



## Biographical Sketch of Michael Kasha

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Michael Kasha was born December 6, 1920, of Ukrainian immigrant parents who had settled in a Ukrainian community in Elizabeth, New Jersey. His parent's educational opportunities were limited to 3 or 4 years of rudimentary education in the Austria-Hungary Empire. On the death of his father at the age of 87, he described his father Stefan as "a man who understood more about the world and knew more arts and crafts than most men I have known." Michael Kasha won scholarships permitting him to attend the Cooper Union Night School of Engineering while working as a laboratory assistant to Karl Folkers and John Keresztesy at the Merck Research Laboratories in Rahway. He was encouraged to go into pure science in a public university and received his B.S. in chemistry degree from the University of Michigan in 1943. He then went to the University of California and completed his Ph.D. in February 1945 on the triplet states of organic molecules. He was the last graduate student to complete work under the famous physical chemist G. N. Lewis.

After some years of postdoctorate research in Berkeley, Chicago, and England, he was appointed professor of physical chemistry in the nascent Florida State University in September 1951, a position in which he is still actively engaged.

In 1960 he founded the Institute of Molecular Biophysics as an interdisciplinary research facility, featuring, especially, research interaction between chemical physics and molecular biology. Among the most powerful influences in Kasha's interests in these directions was a series of international conferences of interdisciplinary character, especially the great Boulder, Colorado, conference organized by Francis Schmitt in 1958 (published as the *Biophysical Science Study Program*) and the Institut de la Vie conference series organized by Maurice Marois, continuing today (*From Theoretical Physics to Biology*). Equally influential were summer conferences organized by Albert Szent-Gyorgyi at the Marine Biological Laboratories in Woods Hole, Massachusetts, which Kasha participated in over the decade from 1960 to 1970.

Michael Kasha has worked consistently on research extending the application of molecular electronic theory and spectroscopic interpretation to biomolecular systems. His work covers fundamental orbital and spin characterization of molecular electronic states, molecular energy transfer, and the molecular exciton model applied to molecular dimers, polymers, and lamellar structures, elucidation of the role of singlet molecular oxygen in chemistry and biology, and, currently, proton-transfer phenomena in organic and biological systems, especially the effect of hydration on proton-relay systems.

One of the strongest contributions made to science by Michael Kasha is the impressive list of scientists who came under his inspiring influence. Among those who earned their Ph.D. degrees under his guidance are Ralph Becker (Houston), Donald S. McClure (Princeton), Seán P. McGlynn (Louisiana State University), Eion G. McRae (Bell Laboratories), Mostafa El-Sayed (UCLA), M. Ashraf El-Bayoumi (Alexandria University), Stewart J. Strickler (Colorado), Bryan R. Henry (Guelph University), and Pi-Tai Chou (University of South Carolina). Among his postdoctorate research associates are included Enrico Clementi (IBM, Kingston), Glenn A. Crosby (Washington State University), Lionel Goodman (Rutgers), Jozef Heldt (University of Gdańsk), Robin Hochstrasser (University of Pennsylvania), Ahsan U. Khan (Boston), William Rhodes (Florida State University), and Ian G. Ross (Canberra).

The essay "Four Great Personalities of Science—G. N. Lewis, J. Franck, R. S. Mulliken, and A. Szent-Györgyi" by Michael Kasha reveals his remarkable good fortune of having worked (or interacted) for a long period with four of the world's greatest scientists, three of them Nobel Laureates, and the fourth (Lewis) considered a Nobel Laureate by all of his colleagues, disciples, and professional admirers.

In 1963 Kasha was elected a Fellow of the American Academy of Arts and Sciences (established in Boston by Benjamin Franklin). In 1971 he was elected to membership in the National Academy of Sciences (established in Washington by Abraham Lincoln). He is the first Floridian to be elected to these national academies and also was elected by President Jimmy Carter to the National Science Board (1979–84) as the first Floridian to serve. Since 1962, he has been the Robert O. Lawton Distinguished Professor at the Florida State University.

In 1948 Michael Kasha married Lilli Cohn, a native of Danzig. She is a linguist, with a pragmatic knowledge of nine languages, including Japanese and Chinese. Their son, Nicolas, has earned degrees in environmental science, physics, and music.

Michael Kasha is known for the diversity of his interests aside from physical chemistry. On the FSU campus he was director of the Institute of Molecular Biophysics for 20 years. He was chairman of the Science Center Planning Committee for 10 years, whose function was laying out in the 1960s the \$40 million FSU Science Center. Kasha holds patents in acoustical and mechanical engineering and is active in design work. His work on string-soundboard instruments is widely known and covers instruments from guitars to the violin family and to

the harp, harpsichord, and piano. His interest in flower color genetics led him to a treatment of Mendelian genetics by tensor matrix product algebra, to a long series of flower pigment researches culminating in a genetic synthesis scheme for producing blue flowers, and ultimately to the development of the proton-transfer laser based on a flavonol co-pigment.

Dr. Kasha also has had a life-long interest in technical photography and is developing an archeological art project involving various photo-silk-screen printing techniques. His 39 years of residence in a former wilderness area on a lake near Tallahassee has led him to undertake writing a book of 35 chapters on natural history and cultural history of the area, illustrated by a graphic artist in St. Augustine, Florida, where sample installments were published serially.

### **Biographical Professional Summary**

**Date and Place of Birth:** December 6, 1920; Elizabeth, New Jersey

#### **Education and Professional Experience:**

B.S., Chemistry, University of Michigan, Ann Arbor, 1943.

Ph.D., Chemistry, University of California, Berkeley, 1945.

Research Chemist, Plutonium Project, Berkeley, February 1944–46.

Research Associate, University of California, Berkeley, 1947–49.

Postdoctoral Fellow, Department of Physics, University of Chicago, 1949–50.

Professor of Chemistry, Florida State University, 1951 to present.

Chairman, Department of Chemistry, FSU, 1959–62.

Founder, Institute of Molecular Biophysics, FSU, 1960. Director, 1960–80.

National Science Board (Appointment by President Carter), 1979–84.

Conference Committee, Institut de la Vie, Paris, France, 1980–89.

Foreign Councilor, Institute of Molecular Science, Okazaki, Japan, 1982–85.

Scientific and Technical Advisory Committee, Argonne National Lab., 1983–84.

Science Advisor to Governor Bob Graham, Florida, 1983–87.

#### **Distinctions and Awards:**

Robert O. Lawton Distinguished Professor, FSU, 1962–.

Fellow of the American Academy of Arts and Sciences, 1963.

National Academy of Sciences, 1971.

Corresponding Member, Brazilian Academy of Sciences.

Doctor of Science, *Honoris-causa*, Gonzaga University, May 1988.

John Simon Guggenheim Memorial Fellow, 1950–51.

Unrestricted Venture Grant, Petroleum Research Fund, ACS, 1958.

Florida Award, Florida Section, ACS, 1961.  
Charles F. Kettering Research Award, 1963–69.  
Evans Award, Teaching and Research in Chemistry, Ohio State University, 1969.  
Southern Chemist Award, Memphis Section, ACS, November 1974.  
Kasha Symposium on Electronic Proc. and Energy Trans., J. Phys. Chem., Sept. 1976.  
Outstanding Scientist of the Year Medal, Florida Academy of Sciences, 1977.  
Randolph T. Major Medal, Univ. of Connecticut and Merck and Co., Inc., 1981.  
George Porter Medal, Selection by Panel from European, Interamerican and Japanese Photochemical Societies, 1990.

#### Visiting Professorships:

Special Lecturer, University of Manchester, England, 1950–51.  
Visiting Professor of Chemistry, Harvard University, 1959–60.  
Visiting Professor of Biophysics, Harvard University, October 1961.  
Visiting Professor of Chemistry, University of Michigan, November 1969.  
Visiting Professor, Inst. de Fisica e Quimica, São Carlos, Brazil, Summer 1976.  
Japan Society for Promotion of Science (Lecturer in 24 universities), Fall 1979.  
Guest Professor, Ørsted Institute, University of Copenhagen, May–June 1982.  
Visiting Professor, Inst. of Chem. Physics, Jilin University, Changchun, People's Republic of China, Fall 1984.

#### Honorary Lectureships:

Phillips Lecturer, Haverford College, Pennsylvania, 1959.  
Reilly Lecturer, University of Notre Dame, Indiana, 1959.  
S. C. Lind Lecturer, Oak Ridge National Laboratory, 1961.  
Dreyfus Lecturer, Dartmouth College, May 1971.  
Venable Lecturer, University of North Carolina, November 1971.  
IBM Lecturer, Williams College, September 1972.  
Coover Lecturer, Iowa State University, October 1974.  
Mead-Swing Lecturer, Oberlin College, May 1980.  
Jean Day Lecturer, Rutgers University, May 1983.  
Probst Lecturer, Southern Illinois University, May 1983.  
G. N. Lewis Lecturer, University of California, Berkeley, October 1983.  
Marie Curie Lecturer, Texas Woman's University, Denton, November 1983.  
Welch Lecturer, Houston, Kingsville, San Antonio (Texas), March 1985.  
W. M. Spicer Memorial Lecture in Chemistry, Georgia Inst. of Tech., March 1985.

Frontiers of Chemistry Lecture, Wayne State University, September 1986.  
Lemieux Lecturer, University of Ottawa, December 1986.  
O'Leary Distinguished Scientist Lecturer, Gonzaga University, April 1987.  
DOW Senior Lecturer, University of Western Ontario, May 1988.  
Milton Kahn Lecturer, University of New Mexico, May 1988.  
Robert Mulliken Lecturer, University of Georgia, May 1990.

### Publications

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2. Phosphorescence in fluid media and the reverse process of singlet-triplet absorption, G. N. Lewis and M. Kasha, *J. Am. Chem. Soc.* **67**, 994 (1945).
- \*3. Ionic equilibria and reaction kinetics of plutonium ions in hydrochloric acid solution, M. Kasha and G. E. Sheline, chap. 3.14, pp. 180–226.
- \*4. The pentapositive oxidation state of plutonium, R. E. Connick, M. Kasha, W. H. McVey, and R. E. Sheline, chap. 3.15, pp. 227–240.
- \*5. Reactions between plutonium ions in perchloric acid solution: Rates, mechanisms, and equilibria, M. Kasha, chap. 3.100, pp. 295–334.
- \*6. Spectrophotometric studies of plutonium in aqueous solution, R. E. Connick, M. Kasha, W. H. McVey, and G. E. Sheline, chap. 4.20, pp. 559–601.
- \*7. Magnetic susceptibilities of plutonium in various oxidation states in aqueous solution, M. Calvin, M. Kasha, and G. E. Sheline, chap. 4.23, pp. 632–634.
8. (Note) On the correlation of the spectroscopic and thermal energy differences between the fluorescence and phosphorescence levels of dye molecules, M. Kasha and R. E. Powell, *J. Am. Chem. Soc.* **69**, 2909 (1947).
9. Phosphorescence and the role of the triplet state in the electronic excitation of complex molecules, M. Kasha, *Chem. Rev.* **41**, 401 (1947).
10. (Note) On the properties of gelatin-dye phosphors and the continuum theory of szent-Gyorgyi, M. Kasha, *Science* **107**, 556 (1948).
11. Chemical notes on the coloring matter of Chihuahua textiles of pre-Columbian Mexico, M. Kasha (*Textiles of Pre-Columbian Chihuahua*, by Lila M. O'Neale), Carnegie Institution of Washington, Publication 574, Contribution 45, p. 151 (1948).
12. Transmission filters for the ultraviolet, M. Kasha, *J. Opt. Soc. Am.* **38**, 929 (1948).
13. Fabrication of boric acid glass for luminescence studies, M. Kasha, *J. Opt. Soc. Am.* **38**, 1908 (1948).
14. (Note) Vibrational fine structure in the absorption spectra of uranyl and plutonyl ions in aqueous solution, M. Kasha, *J. Chem. Phys.* **17**, 349 (1949).
15. The metastability of the lowest excited singlet level of naphthalene, M. Kasha and R. V. Nauman, *J. Chem. Phys.* **17**, 516 (1949).
16. Photomagnetism. Determination of the paramagnetic susceptibility of a dye in its phosphorescent state, G. N. Lewis, M. Calvin, and M. Kasha, *J. Chem. Phys.* **17**, 804 (1949).

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\*Research papers in *The Transuranium Elements*, edited by Seaborg, Katz, and Manning (National Nuclear Energy Series; Plutonium Project Record, Division IV), Vol. 14B, Part I, McGraw-Hill, New York, 1949. (Original dates of Manhattan District papers, July 1944–August 1946.)

17. Characterization of electronic transitions in complex molecules, M. Kasha, Faraday Soc. Discuss. **9**, 14 (1950).
18. Collisional perturbation of spin-orbital coupling and the mechanism of fluorescence quenching. A visual demonstration of the perturbation, M. Kasha, J. Chem. Phys. **20**, 71 (1952).
19. *Discussion on Basic Mechanisms in Radiobiology*, M. Kasha, National Research Council Publication No. 305, Nuclear Science Series, Magee, Kamen, and Platzman, Eds. (National Research Council, Washington, DC, 1953).
20. (Note) Lowest triplet levels of the polyacenes, S. P. McGlynn, M. R. Padhye, and M. Kasha, J. Chem. Phys. **23**, 593 (1955).
21. Luminescence spectroscopy of molecules and the photosynthetic system, R. S. Becker and M. Kasha, in *The Luminescence of Biological Systems*, F. H. Johnson, Ed. (American Association for the Advancement of Science, Washington, DC, 1955), pp. 25-47.
22. (Note) Luminescence spectroscopy of porphyrin-like molecules including the chlorophylls, R. S. Becker and M. Kasha, J. Am. Chem. Soc. **77**, 3669 (1955).
23. The role of hydrogen bonding in the  $n \rightarrow \pi^*$  blue-shift phenomenon, G. J. Brealey and M. Kasha, J. Am. Chem. Soc. **77**, 4462 (1955).
24. (Note) Interpretation of the lowest frequency electronic absorption bands of inorganic molecules of type  $\text{XO}_2^{z-}$  as  $n \rightarrow \pi^*$  transitions, S. P. McGlynn and M. Kasha, J. Chem. Phys. **24**, 481 (1956).
25. Confirmation of the anomalous fluorescence of azulene, G. Viswanath and M. Kasha, J. Chem. Phys. **24**, 574 (1956).
26. The lowest triplet state of anthracene, M. R. Padhye, S. P. McGlynn, and M. Kasha, J. Chem. Phys. **24**, 588 (1956).
27. Spin intercombinations in molecules, M. Kasha and S. P. McGlynn, Ann. Rev. Phys. Chem. **7**, 403 (1956).
28. (Note) Spin-orbital perturbation in Cyanine dyes absorbed on surfaces containing high-Z atoms, E. Clementi and M. Kasha, J. Chem. Phys. **26**, 967 (1957).
29. Review: Quantum chemistry, an introduction (by Walter Kauzmann), M. Kasha and G. A. Crosby, J. Chem. Ed. **34**, 412 (1957).
30. Intramolecular energy transfer in ytterbium organic chelates, G. A. Crosby and M. Kasha, Spectrochim. Acta **10**, 377 (1958).
31. The observation and assignment of the lowest multiplicity-forbidden transition in pyrazine, L. Goodman and M. Kasha, J. Mol. Spectrosc. **2**, 58 (1958).
32. *Molecular Electronic Bibliography*, L. Kasha and M. Kasha, (Publishers Press [Rose Printing Co.], Tallahassee, FL, 1958), Vol. 1.
33. (Note) The enhancement of phosphorescence ability upon aggregation of dye molecules, E. G. McRae and M. Kasha, J. Chem. Phys. **28**, 721 (1958).
34. Spin-orbital interaction in N-heterocyclic molecules. General results in a cylindrical potential approximation, E. Clementi and M. Kasha, J. Mol. Spectrosc. **2**, 297 (1958).
35. Review: Excited states in chemistry and biology (by C. Reid), M. Kasha, J. Chem. Ed. **35**, 583 (1958).
36. Relation between exciton bands and conduction bands in molecular lamellar systems, M. Kasha, Rev. Modern Phys. **31**, 162 (1959). (Also in *Biophysical Science*, Oncley, Schmitt, Williams, Rosenberg, and Bolt, Eds. (Wiley, New York, 1959), p. 162.
37. Paths of molecular excitation, M. Kasha, in *Bioenergetics*, L. G. Augenstine, Ed., Radiat. Res., Suppl. **2**, 243 (1960).
38. Ultraviolet radiation effects: Molecular photochemistry, M. Kasha, in *Comparative Effects of Radiation*, M. Burton, J. S. Kirby-Smith, and J. L. Magee, Eds. (Wiley, New York, 1960), pp. 72-96.
39. (Note) Basic theories of energy and electron transfer, M. Kasha, in *Fast Fundamental Transfer Processes in Aqueous Biomolecular Systems*, F. O. Schmitt, Ed., Bulletin (Dept. of Biology, Massachusetts Institute of Technology, Cambridge, 1960), pp. 3-6.

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41. Introductory Remarks at the Symposium on Molecular Electronic Spectroscopy, M. Kasha, *J. Mol. Spectrosc.* **6**, 1 (1961).
42. The nature and significance of  $n \rightarrow \pi^*$  transitions, M. Kasha, in *Light and Life*, W. D. McElroy and B. Glass, Eds. (Johns Hopkins University Press, Baltimore, MD, 1961), pp. 31–64.
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49. *Theory of Molecular Excitons*, A. S. Davydov (McGraw-Hill, New York, 1962), 184 pp. (Translated from the Russian text by M. Oppenheimer, Jr. and M. Kasha.)
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51. *Horizons in Biochemistry*, M. Kasha and B. Pullman, Eds. (Academic Press, New York, 1962), 606 pp.
52. Quantum chemistry in molecular biology, M. Kasha, in *Horizons in Biochemistry*, M. Kasha and B. Pullman, Eds. (Academic Press, New York, 1962), pp. 583–599.
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64. (Note) Physical theory of chemiluminescence in systems evolving molecular oxygen, A. U. Khan and M. Kasha, *J. Am. Chem. Soc.* **88**, 1574 (1966).



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66. Theory of molecular luminescence, M. Kasha, in *Proceedings of the International Conference on Luminescence*, G. Szigeti, Ed. (Akademiai Kiado, Budapest, 1968), Vol. I, pp. 166-182.
67. Radiationless molecular electronic transitions, B. R. Henry and M. Kasha, *Annu. Rev. Phys. Chem.* **19**, 161 (1968).
68. Criteria for maximizing steady state population of the lowest excited triplet state, B. R. Henry and M. Kasha, *J. Mol. Spectrosc.* **26**, 536 (1968).
69. Luminescence spectra of transition metal complexes, F. Zuloaga and M. Kasha (Caracas Symposium), *Photochem. Photobiol.* **7**, 549 (1968).
70. Correlation of orbital classification of molecular electronic transitions with transition mechanism: The aromatic amines, M. Kasha and H. R. Rawls, *Photochem. Photobiol.* **7**, 561 (1968).
- 70a. A new look at the history of the classic guitar, M. Kasha, *Guitar Rev.* **30**, 2 (1968).
71. Electronic consequences of vibrational deficiency in polyatomic molecules, P. J. Gardner and M. Kasha, *J. Chem. Phys.* **50**, 1543 (1969).
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74. Excited-state two-proton tautomerism in hydrogen-bonded N-heterocyclic base pairs, C. A. Taylor, M. A. El-Bayoumi, and M. Kasha, *Proc. Natl. Acad. Sci. U.S.A.* **63**, 253 (1969).
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77. Observation of simultaneous electronic transitions for rare-earth ion pairs in aqueous solution, J. M. Marrs and M. Kasha, *Chem. Phys. Lett.* **6**, 235 (1970).
78. The physics, chemistry, and biology of singlet molecular oxygen, M. Kasha and A. U. Khan, *Ann. N.Y. Acad. Sci.* **171**, 5 (1970).
79. An optical-residue singlet-oxygen theory of photocarcinogenicity, A. U. Khan and M. Kasha, *Ann. N.Y. Acad. Sci.* **171**, 24 (1970).
80. The competition among biprotonic phototautomerism, excimer formation, and proton tunnelling in DNA base-pairs, M. Kasha, P. Horowitz and M. A. El-Bayoumi, in *Molecular Spectroscopy, Modern Research*, K. N. Rao and C. W. Mathews, Eds. (Academic Press, New York, 1972), pp. 287-296.
81. Photo-excitation effects in molecules concealed in normal action spectra, M. Kasha, in *Horizons of Bioenergetics*, A. San Pietro and H. Gest, Eds. (Academic Press, New York, 1972), pp. 171-172.
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83. The charge-transfer band in the ultraviolet spectrum of cyclopropyl ketones, A. Y. Meyer, B. Muel, and M. Kasha, *J. Chem. Soc., Chem Commun.*, 401 (1972).
84. Role of photons in the storage of energy and resonance transfer, M. Kasha, in *From Theoretical Physics to Biology*, M. Marois, Ed. (S. Karger, Basel, 1973), pp. 156-164.
85. Multiple excitation in composite molecules: Biprotonic phototautomerism, M. Kasha, in *Excited States of Matter*, C. W. Shoppee, Ed. (Grad. Studies, Texas Tech University, 1973), pp. 5-19.
86. A survey of multiple excitation in composite molecules, M. Kasha and M. A. El-Bayoumi, in *Physical Mechanisms in Radiation Biology*, R. D. Cooper and R. W. Wood, Eds. U.S. Atomic Energy Commission, CONF-721001, (1974), pp. 126-130.
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### Research Contributions

Michael Kasha has made his main research career in molecular electronic spectroscopic phenomenology. In his research span of 40 years, he has contributed a stream of key discoveries that have had an enormous influence on the development of the subjects of molecular electronic spectroscopy and molecular

photochemistry, with countless applications to organic chemistry, biophysics, and technology. The phenomena, concepts, language, and theoretical treatment developed in his laboratory could literally be described as the backbone of molecular electronic phenomenology as it has evolved in the last three decades.

### *Triplet State of Organic Molecules — Photomagnetism*

In 1944 and 1945, two key papers were published by G. N. Lewis and M. Kasha on triplet states of organic molecules. To understand the atmosphere of scientific hostility with which these papers were greeted, and the later impact they had on the field, a few retrospective observations are needed. From the experimental point of view, the observation of phosphorescence of organic molecules in crystals at liquid air temperature, or dissolved in rigid glass media, was known since the experiments of Dewar before 1900. These strange phenomena were ranked with those of inorganic doped solids as representing solid-state phenomena associated with impurity traps, etc. In the early 1940s, no spectroscopist or quantum chemist considered making use of such data as representing an intrinsic molecular property. Another aspect of the period was the firm conviction by theoreticians of the unobservability of singlet-triplet transitions in molecules made up of low atomic number atoms (C,N,O). H. Sponer and E. Teller in a famous paper in *Reviews of Modern Physics* (1941) stated the case strongly, on the basis of calculations of transition probabilities, that triplet states of ordinary organic molecules could not be excited by light.

The Lewis and Kasha papers were received in an atmosphere of great skepticism at best. J. Franck and R. Livingston argued publicly against them, as did E. Teller and R. Rabinowitch. But the Lewis and Kasha work has survived and launched a whole new era of spectroscopic observation including flash spectroscopic studies and EPR studies of the triplet state. Lewis and Kasha proved through their spectroscopic studies of some 90 molecules that the phosphorescence glow of organic molecules corresponded to the lowest triplet state to ground singlet state emission and was an intrinsic property of each molecule, independent of environment, and was even observable in vapor states in appropriate cases; that the vibrational structure, lifetime, and frequency of emission could be correlated with molecular structure and that the Einstein A and B coefficient relationship held sufficiently well to correlate triplet-singlet emission with singlet-singlet absorption. In a posthumous paper of Lewis, Lewis, Calvin, and Kasha determined the magnetic moment of the lowest triplet state of fluorescein dye—a determination cited by Kittel in the first edition of his *Solid State Physics* as the most sensitive paramagnetic susceptibility measurement in the history of physics.

### *Radiationless Transitions and Spin-Orbital Perturbation Studies*

After Lewis's death in 1946, Kasha continued his spectroscopic studies for 3 years in Berkeley. His main objective was to overcome the criticism of the established workers in the field and to extend the work in the direction of quantum

theoretical requirements. This proved to be a very fertile period of new ideas being spawned. The study of radiationless transitions, some 30 years before the current rapid redevelopment of this field, laid down some phenomenological concepts and terminology that are a standard part of the understanding of this field today. His study of the excited-state radiationless process led to the formulation of what are known as Kasha's Rules (the emitting state of a given multiplicity is the lowest state in the manifold of excited states; the nonradiative states are characterized by a diffuse band structure as a result of uncertainty broadening), which facilitate the correlation of experiment with theoretical analysis. Kasha showed that the Teller restriction on singlet-triplet observability was based on radiative arguments and that even though there was a millionfold restriction of transition probability for "spin-forbidden" transitions in C,N,O atom molecules, the radiationless *intersystem crossing* could compete with normal fluorescence and, in one special case ( $n \rightarrow \pi^*$  transitions, see below), triplet-state excitation and subsequent phosphorescence under conditions of observability would actually dominate the luminescence properties, with quantum yields approaching unity. These researches were accompanied by many novel approaches to spin-orbital perturbation of radiationless transitions. Kasha showed that heavy-atom substitution in organic molecules has the theoretically expected Z-effect on intersystem crossing. He also introduced a new effect, the "external heavy atom effect" or *collisional perturbation of spin-orbital coupling*, showing that singlet-triplet transitions of organic molecules could be induced by a heavy-atom-containing solvent. This widely used phenomenon has been named the Kasha Effect.

### *Orbital Classification of Electronic Transitions of N-Heterocyclic Spectroscopy*

In attempting to correlate his research with the wider world of theoretical spectroscopy, Kasha took a clue from Robert Mulliken's writings and introduced the  $n \rightarrow \pi^*$  and  $\pi \rightarrow \pi^*$ , and later the  $l \rightarrow a_\pi$ , classification of organic molecule and inorganic polyatomic molecule transitions, starting in 1950. This work initiated a whole new era in finer understanding of molecular spectroscopy and especially photochemistry. The last 20 years of active redevelopment of organic photochemistry from a kinetic emphasis to an electronic spectroscopic mechanisms emphasis was strongly influenced by Kasha's contributions.

The orbital classification scheme led to the first understanding of hydrogen-bonding effects in molecular spectra (the " $n \rightarrow \pi^*$  Blue Shift"), the theoretical demonstration that spin-orbital matrix elements for  $n \rightarrow \pi^*$  triplets should be greater than that of  $\pi \rightarrow \pi^*$  triplets, and that the unusual quantum yields of phosphorescence and consequent photochemical effects could be understood.

This section would be incomplete without stating that again Kasha's first attempts to introduce these topics into N-heterocyclic chemistry met some opposition by the established workers. Robert Mulliken estimated that the higher ionization potential of the "lone-pair" electrons in N-aromatics would make optical transitions involving these electrons unobservable in such molecules. Kasha

found that such molecules behaved very much like the carbonyl compounds studied theoretically by Mulliken, and so extended the concept of "lone-pair" or "nonbonding" electron excitation ( $n \rightarrow \pi^*$ ) to all heteroaromatic molecules containing functional groups with N,O,S and similar atoms. Published work by H. Sponer and by H. Stücklen already treated nitrogen heterocyclics as having only  $\pi$ -electron transitions, as Mulliken predicted, and new work being developed by Halverson and Hirt also assumed this to be the case. Kasha reversed all these assignments and proved that even in cases like pyridine the  $n \rightarrow \pi^*$  transitions lie lowest and predominate.

### *Molecular Exciton Effects and Excitation Mechanism*

Starting in 1959, Kasha and his students published an extensive series of papers on molecular exciton theory applied to molecular aggregates. Kasha had published English translations of A. S. Davydov's now famous first paper on naphthalene crystal spectra (1948) and later his book *Theory of Molecular Excitons* (with Max Oppenheimer, Jr., 1962), which was largely in the realm of crystal physics. Kasha's interest in molecular exciton application to small molecular aggregates was provided by Albert Szent-Gyorgyi, who presented the paradox that dye molecules frozen in alcohol exhibited fluorescence, whereas the same molecules frozen in water (ice) exhibited only phosphorescence—seemingly in conflict with all known quantum theories of spin-orbital effects. McRae and Kasha showed that molecular exciton effects in the singlet manifold of dye aggregates could explain the phenomenon as a non-spin-orbital enhancement effect of triplet-state excitation (parallel work by William T. Simpson appeared simultaneously). They thus generalized the effect to explain several other related phenomena of photosensitization effects noted in the literature, especially in the experiments of H. Kautsky.

This work has been greatly extended in recent years to include hydrogen-bonded dimers, simple polymers, and lamellar molecular arrays. Extensive applications to biophysical problems have been made. Nevertheless, a great gap remains to be bridged between the experimental biologist and biochemist, working with elegant techniques but rudimentary molecular concepts, and the real understanding of sophisticated concepts of molecular quantum theory, when applied to molecular aggregates.

### *Vibrational Deficiency Theorem for Small Polyatomics*

Starting in 1964, the Group Theoretical study of electronic-vibrational perturbation in small molecules led Kasha and his students to the development of a new Group Theoretical theorem: A molecule may be considered to be *vibrationally* deficient if its normal modes of motion fail to span the irreducible representations of the Point Group to which it belongs. It was shown that a whole new class of environmentally sensitive electronic transitions may exist in small symmetrical molecules (including all diatomics) as a consequence of this theorem, explaining some unusual intensity and solvation anomalies previously neglected.

### *Singlet Molecular Oxygen*

In 1963 Khan and Kasha opened up a new field of oxygen chemistry by their discovery of the *chemical* production of (excited) singlet molecular oxygen from hydrogen peroxide and hypochlorite in aqueous media. Since 1924, singlet molecular oxygen was known largely as a rare species observed in atmospheric or stratospheric spectroscopy, with zero, one, or two publications appearing per year by astrophysicists. Since 1963, more than 2000 research papers on singlet oxygen have appeared, covering every aspect of physics, inorganic and organic chemistry, biochemistry and biology, photochemistry, radiation chemistry, and technology. Five international conferences have been held on this subject, and several special research volumes have been published.

The dramatic development of this field came from the sudden chemical availability of a new and highly reactive species of molecular oxygen, and a fuller understanding of its ubiquitous role in phenomena that had become ossified in old concepts. For example, photooxidation of organic molecules in the presence of dyes (as the light absorber) and ordinary (triplet) molecular oxygen was long accepted as involving a dye-peroxy complex. Now it was unambiguously demonstrated (C. Foote) that the molecular oxygen was excited by energy transfer from the dye to the highly metastable singlet molecular oxygen state and that this latter form was the reactive species. This cast an entirely new view on a whole series of "oxygen effects" in chemistry and biology. This field is developing currently at an even more rapid pace and has reached into mechanisms of certain human diseases and laser cancer treatment (T. Dougherty) and, most recently, into peroxidase enzyme mechanisms (A. U. Khan).

### *Spectroscopic Theory of the Solvent Cage*

In 1975 Dellinger and Kasha applied the Born–Oppenheimer approximation to the old problem of the kinetic solvent cage. They demonstrated the feasibility of resolving the vibrational motion of molecules within a solvent cage, from the motion of molecules making up the solvent cage. The method applies a potential curve perturbation for the solute molecule, not at the equilibrium coordinate  $Q_e$ , as is usually done, but at a new position  $Q_0$ , where the derivative of the potential with respect to coordinate approaches 0, that is, near the top of a barrier or near a dissociation limit. This gives a new tool for the spectroscopic studies of molecules in a solvent cage, and the schematic theory published has been applied to several different molecular potentials, representing different kinds of molecular motion. Results are already beginning to appear in the literature confirming some of the novel predictions made. Dellinger and Kasha have themselves completed research on hydrogen-bonded molecular complexes showing the action of the solvent-cage viscous-flow barrier to photodissociation of the complexes. New phenomena are thereby predicted.

### *Excited-State Proton Transfer Spectroscopy*

One of the main currents of new development in Kasha's laboratory lies in studying the electronic consequences of excited-state proton transfer. One of the

most exciting discoveries in Kasha's laboratory was the observation in 1969 of bi-protonic phototautomerism in H-bonded base pairs (7-azaindole). This work has led to a series of research in other laboratories confirming and enhancing the understanding of the cooperativity of the simultaneous transfer of two protons in excited states of base pairs and on the nature of the double-minimum potential and its kinetic aspects.

Recently, this work has been greatly extended to the observation of five distinct types of intramolecular proton transfer, in addition to the well-known previous cases of intermolecular proton transfer (to solvent). In the new cases, e.g., 3-hydroxyflavone, intramolecular proton transfer has been demonstrated (1984) to be strongly modified or prevented by external H-bonding. Several new studies show catalytic promotion of intramolecular proton transfer, including a proton-relay mechanism involving one, two, or three H-bonded water molecules as bridges. Taken together, these new proton-transfer researches offer a greatly expanded range of observable spectroscopic phenomena for the study of the normal and tautomeric forms of a molecule and their dynamic relations. Applications include the mechanisms of proton-transport in membranes and a new highly efficient *proton-transfer laser* published by Kasha and his colleagues in recent years.

### *Summary and Overview*

Kasha's work can be characterized as discovery of new molecular phenomena by the most detailed study of the interplay of quantum theory and spectroscopic experimentation.

One of Michael Kasha's greatest contributions to science is the long series of inspired scientists he has educated in his 39 years at the Florida State University. The penetrating analysis, search for novelty, and enthusiasm generated in his laboratory have been amply transmitted to his large group of productive students and associates now distributed in key positions in universities and in industrial and government laboratories in this country and abroad.

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