force Base in California concerning the use of EPR techniques. The PI also had discussions with Dr. Frederick Hedberg (Bolling Air Force Base) on the possible use of

photochemical methods and adsorption on zeolites as a possible means of destroying hazardous chemicals.

Year 2. The PI had several conversations with Dr. Hedberg of AFOSR concerning the possible use of zeolites to terminate fires and as substrates for photomineralization of hazardous chemicals of interest to AFOSR. Two AASERT fellows performed summer undergraduate research in the PI's laboratory during the summer of 1993.

Year 3. The PI attended a meeting at Tyndall Air Force Base, Florida on May 12 and 13, 1994 to discuss AFOSR's mission in "Subsurface Fate & Transport in Environmental Quality." He had numerous fruitful conversations with AFOSR researchers concerning the strategy for investigating the fate, transport, and assessment program for toxic materials. During this meeting, a number of connections were made with other PIs, in particular Dr. Howard Hanley, who will be supplying the Columbia group with clay materials and will be collaborating on neutron scattering investigations, Dr. Theodore Mill and Professor John Hassett who will be collaborating on the fate of quadracyclane adsorbed in simulated natural waters and adsorbed on clays.

On August 23, 1994 the PI met with Dr. Frederick Hedberg, Dr. Ted Mill, Professor Patrick Sullivan and Professor John Hassett at SRI's Washington Office in Arlington, VA . At this meeting the preliminary results with quadracyclane were discussed and a priority was placed on the next series of experiments. Plans were also made to obtain from Dr. Robert Schmitt of SRI International samples of potassium dinitramide for testing in our laboratory.

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STINFO Program Manager

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