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Resonance Enhanced Multiphoton Ionization Spectra of Molecules and Molecular Fragments

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● Background and Objectives.

Resonance Enhanced Multiphoton Ionization (REMPI) utilizes pulsed laser radiation to prepare a molecule in an excited state via absorption of one or more photons and *to subsequently ionize that state* before it can decay. A remarkable feature of REMPI, and one that is very basic to many of its applications and uses, is that the very narrow bandwidth of the "pump" laser makes it possible to select a specific vibrational and rotational level in the initial state and to prepare the excited state of interest in a single vibrational and rotational level. Thus, by suitable choice of the photon pump transition, it is possible to selectively ionize a species of interest without ionizing any other species that might be present. This feature makes REMPI one of the most *powerful tools for ultrasensitive detection of species.* With REMPI it is also possible to study the photoionization dynamics of a *single rotational level of an excited electronic state.* Such state-resolved studies can certainly be expected to provide significant insight into the underlying dynamics of molecular photoionization. Some significant applications of REMPI, which are relevant to our choice of objectives, include:

(*i*) ultrasensitive and state-specific detection of unstable, reactive, or trace species

(*ii*) production of molecular ions with specific vibrational and rotational distributions for use in studies of ion-molecule reactions

(*iii*) probing the intramolecular dynamics of highly excited states, photofragmentation, and gas-surface scattering,

(*iv*) exploring excited state chemistry and physics at a quantum-state specific level.

The overall objective of this effort is to carry out theoretical studies of these REMPI processes in molecules and molecular fragments which are designed to provide a robust analysis and prediction of key spectral features of interest in several experimental studies and applications of this technique. A specific and very important objective of our effort is to predict the vibrational and rotational ion distributions which result from REMPI of representative molecules and to understand the underlying mechanisms that give use to these ion distributions.

- For example, a key question is: when can we expect to see anomalous vibrational distributions of ions in REMPI of molecules via a resonant Rydberg state? Photoionization of such Rydberg states is generally expected to lead to Franck-Condon ion distributions and to occur with preservation of vibrational quantum number, i.e. $\Delta v = v^+ - v' = 0$. Anomalous behavior in these ion distributions introduces serious complications into the use of REMPI for determination of state populations from ion signals and production of ions in specific states. We will see below that, in addition to shape and autoionizing resonances[1,2] which are final-state mechanisms responsible for anomalous ion vibrational distributions, we have shown that rapid orbital evolution of the resonant Rydberg state, i.e. changing angular momentum character with internuclear distance, is an important mechanism leading to such anomalous ion distributions. Furthermore, it will also be seen that *Cooper minima*, which can often be, but not necessarily, associated with such orbital evolution, *have a profound and far-reaching influence on rotational ion distributions resulting from REMPI.*

- Other important objectives of our work are to study rotational ion distributions resulting from REMPI of molecules, to identify the underlying and important mechanisms responsible for these distributions, and to exploit this insight to tune and influence rotational ion distributions. To put the significance of this objective into perspective, *it is important to recognize that, while a specific rotational level of a resonant state is photoionized in REMPI, the resulting rotational distributions of ions is strongly controlled by the angular momentum coupling present in the molecular photoelectron orbitals.* In contrast to atomic photoelectron orbitals, molecular photoelectron orbitals are not angular momentum eigenfunctions, i.e. they are not s, p, d, or f functions, but are admixtures of angular momentum functions. This mixing of angular momentum in molecular photoelectrons is brought about the torques associated with the nonspherical potentials, $V(r, \theta, \phi)$, of molecular ions. One may think of this as though the photoelectron "arrives" in the photoionization continuum with a specific angular momentum ℓ which is quickly mixed with other ℓ's as the photoelectron is rescattered or "knocked around" by the molecular ion potential. *For a very simple picture, we could think of rotational ion distributions as reflecting how angular momentum is being shared between the photoelectron and the molecular ion.* This certainly suggests that rotational ion distributions will reveal rich dynamical structure and often show features which cannot be accounted for on the basis of an atomic-like model of photoionization, even if photoionization occurs out of a molecular Rydberg orbital of *almost pure s or p or d character.*

A significant development here has been our identification of the significant role that Cooper minima[3] (or Cooper zeros) exert on rotational ion distributions.[4-8] A Cooper minimum occurs at a photoelectron kinetic energy where a specific angular momentum component of the photoionization matrix element goes through a zero. When this happens, *that angular momentum is no longer operative in the photoionization process* and hence rotational levels in the ion spectrum which are "driven" by this angular momentum component of the photoelectron can be strongly influenced. In fact, we have seen a very significant example of this behavior recently in a joint theoretical-experimental study of rotational ion distributions in REMPI of OH where a Cooper minimum leads to primarily a ΔN = even distribution of ionic rotational states, in contrast to the expected ΔN = odd distribution.[7] Here ΔN is the change in angular momentum, exclusive of spin, between the state being ionized and the ion. Furthermore, we have also demonstrated that Cooper minima can be exploited to achieve a high degree of rotational selectivity in molecular ion distributions produced via REMPI[4] and that circular dichroism in the photoelectron angular distributions[8] (CDAD) can be used to locate and identify Cooper minima in REMPI spectra.

● Research Accomplishments: Progress and Highlights

I will now highlight the progress we have made in our studies of the features and applications of REMPI of molecules which we have outlined above. A significant feature of these studies is that they were carried out using molecular photoelectron orbitals which incorporate the angular momentum coupling present in such orbitals. This angular momentum coupling, which results from the torques associated with nonspherical molecular ion potentials, exerts a strong influence on the dynamics of the photoionization processes of interest here. In these studies we take these orbitals to be solutions of a one-electron Schrödinger equation containing the Hartree-Fock potential associated with the charge density produced by the molecular orbitals of the ion. These photoelectron orbitals are obtained by numerical solution of the associated Lippmann-Schwinger equation.[9]

- *The influence of Cooper minima on rotational ion distributions*: In a joint theoretical and experimental study we have demonstrated, for the first time, the dramatic influence of a Cooper minimum on the rotational ion distributions resulting from REMPI of a resonantly prepared Rydberg state of a molecule.[7] The D $^2\Sigma^-(3p\sigma)$ Rydberg state was prepared by two-photon absorption from the X $^2\Pi$ ground state of OH produced by photolysis of H_2O_2. Fig. 1a shows the rotational ion distribution resulting from photoionization of the $N' = 9$ rotational level of this $3p\sigma$ Rydberg state. For ionization of such a Rydberg level the ion rotational distribution (N^+) is

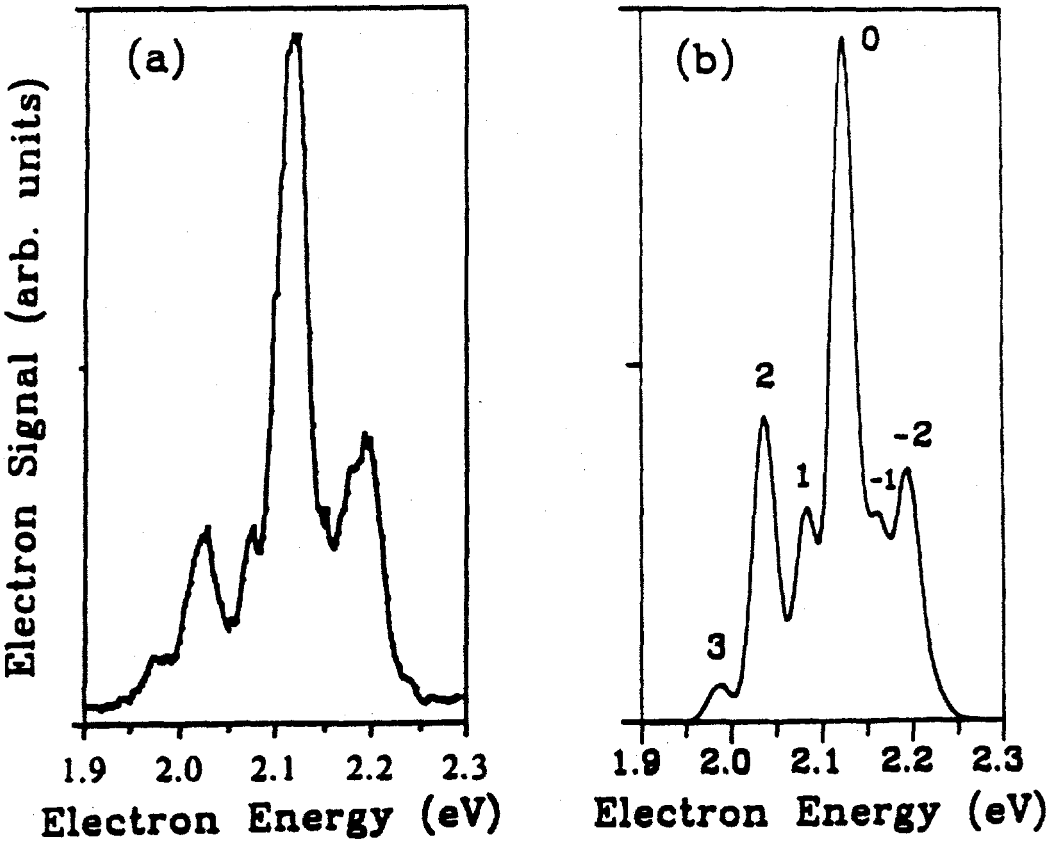

Fig. 1. Experimental and calculated rotationally resolved photoelectron spectra for (2+1) REMPI of OH: (a) measured spectra for the $N' = 9$ rotational level of the D $^2\Sigma^-(3p\sigma)$ Rydberg resonant state; (b) calculated photoelectron spectra.

governed by the selection rule $\Delta N + \ell = \text{odd}$, where $\Delta N = N^+ - N'$ is the change of rotational quantum number and ℓ is an angular momentum component of the photoelectron orbital. We observe strong $\Delta N = \text{even}$ signals in these spectra, in strong contrast to the $\Delta N = \text{odd}$ distribution expected for ionization of a $3p\sigma$ Rydberg orbital in an atomic-like picture, i.e. $3p\sigma \rightarrow ks$ and kd ($\ell = 0, 2$).

Fig. 1b shows our calculated rotational ion distributions.[7] The calculated spectra do, in fact, show a dominant $\Delta N = \text{even}$ distribution. Analysis of these results shows that this rotational distribution arises from a Cooper minimum in the $3p\sigma \rightarrow k\pi$ ($\ell = 2$) ionization channel. Since the $\ell = 2$ component of the photoelectron continuum becomes depleted upon formation of the Cooper minimum, the odd photoelectron waves from the $3p\sigma \rightarrow k\sigma$ channel begin to dominate. In fact, the strong $\Delta N = 0$ peak seen in the spectra is associated mainly with the $\ell = 1$ (p wave) component of the photoelectron orbital. The agreement between the measured and calculated spectra is encouraging and serves to highlight the implications of Cooper minima for rotational ion distributions resulting from REMPI.

We have also shown that the intense and unexpected $\Delta N = 0$ peak, observed in early and seminal measurements[11] of the rotational ion distributions resulting from (2 + 1) REMPI of NO via the D $^2\Sigma^+(3p\sigma)$ state, primarily arises from a Cooper minimum around 0.33 eV in the $3p\sigma \rightarrow k\sigma$ ($\ell = 2$) photoionization channel.[12] According to the selection rule $\Delta N + \ell = \text{odd}$, this *unusual rotational peak should derive predominantly from odd angular momentum components* (ℓ), *particularly* $\ell = 1$ (p-wave), *of the photoelectron.* Calculations show that the $3p\sigma$ Rydberg orbital of the D $^2\Sigma^+(3p\sigma)$ state is about 99% $\ell = 1$. An atomic-like propensity rule, which would predict $\ell = 0$ and $\ell = 2$ character for the photoelectron, i.e. $\ell = \ell_0 \pm 1$, where ℓ_0 is an angular momentum component of the Rydberg orbital, thus seems wholly inadequate. An analysis[12] shows that it is the depression of the $\ell = 2$ wave brought on by a Cooper minimum around 0.33 eV and the subsequent enhancement of the relative importance of the $\ell = \text{odd}$ waves that results in the unusual $\Delta N = 0$ peak in the ion rotational distribution and the dependence of these distributions on photoelectron energy.

- *Locating and identifying Cooper minima in resonance enhanced multiphoton ionization spectra*: In view of the important role that Cooper minima can be expected to play in REMPI rotational ion spectra, it is clearly desirable to develop a convenient experimental strategy for locating and identifying Cooper minima in photoionization of excited molecular Rydberg states. Due to their weak influence on total cross sections – particularly with the larger number of partial waves (angular momentum components) contributing to photoionization of molecules compared to atoms – Cooper minima have traditionally been detected in photoelectron angular distributions in photoionization of ground electronic states. A Cooper minimum has yet to be observed in ionization of an excited molecular Rydberg state. In REMPI, however, such an excited Rydberg state would be aligned due to the absorption of linearly polarized light in the multiphoton excitation step of REMPI. We have previously shown that the angular distributions of photoelectrons resulting from photoionization of such aligned linear molecules – with their unequal populations of M_J sublevels of a rotational level J – exhibit a dichroic asymmetry, i.e. angular distributions from aligned states are different for photoionization with left or right circularly polarized light. Furthermore, we have also shown that this effect (CDAD) – circular dichroism in photoelectron angular distributions – arises in the electric dipole approximation[14] and can be readily measured.[8]

To exploit this CDAD behavior to probe Cooper minima, we proceed as follows.[6] First, we note that CDAD arises solely from interference between different channels in the photoionization continuum. We can see this clearly in the example of photoionization of an aligned atomic 3p orbital. In this case photoionization occurs into the s($\ell = 0$) and d($\ell = 2$) continua and the CDAD signal is due to interference between these s and d continua. The magnitude of this signal at an angle θ is determined by the quantity β where

$$\beta = \langle 3p\,|r|\,ks\rangle \cdot \quad \langle 3p\,|r|\,kd\rangle \cdot \quad \sin(\delta_s - \delta_d)$$

where $\langle 3p\,|r|\,k\ell\rangle$ is the radial matrix element for photoionization of a 3p orbital into the ℓ continuum and δ_ℓ is its phase shift. *This shows that CDAD will change sign whenever one of the radial matrix elements goes through zero and flips sign, i.e. a Cooper minimum!* To illustrate this behavior, Fig. 2 shows the calculated CDAD signal for (1 + 1') REMPI via a R branch of the D $^2\Sigma^+$ (3pσ) state of NO as a function of photoelectron energy at 45°. The zero seen in the CDAD signal at 3.2 eV results from a Cooper minimum in the $\ell = 2$ angular momentum component of the photoelectron continuum. We expect this technique to be generally applicable for detection of molecular Cooper minima associated with REMPI.

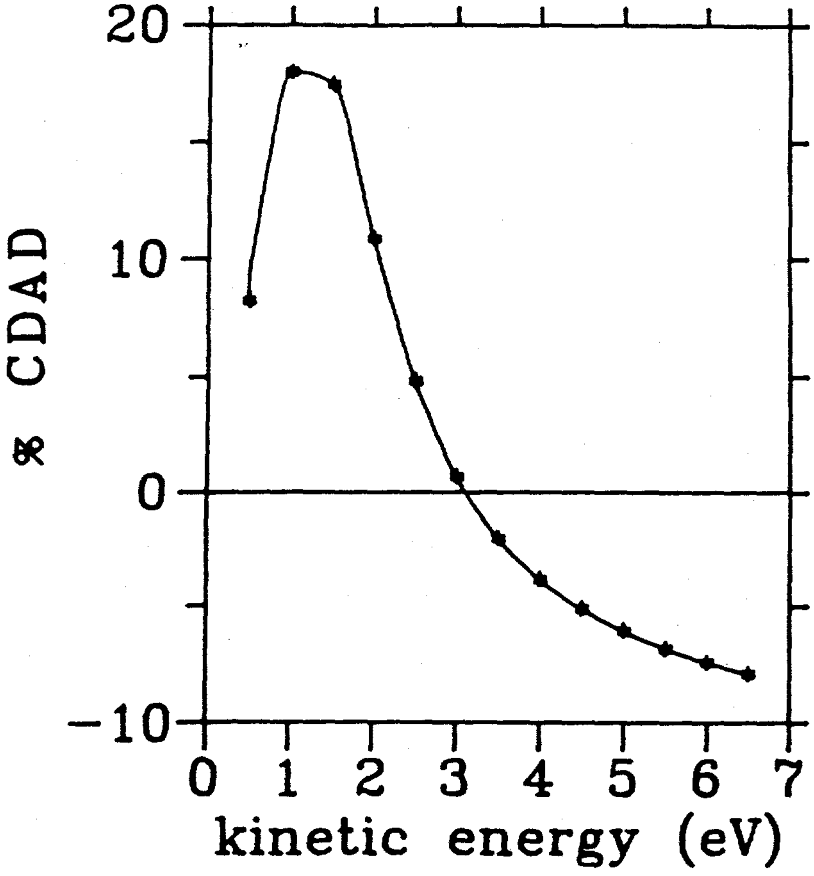

Fig. 2 Illustration of the use of circular dichroism in photoelectron angular distributions (CDAD) to locate Cooper minima in molecular photoelectron spectra. This example is for resonance enhanced multiphoton ionization of the D $^2\Sigma^+$ (3pσ) state of NO.

- *Rotational ion distributions resulting from REMPI of HBr*: Recently Xie and Zare[15] have used laser induced fluorescence (LIF) to determine the rotational distribution of HBr^+ ions resulting from (2 + 1) REMPI via the S(2) branch of the F $^1\Delta_2$ (npπ) state. These LIF studies provide higher resolution than energy-resolved photoelectron studies and made it possible to monitor the populations of individual Λ components of rotational levels of the ion. Since these Λ-doublets result from a splitting of rotational levels due to the interaction between the rotation of the nuclei and the electronic angular momentum, such highly resolved spectra can be expected to provide significant insight into the underlying dynamics of these processes. These results show a strongly parity - favored ion distribution with about 80% of the ions in the (–) component of the Λ-doublet of the J^+ rotational level and the remaining 20% of the population in the (+) component. Furthermore, on the basis of parity selection rules it can be seen that the population of the (–) component results from even angular momentum components of the photoelectron matrix element, while the (+)

population arises from odd angular momentum components. Fig. 3 compares our calculated rotational ion distribution[16] with the measured ion spectra of Xie and Zare.[15] The agreement is encouraging. It is worth stressing here that the odd angular momentum components of the photoelectron matrix element, which are responsible for the population in the (+) parity component, arise from angular momentum mixing in the continuum, i.e. it is intrinsically non-atomic-like.

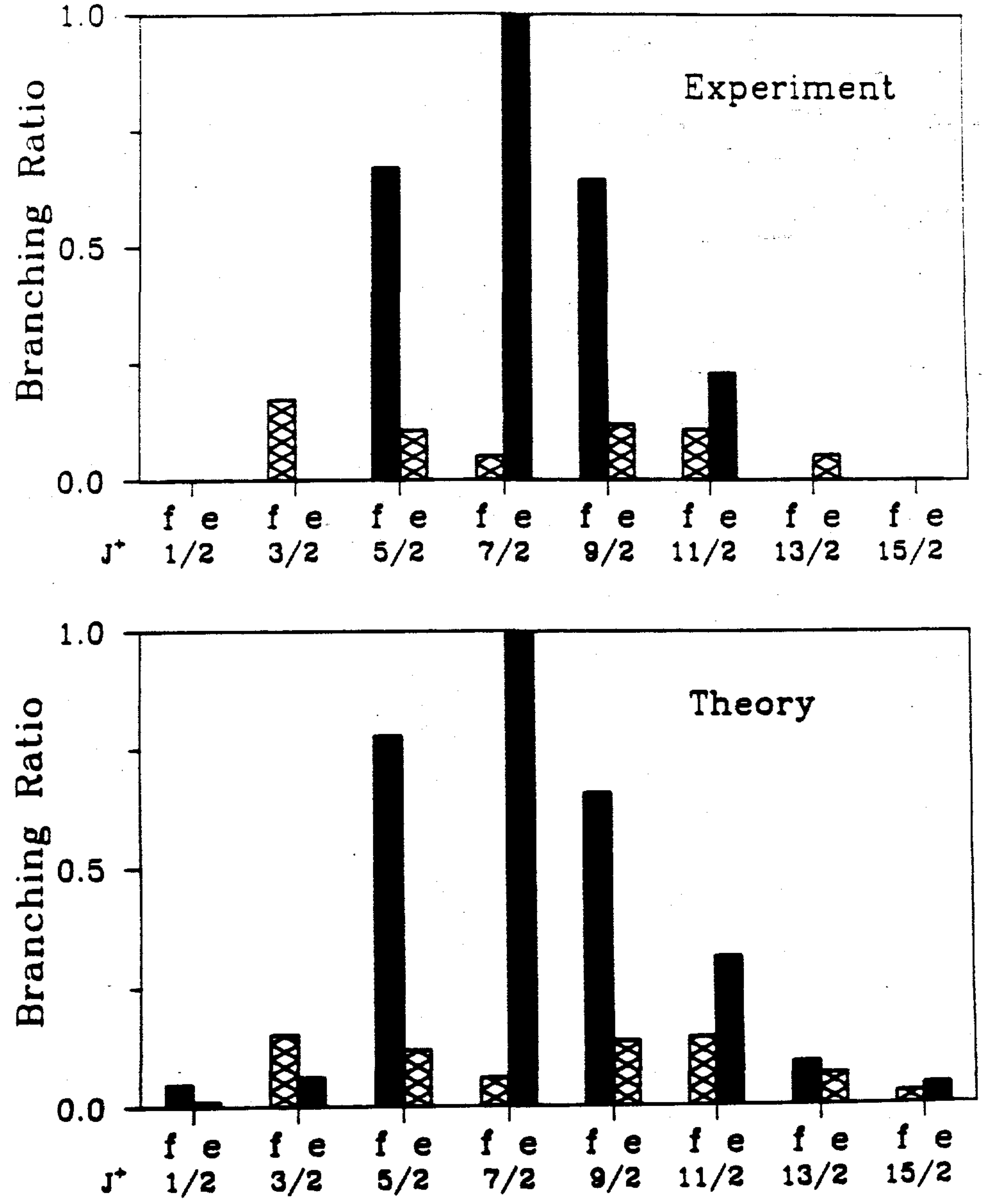

Fig. 3. Measured (ref. 15) and calculated (ref. 16) rotational distributions for HBr$^+$ (X $^2\Pi_{\frac{1}{2}}$) resulting from (2 + 1) REMPI of HBr via the S(2) branch of the $F^1\Delta_2$ Rydberg state. Solid and cross-hatched bars indicate the (–) and (+) parity components of the Λ-doublet of a J^+ rotational level, respectively.

- *Rotational ion distributions resulting from photoionization of H_2O by coherent vacuum ultraviolet (VUV) radiation*: A novel approach to very-high-resolution photoelectron spectroscopy, which is based on the detection of electrons with zero kinetic energy (ZEKE), has recently been developed.[17] ZEKE photoelectron detection achieves a resolution of the order of laser bandwidths and hence provides very high resolution in molecular ion detection. ZEKE photoelectron detection clearly opens up completely new opportunities for studies of rotational ion spectra which would not be possible otherwise.

Recently M.G. White and collaborations[18] at Brookhaven National Laboratory have used a slight modification of this ZEKE technique - pulsed field ionization (PFI) of near-threshold Rydberg states – to measure the rotational distribution of H_2O^+ ions resulting from photoionization of H_2O by coherent VUV radiation. Briefly water is expanded in a supersonic jet where it is cooled to about 15°K. Tunable coherent VUV radiation is used to excite these H_2O molecules via absorption of a single photon to very high Rydberg states which, after a time delay, are ionized by a weak electric field. Such field ionization of Rydberg levels just below threshold should provide a measure of the direct near-threshold photoionization cross sections.[19]

Fig. 4 shows the measured rotational distributions of H_2O ions obtained in these experiments[18] along with the results of our calculations of these rotationally resolved spectra.[20] Assignments are shown above the rotational ion peaks in the calculated spectra. There are several features worth noting about these spectra. First, that such high-resolution rotational ion spectra (note the energy scale!) can be obtained from photoelectron spectroscopy clearly shows the enormous potential that ZEKE electron detection offers for future studies of molecular photoionization at a highly state-selective level. Secondly, the agreement between the measured and calculated ion spectra is encouraging. In these calculations the photoionization matrix elements were obtained using photoelectron orbitals which are true continuum solutions of a one-electron Hamiltonian containing the nonspherical potential of the H_2O^+ ion. These orbitals hence include the angular momentum coupling brought about by such potentials. This coupling gives rise to features in the ion spectra which are entirely molecular and non-atomic-like in character. For

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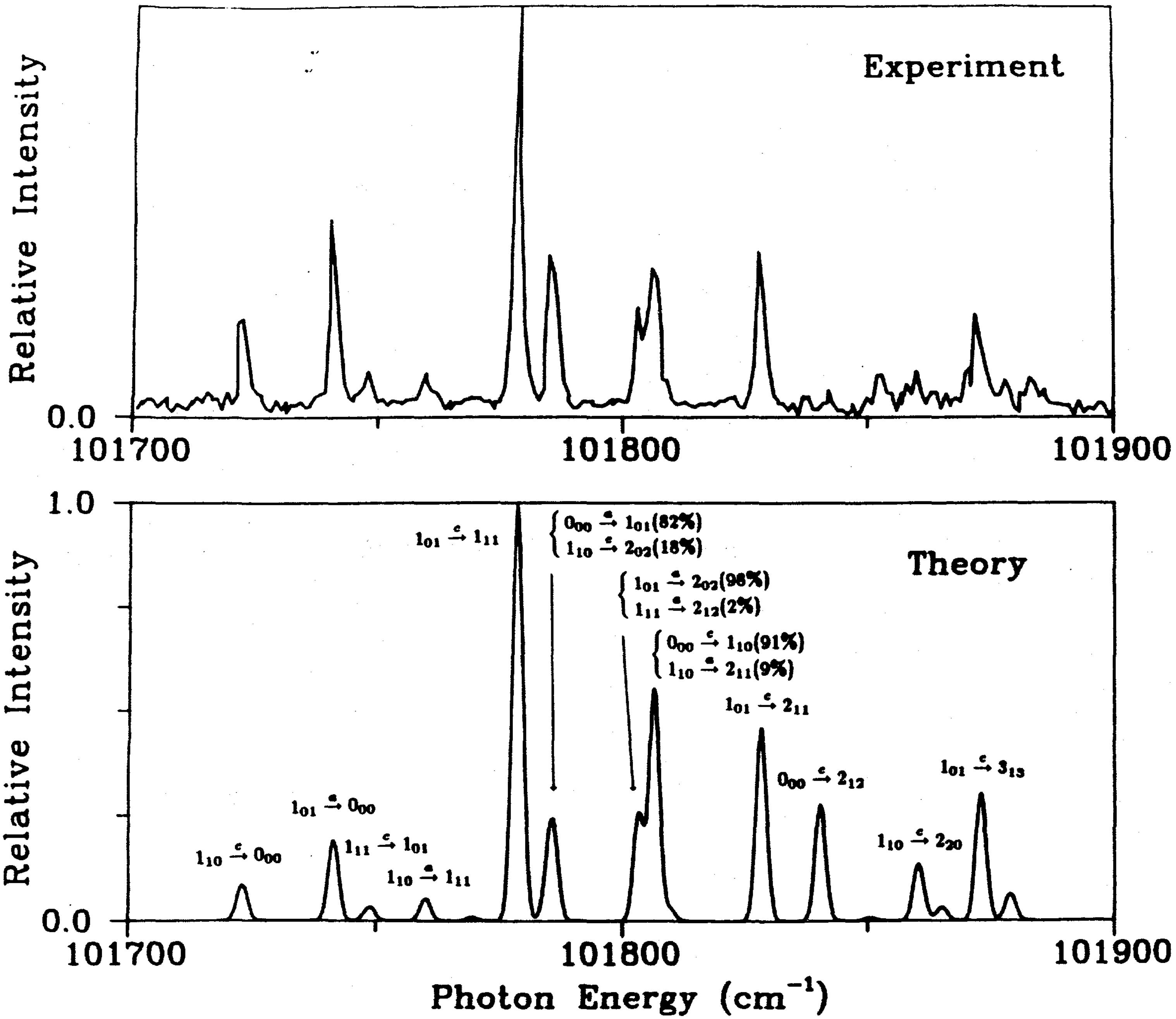

Fig. 4 Measured (ref. 18) and calculated rotational ion distributions resulting from photoionization of H_2O by coherent VUV radiation. Note that rotational transitions labelled a (*type a*) are truly molecular in origin and cannot be accounted for on the basis of any atomic-like model of photoionization.

example, *type a* transitions in fig. 4 are due to such angular momentum coupling in the photoelectron orbital and would not be present in an "atomic" picture of this process. It is clearly important to use the appropriate molecular description of the photoelectron in any analysis of these rotationally resolved spectra.

Future studies of rotational ion distributions resulting from resonance enhanced multiphoton ionization of H_2O are underway.

● *Resonance enhanced multiphoton ion spectra of molecules containing heavier atoms*: To date our studies of vibrational and rotational ion distributions resulting from REMPI have focussed on molecules containing first-row atoms, e.g. OH, NH, CF, and H_2O. We are now extending these studies to molecules containing heavier atoms such as VO, SiH, SiF, and SiF_2. Such studies can be expected to be useful in the use of REMPI for state-specific detection and diagnostics of media containing such species. These molecules with their heavier atoms will reveal a richer and more complex behavior in their ion distributions. For example, our studies of the rotational distributions of VO^+ ions resulting from REMPI of VO have been stimulated by a preliminary report of evidence in the ion spectra of very large changes in rotational angular momentum ($\Delta N = N^+ - N'$) upon photoionization at very low photoelectron energies.[20] These large ΔN (up to 7) imply that *very high angular momentum* (ℓ) *components*, e.g. i waves, are present in the low-energy photoelectron wavefunction. Such behavior clearly has far reaching implications for our understanding of rotational ion distributions formed virtually at the ionization threshold. Studies of these rotational ion distributions in REMPI of VO and SiH are well underway.

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- **The research accomplishments outlined above are described in the following publications.**

1. Rotational branching ratios and photoelectron angular distributions in resonance enhanced multiphoton ionization of diatomic molecules
   K. Wang and V. McKoy
   J. Chem. Phys. (in press)

2. Rotationally resolved photoelectron spectroscopy of the $^2\Sigma^-$ Rydberg states of OH: the role of Cooper minima
   E. de Beer, C.A. de Lange, J.A. Stephens, K. Wang, and V. McKoy
   J. Chem. Phys. **95**, 714 (1991)

3. Effects of Cooper minima in resonance enhanced multiphoton ionization-photoelectron spectroscopy of NO via the D $^2\Sigma^+$ and C $^2\Pi$ Rydberg states
   K. Wang, J.A. Stephens, and V. McKoy
   J. Chem. Phys. (in press)

4. Cooper minima and circular dichroism in photoelectron angular distributions
   H. Rudolph, R.L. Dubs, and V. McKoy
   J. Chem. Phys. **93**, 7513 (1990)

5. Orbital evolution and promotion effects in the photoionization dynamics of $^2\Sigma^-$ Rydberg states of OH
   J. A. Stephens and V. McKoy
   J. Chem. Phys. **93**, 7863 (1990)

6. Non-Franck-Condon effects in photoionization of the 3 $^3\Pi$ Rydberg state of NH
   Kwanghsi Wang, J. A. Stephens, and V. McKoy
   J. Chem. Phys. **93**, 7874 (1990)

7. (2 + 1') rotationally resolved resonance enhanced multiphoton ionization via the E $^2\Sigma^+$ (4s, 3d) and H $^2\Sigma^+$ (3d, 4s) Rydberg states of NO
   H. Rudolph and V. McKoy
   J. Chem. Phys. **93**, 7054 (1990)

8. Rotational branching ratios and photoelectron angular distributions in resonance enhanced multiphoton ionization of HBr via the F $^1\Delta_2$ Rydberg state
   K. Wang and V. McKoy
   J. Chem. Phys. (in press)

9. Rotationally resolved photoelectron spectra in resonance enhanced multiphoton ionization of $HC\ell$ via the $F\ ^1\Delta_2$ Rydberg state
   K. Wang and V. McKoy
   J. Chem. Phys. (submitted for publication)

10. Rotational ion distributions resulting from resonance enhanced multiphoton ionization of NH
    K. Wang, J.A. Stephens, V. McKoy, E. de Beer, C. deLange, and N.P.C. Westwood
    J. Chem. Phys. (manuscript in preparation)

11. Rotationally resolved photoelectron spectra of H_2O at very low photoelectron energies
    M-T. Lee, K. Wang, V. McKoy, R.G. Tonkyn, and M.G. White
    J. Chem. Phys. (manuscript in preparation)

● Education, Training, and Professional Personnel

- Graduate students supported
  1. Matthew Braunstein

- Degrees granted
  1. Matthew Braunstein
     Thesis title: *Photoionization Dynamics and Ion State Distributions in Single-Photon and Resonance Enhanced Multiphoton Ionization of Molecules* (June, 1990)

- Postdoctoral fellows
  1. J.A. Stephens
  2. K. Wang

- **In addition to invited seminars at several institutions, the following presentations were or will be made:**

1. Rotational and Vibrational Distributions in Molecular REMPI
   Gordon Research Conference on Multiphoton Processes,
   June 11-15, 1990 in New London, New Hampshire

2. State-Selective Photoelectron Spectroscopy of Molecules.
   An invited paper at the European Research Conference on Very-High-Resolution Spectroscopy with Photoelectrons: ZEKE Spectroscopy.
   Kreuth near Munich, October 28-31, 1991.
   Note: These conferences are modelled on the Gordon Research Conferences and are sponsored by the European Science Foundation and the Commission of the European Communities.