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N. Feautrier
Chairman of Working Group

WORKING GROUP 5: MOLECULAR STRUCTURE AND TRANSITION DATA

Research in molecular spectroscopy has continued to grow over the past three years. The spectral range has expanded from the far ultraviolet to millimeter wavelengths. The report has been limited to molecular spectroscopy of relevance to astronomy and has been compiled from edited contributions sent to me in the fall of 1987.

Sumner P. Davis and John G. Phillips have reported studies at the Berkeley laboratory on the molecules C₂, CN, FeH, InI, SH, Si₂, SiC₂, TiCl₂, TiO, ZrO, ZrS of either analysis of spectral structure or measurements of lifetimes and oscillator strengths. FeH is of special interest. A complete table of excitation energies for its complex infrared system has been prepared, and a set of tables for far infrared and radio wavelengths assembled to predict spectrum lines which may exist in stellar spectra or in the spectrum of matter in interstellar space. Underway are analysis of the blue-green system of the FeH molecule, measurements of radiative lifetimes for FeH, radiative lifetimes of CaH, transition strength for A-X and B-X systems of CaH, measurement of CaCl transition strength (collaboration with J.E. Littleton), analysis and tabulation of infrared OH and OD bands (collaboration with R. Engleman) and analysis of ZrS in the infrared (collaboration with R. Winkel).

The bimonthly Berkeley Newsletter on Analyses of Molecular Spectra compiled by Davis, Phillips and Eakins continues to be the invaluable, timely bibliography of molecular spectra. There is about 150 recipients of the Newsletter.

Takeshi Oka has reported from the University of Chicago, laboratory spectra observed of the following molecular ions, H₃⁺, H₂D⁺, HeH⁺, NeH⁺, ArH₃⁺, NH₄⁺, NH₃⁺, NH₂⁺, H₃O⁺, H₂O⁺, OH⁺, HCNH⁺, CH₃⁺, HCCH⁺, C₂H₃⁺, OH⁻, C₂⁻. A search for interstellar infrared absorption spectrum of H₃⁺ was conducted; the result is inconclusive though promising.

Kurt Dressler has reported from ETH Zurich work on the electronic transition

moments for the Lyman and Werner bands of $H_2(1)$. Stephens and Dalgarno's (2) calculated radiation lifetimes of the B & C states are confirmed and its suggested that recent measurements of longer lifetimes may be affected by radiation trapping.

The Molecular Spectroscopy Division of the National Bureau of Standards, Gaithersburg, MD, carries out experimental studies in the microwave, infrared and visible regions and develops critical reviews on microwave spectra of interstellar molecules and tables of infrared absorption lines for calibration of diode laser measurements. F.J. Lovas has reported that during the past three years several critical evaluations of microwave rotational spectra have been published. The twenty-second article in the series "Microwave Spectra of Molecules of Astrophysical Interest" treats the spectrum of SO_2 (3). The 1985 Revision to "Recommended Rest Frequencies for Observed Interstellar Molecular Microwave Transitions" was published in 1986 (4). Two compilations treating vibrational spectral and properties have also been published. Pine *et al.* (5) report on the high temperature water vapor spectrum in the 3000 4000 cm^{-1} region and Jacox (6) has compiled the ground state vibrational energy levels for polyatomic transient molecules.

The properties and spectra of diatomic species have been examined both theoretically and experimentally. Theoretical electronic structure calculations on FeO RuO (7). AgH and AuH (8), and transition moments for excited states of NaK (9) have been reported. Jacox (10) reports on comparisons of vibrational fundamentals of diatomic molecules in the gas phase and in inert solid matrices. Experimental studies on diatomic species have treated collisional effects on the rovibrational transitions of HD (11) and infrared spectra of high temperature species LiI (12). GeO (13) LiCl (14) and PbO (15).

The effort to develop tables of accurate infrared transition frequencies continues with heterodyne measurements on CO_2 (16), N_2O (17) and OCS (18) by Wells, Maki and co-workers. Also high resolution infrared studies of the hydrocarbons, ethane (19) and allene (20) have been reported.

A.R.W. McKellar has reported from the Herzberg Institute of Astrophysics, NRC, Canada that spectroscopic studies of molecules of astronomical interest have continued with an emphasis on unstable species in general and molecular ions in particular. Electronic spectra have been studied for H_2 (21-23), He_2 (24), NO (25) XeH (26), $ArXe^+$ (27) and NF (28) with special attention to O_2 (29-33), S_2 (34), and SeS (35). Electron impact ionization studies have been made for the hydrogen halide systems HCl DCI (36) and HBr DBr (37). Infrared vibration-rotation and pure rotational studies have been made for H_2 (38), HD (39-42) SiN (43), FO (44), and HF (45).

Considerable efforts have been applied to the study of polyatomic molecular ions in the infrared, including DCO^+ (46), N_2O^+ (47-49), HOC^+ (50-52), $HOCO^+$ (53 54), D_3O^+ (55), $HCNH^+$ (56), NH_3D^+ (57), SH_3^+ (58), and especially H_3^+ and its isotopes (59-67). Other polyatomic molecules studied in the infrared include H_2S (68,69), DO_2 (70), OCS (71), H_2O (72,73), CO_2 (74), CH_2 (75,76), ND_3 (77,78), C_3O_2 (79), D_2CO (80), and cyclopropane (81-83). Theoretical studies have emphasized a combination of high accuracy *ab initio* force fields and sophisticated vibration-rotation calculations to predict molecular energies (76,84-89).

The technique of microwave-optical double resonance has been extensively utilized to study the molecules HNO (90,91), and H_2CS (92-95). Other polyatomic molecules studied by electronic spectroscopy include ND_2 (96), CF_2 (97), CH_2 (98), C_2H_2 (99,100), and NH_3 (101). The electron impact ionization technique has been applied also to He (102) C_2H_2 (103), and C_2H_4 (104).

R.W. Nicholls has reported from the Center for Research in Experimental Space

Science at York University, Toronto, that work has continued on experimental and theoretical aspects of astrophysically important spectra. Shock tube studies on NbO and LaO have recently been initiated. Much effort has been devoted to the realistic numerical high resolution synthesis of molecular spectra, to reduce line shapes (105), and also for diagnostic application to laboratory (106), atmospheric and astrophysical circumstances including the interstellar extinction function (107). Non-dispersive digital correlation spectrometers have been developed for diagnoses of atmospheric conditions. The behaviour of dispersive correlation spectrometers has been modelled (108). An extensive study of simple analytical representations for molecular Franck-Condon factors for astrophysical band systems whose constants are incompletely known has been extended to include bound-free and free-free transitions. An extended Birge-Mecke rule linking the vibrational and rotational constants of molecular states, which fits extensive experimental data has recently been derived (109).

Research in molecular spectroscopy at the Harvard-Smithsonian Center for Astrophysics Cambridge MA is directed towards processes of importance to atmospheric physics and astrophysics. The wavelength range studied extends from the ultraviolet to the millimeter-wave spectroscopy. W.H. Parkinson has reported that K. Yoshino and colleagues have concentrated on a number of high resolution spectroscopic studies with the 6.65 m vacuum scanning spectrometer/spectrograph. Ab initio studies of potential energy curves and dipole moments for important small molecules of astrophysical interest have been undertaken by K. Kirby and colleagues.

Absorption cross sections and band oscillator strengths of the Schumann-Runge (S-R) bands of $^{16}\text{O}_2$, $^{18}\text{O}_2$ and $^{16}\text{O}^{18}\text{O}$ at 79 K have been measured between 179.3 and 198.0 nm with an instrumental full width at half maximum (FWHM) of 0.0013 nm. Oscillator strengths of the bands have been obtained by direct integration of the measured cross sections (110).

Spectroscopic constants of the $\text{B } ^3\Sigma_u^-$ state of $^{16}\text{O}_2$ (111), $^{18}\text{O}_2$ and $^{16}\text{O}^{18}\text{O}$ have been determined from the experimental data on the high resolution absorption spectrum of the S-R bands. These constants will be used in the ongoing effort to extract the predissociation linewidths of the S-R bands from the absolute cross section measurements. Level shifts in the $\text{B } ^3\Sigma_u^-$ state of these isotopes due to various repulsive continuous states are being calculated. Parameters of these states are being determined and will be used to examine the vibrational and rotational dependence of the resonance linewidths.

The absorption cross section of O_2 has been measured between 195 and 241 nm at 300 K. This region contains the Herzberg I transition of O_2 and of the O_2 dimer. Our Herzberg continuum cross sections are significantly smaller than those previously used in many photochemical stratospheric modelling calculations (112).

Absolute cross sections of O_3 at the temperature of 195 K, 228 K and 293 K have been measured at several discrete wavelengths in the region 238-335 nm (113).

A high resolution vuv absorption study and vibrational analysis of the $\text{B } ^1\Sigma^+ - \text{X } ^1\Sigma^+$ system of CS was performed in the wavelength region 128-155 nm (114).

High resolution spectra and photoabsorption coefficients for CO absorption bands in the 91-112 nm region have been measured at the Photon Factory of the National Laboratory for High Energy Physics (KEK), Japan.

Potential energy curves, electronic wavefunctions and electric dipole moments for the $\text{X}^3\Sigma^-$, $\text{A}^3\Pi$, $2^3\Sigma^-$, $2^3\Pi$ and $1^5\Sigma^-$ states of NH (116). The $2^3\Sigma^-$ and $2^3\Pi$ states are repulsive and have been identified as important photodissociation pathways.

Spectroscopic constants have been obtained for the bound states. The $1^5\Sigma^-$ is shown to cross the $A^3\Pi$ at a substantially higher energy and larger internuclear separation than previously postulated and therefore is probably not the cause of observed predissociations in low-lying vibrational levels of the $A^3\Pi$.

Configuration interaction calculations have been carried out on the low-lying $1^1\Delta$ and $1^1\Sigma^-$ states of CO. The lifetimes of the vibrational levels of the $1^1\Sigma^-$ and $D^1\Delta$ have been computed using transition moments for $1^1\Sigma^- - A^1\Pi$ and $D^1\Delta - A^1\Pi$.

By using a specially-designed millimeter-wave absorption spectrometer, B.J. Connor and H.E. Radford have made laboratory measurements of pressure-broadening coefficients for several spectral lines of ozone (117) and carbon monoxide (118) at wavelengths near 3 mm. The results are useful for the analyses of ground-based observations of the same lines emitted by the earth's atmosphere.

In the period 1985-88 Patrick Thaddeus and Carl Gottlieb have completed the first laboratory millimeter-wave spectroscopy of the carbon chain radicals C_3H (119, 120), C_5H (121), and C_6H (122); the CCD radical (123); the first hydrocarbon ring identified in interstellar space, C_3H_2 (124, 125); the stable three-membered ring cyclopropene, C_3H_2 rotational spectrum led to the identification of numerous astronomical transitions. including the strong ubiquitous lines at 18, 343 MHz and 85, 338 MHz. Following the measurement of its laboratory spectrum, the CCD radical was detected in Orion A with a deuterium enhancement comparable to the strongest enhancement yet found in any molecule. Measurement of the millimeter-wave spectrum of cyclopropene, obtained by attaching two H atoms to the related ring C_3H_2 , allowed them to place upper limits on its abundance in Sgr B2 and TMC-1 indicating that in these sources C_3H_4 isn't much more abundant than C_3H_2 . Laboratory measurements of vibrationally excited C_4H confirmed a tentative astronomical identification. The surprisingly large intensity of the vibrationally excited lines of C_4H relative to the ground state in IRC+10216 suggests the opening of a whole new subdiscipline in which laboratory spectroscopy will be crucial, since up to now little or no spectroscopy has been done on the excited state of small reactive chains and rings.

Research programs at the Observatoire de Paris (Meudon) concern new spectroscopy, photodynamics and transition probabilities of astrophysically interesting molecules. Francois Rostas has reported that a comprehensive effort has been applied to a better understanding of the CO molecule. A new analysis of the $A^1\Pi(v=0)$ perturbations extending to $J = 75$ has been completed (129) using absorption and emission spectra of the A-X transition. The analysis of the $0 < v \leq 4$ levels will be published shortly. The B-X transition has been reanalyzed (130) and a study of its predissociation is in progress. New data concerning the $C^1\Sigma^+$ and $E^1\Pi$ Rydberg states have been analyzed. The photodissociation cross section has been measured between 88.5 and 115 nm (131). It is shown that photodissociation occurs in discrete bands rather than in a continuum with far reaching astrophysical consequences.

The establishment of an atlas of H_2 emission lines between 78 and 168 nm is progressing steadily. This work has suggested calculation of line positions and intensities of the Lyman and Werner systems taking into account rotational coupling between the B and C states (132). These results are in agreement with observations and quantitative intensity measurements and allow a more complete analysis of the spectra.

A new emission band of the N_2^{++} ion has been observed and compared to ab initio calculations (133, 134).

Emission spectra of N_2 have been obtained at very low pressure in the 85 to 108 nm region. Intensity anomalies of previous absorption spectra have been shown

to be due to reabsorption effects (135).

High resolution spectra of CO₂ between 10 and 14 eV have been analyzed. nf transition bands have been characterized by sharp features in the rotational band contours. Calculated quantum defects have been used to assist in the assignments (136).

An extensive theoretical study of C₂ has been pursued in conjunction with a model of the ζ Oph. diffuse molecular cloud. Energy levels, rotational excitation by collisions with H₂ and radiative equilibrium including intercombination transitions have been determined (137-138) and included in a comprehensive chemical and radiative model of the cloud.

A new emphasis is placed on the photophysics spectroscopy and relaxation of molecular ions (139). Doubly charged ions of polycyclic aromatic molecules are studied in detail (140).

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W.H. Parkinson, Chairman of Working Group