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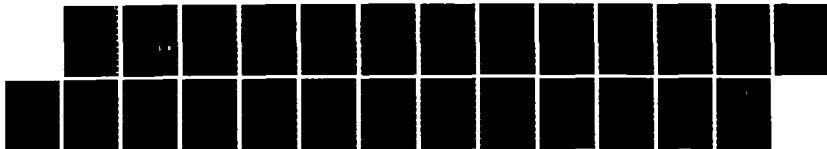
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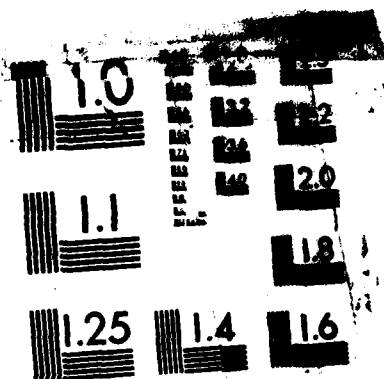
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20. ABSTRACT (Continue on reverse side if necessary and identify by block number) This report summarizes research on resonantly enhanced multiphoton absorption, dissociation, and ionization processes in atoms and molecules. Multiphoton processes are studied using from one to three independently tunable visible and/or UV laser beams in order to establish both the underlying physics and the high degree of selectivity of multiphoton processes. Measurements are made to probe both the formation of excited molecular states and the sub- sequent behavior of excited states either in the presence or in the absence.		

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of further intense laser radiation. Detection methods include laser-induced fluorescence, ion mass analysis, and electron energy analysis.

During the current reporting period, we have studied resonant multiphoton ionization processes^{are studied} for the H_2^+ , I, NO, NeXe, ArXe, KrXe, and Xe_2^+ using both mass spectrometry and photoelectron spectroscopy to analyze the products of the ionizations. In addition, we are developing two new instruments that will provide significantly enhanced resolution, collection efficiency, and versatility in both the ion mass and the electron kinetic energy detection channels. Keywords: $\rightarrow$ to field '9

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ANNUAL SUMMARY REPORT

**HIGH-RESOLUTION SPECTROSCOPY AND DYNAMICS
OF MULTIPHOTON PROCESSES IN ATOMS AND MOLECULES
(Contract No. N00014-86-F-0011)**

Principal Investigators:

**Dr. Patricia M. Dehmer
Argonne National Laboratory
Argonne, Illinois 60439
Tel: (312) 972-4187**

and

**Dr. Joseph L. Dehmer
Argonne National Laboratory
Argonne, Illinois 60439
Tel: (312) 972-4194**

Submitted to:

**Office of Naval Research
Physics Division (Code 1112)
Department of the Navy
800 North Quincy Street
Arlington, Virginia 22217
Attention: Dr. B. R. Junker**

Submitted by:

**Argonne National Laboratory
Argonne, Illinois 60439
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states that are dipole-forbidden in single photon absorption. Second, it is necessary to establish the mechanisms and dynamical parameters governing multiphoton processes. For example, in order to design the most selective and sensitive excitation scheme, one must know the cross sections for each step, the decay mechanisms and the decay rates of the intermediate states, and the cross sections for all competing excitation processes, including nonresonant ones. Such information is largely unknown at this time. Third, this work involves the development of advanced electron energy and ion mass analyzers that are designed specifically for their compatibility with pulsed laser excitation sources.

We presently have operating a 5.1 cm mean radius hemispherical electron spectrometer, a magnetic bottle time-of-flight electron spectrometer, and a time-of-flight mass spectrometer. These instruments are described briefly below. In addition, the following lasers and ancillary laser equipment are presently available in the laboratory: (1) a Molelectron Nd:YAG oscillator and amplifier (Model MY34-10) with 2nd, 3rd, and 4th harmonic generation crystals, an intracavity etalon for line narrowing, and a single-axial mode selector device; (2) A Molelectron dye laser (Model DL18P) with a flowing dye cell amplifier and a doubling crystal which tracks via computer control; (3) a Questec excimer laser (Model 2460); (4) a Lambda-Physik dye laser (Model FL2002E); (5) two computer-controlled, double-grating dye lasers of the modified Littman type -- these ANL-built lasers are capable of producing output with a linewidth of 0.03 cm^{-1} ; (6) two Inrad autotracking frequency doublers; (7) a Lasertechnics Fizeau wavemeter interfaced to a PDP-11/23 computer for measuring the absolute wavelength of a cw laser (to 2 parts in 10^7) or a pulsed laser (to approximately 2 parts in 10^6); (8) a scanning Fabry-Perot interferometer; and (9) an assortment of accessory equipment and parts, such as power meters, mounting hardware, mirrors, prisms, dichroic beamsplitters, and retardation optics. Other equipment, which is readily available, but which has not been used in the initial experiments, include cooled fluorescence detectors, a 0.2 meter vacuum monochromator, a pulsed supersonic molecular beam source, a microwave free radical source, high temperature vapor sources, and VUV capabilities as described in Section V.

The first generation hemispherical electron spectrometer and time-of-flight mass spectrometer have been used for much of this work. The rotatable

electron spectrometer consists of two copper hemispheres of 5.1 cm mean radius, with entrance and exit "zoom" lenses that focus the electron source onto a virtual entrance slit at the entrance to the hemispheres, and then onto the channeltron detector after energy analysis. The electron spectrometer is operated, under computer control, with a constant electron energy through the hemispheres (constant resolution mode). The electron signal is detected using a channeltron electron multiplier and standard pulse counting techniques. The signal is collected only during the 1 μ sec period immediately following a laser pulse. Using a HeI resonance line photoionization source, this spectrometer has achieved a resolution of 5-6 meV. Using the laser photoionization source, the spectrometer has been operated at a resolution of 20 meV. Higher resolution is possible; however, the low duty factor ($\sim 10^{-7}$) associated with the 10 Hz Nd:YAG pulsed laser source usually requires operating at a resolution of at least 20 meV in order to achieve reasonable counting rates.

The time-of-flight mass spectrometer originally was added to the above photoelectron spectrometer in order to perform photoion-photoelectron coincidence studies using a resonance lamp; however, for the laser studies, the mass spectrometer is operated independently of the photoelectron spectrometer. The mass spectrometer consists of 10 stack plates, a 20 cm drift region, and a high current channeltron detector. In the ion counting configuration, a constant voltage is applied to the ion optics to extract and focus the ions onto the multiplier whose output is fed into a gated, charge-sensitive analog-to-digital converter. A variable delay electronic gate, triggered by the laser pulse, is opened for approximately 1 μ sec to accept the ion signal of appropriate mass. The apparatus is completely controlled by a dedicated PDP-11/03 microprocessor.

The magnetic bottle electron spectrometer is a high resolution, high collection efficiency spectrometer. The spectrometer consists of an ionization region, a magnetic lens that parallelizes electrons with different ejection angles, a 50 cm flight tube, and a channel-plate detector. Any electron produced in the ionization region with a finite velocity component in the direction of the detector is transported down the spectrometer axis. Due to the magnetic parallelization of the electron paths, the transit time to the detector is independent of the velocity component transverse to the detector

axis to an accuracy of better than 1% for a 10 eV electron. Hence, the instrument combines extremely high acceptance (~50%) with the high resolution of a time-of-flight device. Another important aspect of this approach is that the acceleration/deceleration of electrons in the weak magnetic field in the flight tube introduces no lens effect, such as that occurring in purely electrostatic lenses. Accordingly, electrostatic retardation can be used to lengthen flight times, thereby improving energy resolution without altering the constant 50% transmission of the device. The combination of resolving power, collection efficiency, kinetic energy range, and constant transmission make this a very important instrumental approach. We estimate that the magnetic bottle electron spectrometer will produce a data collection rate for high resolution electron spectra that is 3-4 orders of magnitude greater than that of our current instrument.

Summarizing, we are able to probe atoms and molecules with a composite laser probe and to measure directly the photoions, photoelectrons, and fluorescent photons. Neutral fragments also can be monitored by subsequent ionization or by laser-induced fluorescence. Spectroscopic and dynamical information is then obtained by monitoring these detection channels as a function of the frequencies and polarization states of one or more of the components of the composite laser probe.

III. PROGRESS

During the most recent contract period, a substantial amount of new work has been carried out. Examples of this work include single and multicolor REMPI experiments on H_2 , I, NO, NeXe, ArXe, KrXe, and Xe_2 using both mass spectrometry (MS) and photoelectron spectroscopy (PES) to analyze the products of the ionization. Highlights of these studies are described below. A related single photon VUV study of autoionization processes in HBr also is described.

1. Photoelectron Angular Distributions and Rotational and Vibrational Branching Ratios for H_2 $C^1\Pi_u$, $v = 0-4$. During the past few years, REMPI-PES has provided an abundance of data on the dynamics of photoionization from excited states of small molecules. The REMPI-PES of molecular hydrogen represents a particularly interesting case as it is both experimentally and theoretically tractable. Recently, we presented photoelectron spectra

following three photon resonant, four photon ($3 + 1$) ionization of H_2 via the $C^1\Pi_u$, $v' = 0 - 4$, $J' = 1$ levels, in which the spectra were recorded along the polarization axis of the laser ($\theta = 0^\circ$). Theoretical calculations performed by others were in good agreement with the experimental results for the lower vibrational levels, although significant differences were observed for the higher vibrational levels. In an effort to clarify these discrepancies and to provide more detailed dynamical information, we now have determined the photoelectron angular distributions for ($3 + 1$) ionization via the same $C^1\Pi_u$, $v' = 0 - 4$, $J' = 1$ levels of the earlier paper. In addition, the present measurements allow the determination of the angle-integrated branching ratios, which could not be evaluated from the $\theta = 0^\circ$ measurements alone.

The $C^1\Pi_u$ electronic state of H_2 is the lowest lying singlet Rydberg state converging to the $H_2^+ \times {}^2\Sigma_g^+$ ground electronic state. As expected on the basis of Franck-Condon arguments, photoionization via a particular vibrational level, v' , of the $C^1\Pi_u$ state leads to a photoelectron spectrum strongly peaked at the ionic vibrational level $v^+ = v'$. The $C^1\Pi_u$, $v' = 0 - 4$ levels show significant Λ -doubling due to the interaction of the Π^+ component with the $B^1\Sigma_u^+$ electronic state. In the present experiments, these perturbations are expected to lead to enhancement of the $v^+ \neq v'$ photoelectron peaks. In an effort to avoid these perturbations, the earlier spectra and the angular distributions reported here were recorded via the $Q(1)$ branch, i.e., via the unperturbed Π^- levels. However, it is of interest to note that the spectra recorded via the $R(0)$ and $R(1)$ branches, which access the Π^+ component of the $C^1\Pi_u$, $v' = 4$, $J' = 2$ level, display strongly v^+ -dependent rotational branching ratios. In these spectra, the $v^+ \neq v'$ rotational branching ratios show dramatically different behavior than the $v^+ = v'$ branching ratios, indicating different photoionization dynamics for the diagonal ($v^+ = v'$) and the off-diagonal ($v^+ \neq v'$) vibrational peaks.

The $H_2 C^1\Pi_u$, $v' = 0 - 4$, $Q(1)$ photoelectron angular distributions determined here provide a detailed basis for comparison with theoretical calculations aimed at producing an understanding of the excited state photoionization dynamics of molecular hydrogen. Although the only currently available calculations are in excellent agreement with the $v^+ = v'$ photoelectron angular distributions, the agreement with the off-diagonal angular distributions is relatively poor.

2. Rotational and Vibrational Branching Ratios for H_2 C $^1\Pi_u$ Using the Magnetic Bottle Electron Spectrometer. We are presently taking advantage of the 2π collection angle, the high resolution, and the high sensitivity of the recently commissioned magnetic bottle electron spectrometer to determine rotational and vibrational branching ratios for a much broader set of excited levels of the C $^1\Pi_u$ state than would be feasible with the dispersive analyzer used in the experiments described in Item 1. Such complete and comprehensive studies are necessary for a complete understanding of the photoionization dynamics of excited molecular states.

3. Photoelectron Branching Ratios for H_2 B' $^1\Sigma_u^+$ and D $^1\Pi_u$. The studies described in Item 1 above have been extended to the higher energy B' $^1\Sigma_u^+$ and D $^1\Pi_u$ electronic states using three photon resonant, four photon ($3 + 1$) ionization. The D $^1\Pi_u$ electronic state is the $3p\pi$ member of the $np\pi$ Rydberg series for which the C $^1\Pi_u$ state is the lowest member ($2p\pi$). Photoelectron spectra obtained via the D $^1\Pi_u$, $v' = 0$ and 1 levels closely resemble the corresponding C $^1\Pi_u$, $v' = 0$ and 1 photoelectron spectra, with the $v' = v^+$ photoelectron peak dominating the spectra. The B' $^1\Sigma_u^+$ state corresponds to the $3p\sigma$ state, which dissociates to the same limit as the B $^1\Sigma_u^+$, $2p\sigma$ state ($H(1s) + H^*(2s)$). The photoelectron spectra obtained via the B' $^1\Sigma_u^+$, $v' = 0-2$ levels show that H_2^+ is formed in a wide distribution of vibrational levels for each of the resonant intermediate levels. This is not unexpected, as the B' $^1\Sigma_u^+$ state contains significant ion-pair character, and the Franck-Condon factors show good overlap with a wide distribution of ionic vibrational levels. However, owing to the more complex nature of the B' $^1\Sigma_u^+$ electronic state, these photoelectron spectra should provide a challenging test for theoretical calculations.

4. Two Photon Spectroscopy of Autoionizing Rydberg States of NO. The two photon ionization spectrum of rotationally cooled nitric oxide (NO) was determined at high resolution in the region between the $NO^+ X \ ^1\Sigma^+$, $v^+ = 0$ and 2 thresholds (2500-2680 Å). The present two photon ionization spectrum is completely different from the corresponding single photon ionization spectrum owing to the difference in selection rules (or, in this case, propensity rules) for one and two photon absorption. In particular, the $n = 7 - 11$ members of two Rydberg series converging to the $NO^+ X \ ^1\Sigma^+$, $v^+ = 1$ and 2 thresholds are observed and are assigned as previously unobserved members of

the nd series. In addition, the 5d $v = 2$ band is observed at somewhat lower energy. All of the nd bands show sharp rotational structure, which is currently being analyzed. Several other bands also are observed, which have not yet been assigned.

5. Wavelength Dependent Photoelectron Spectra of Rotationally Resolved Autoionizing Levels of Nitric Oxide. Nitric oxide (NO) has been the subject of a number of recent studies using resonantly enhanced multiphoton ionization-photoelectron spectroscopy (REMPI-PES). For the most part, these studies have focused on the photoionization dynamics of the bound, resonant intermediate level, although evidence for the influence of autoionizing levels on both the photoelectron spectra and the photoelectron angular distributions has also been discussed. Two studies have been performed more recently using two-color REMPI-PES to excite autoionizing Rydberg states of NO and to determine the vibrational branching ratios. These studies show strong deviations from the propensity rule for pure vibrational autoionization, which predicts that processes with the smallest possible change in the vibrational quantum number of the ion core of the Rydberg state will be the most intense.

In the present study, direct (non-resonant), two photon excitation was used to probe the previously unobserved NO $9d\sigma\pi$, $v = 2$ band, which shows sharp rotational structure and which can autoionize into both the $\text{NO}^+ X^1\Sigma^+$, $v^+ = 0$ and $v^+ = 1$ continua. The direct ionization continua for two photon ionization into the $\text{NO}^+ v^+ = 0$ and $v^+ = 1$ levels are reasonably intense in this region, and it is possible to determine photoelectron spectra both for the $9d\sigma\pi$ autoionizing levels and for direct ionization into the $v^+ = 0$ and $v^+ = 1$ continua. By contrast, in two color excitation schemes the resonant intermediate level is generally a Rydberg state, and the $\Delta v = -1$ transition to the direct ionization continuum is usually much weaker than the corresponding $\Delta v = 0$ transition to the autoionizing levels, making such on-resonance, off-resonance comparisons difficult. We determined wavelength dependent photoelectron spectra and, in addition, photoelectron angular distributions at a number of wavelengths within the band. Both the photoelectron branching ratios and the angular distributions show strong variations across the rotationally resolved autoionization profiles. The data display a strong increase in the $\text{NO}^+ X^1\Sigma^+$, $v^+ = 1$ branching ratio on resonance, in accord with the $\Delta v = -1$ propensity rule for vibrational autoionization. An

analysis of the data indicates that the autoionization mechanism leads to approximately 96% $\text{NO}^+ \ ^1\Sigma^+$, $v^+ = 1$.

6. Autoionizing Rydberg States in Atomic Iodine. During the past few years, considerable progress has been made in the experimental study of photoionization of halogen atoms; however, the best resolution attained to date in such experiments ($\sim 20 \text{ cm}^{-1}$) is not sufficient to resolve all of the features of interest. For this reason, we investigated the possibility of using laser techniques for the study of this problem. We used two color multiphoton ionization mass spectrometry to determine the spectra of the optically allowed autoionizing states of atomic iodine. In these experiments, the first laser is used to produce atomic iodine by the photodissociation of methyl iodide and to pump the iodine atoms to a low lying $\dots 5p^4 6p$ state via a two photon transition. A second laser is used to probe single photon transitions from these $\dots 5p^4 6p$ states to autoionizing $\dots 5p^4 ns$ and nd Rydberg states converging to the 1D_2 ionization limit. Because a total of three photons is used, this process accesses states of the same parity as those accessed by single photon excitation. However, the resolution in the present experiments is limited only by the linewidth of the laser. Thus, it is possible to study the photoionization spectrum of atomic iodine and other open shell atoms with unprecedented detail.

Spectra were obtained by pumping the two photon transition from the $^2P_{1/2}$ state to the $(^1D_2)6p[3]_{5/2}$ and $(^1D_2)6p[1]_{1/2}$ states and probing transitions to the $(^1D_2)ns$ and nd Rydberg series converging to the 1D_2 ionization limit. The former spectrum displays three sharp series that can be resolved to high principal quantum numbers ($n \sim 35$), while the latter spectrum displays a single sharp series and a single broad series. The symmetry of several of these series was deduced from angular momentum coupling rules.

7. Photoelectron Spectroscopy of Rydberg States of Atomic Iodine.

Several recent REMPI-PES studies have shown that ionization into a structureless continuum via an unperturbed Rydberg state usually proceeds by the ejection of the Rydberg electron without a change in the electronic or vibrational state of the ion core. Thus, in many instances REMPI can be used as source of state-selected ions. However, the results can be quite different if the resonant intermediate level is perturbed, because the character of the unperturbed level will then be mixed with that of the perturbing level. If

the two interacting levels have different ion cores, the perturbation is revealed in the photoelectron branching ratios following REMPI. In this work, REMPI-PES was used to study a perturbation in the odd parity Rydberg series of atomic iodine and to determine the feasibility of producing state-selected I^+ ions using REMPI. It was shown that, while state selected ions can be produced in some cases, the state selecting capability of REMPI is dramatically reduced for perturbed levels. For example, the photoelectron spectra obtained via the $(^3P_2)nf[3]_{5/2}$ levels show the effect of a perturbation by the $(^3P_0)4f[3]_{5/2}$ level.

As a consequence of the small ionization cross sections of the Rydberg levels in these experiments, two processes were observed that compete with REMPI. The first involves fluorescence of the resonant intermediate level to a lower lying level, which is then ionized with greater efficiency, and the second involves energy transfer (either collisional or radiative) between $I^* 2P_{1/2}$ atoms and the Rydberg atoms. Although these two processes would be indistinguishable from direct REMPI using mass spectrometric techniques alone, the use of REMPI-PES allows the separation of the contributions of these different mechanisms and provides a more complete understanding of the overall process.

8. Analysis of Rydberg State Spectra of NeXe, ArXe, KrXe, and Xe₂.

Vibronic structure in the REMPI-MS spectra of NeXe, ArXe, KrXe, Xe₂ were analyzed in the energy region containing the atomic Xe^{*} 5d and 6p states. Several new bands were observed for each species, providing new information on the excited state potential curves and on the binding in van der Waals molecules. In the case of Xe₂, the excited states have gerade (g) symmetry and, therefore, cannot be observed in single photon studies. In the case of the heteronuclear van der Waals molecules, all but two of the observed band groups occur in the neighborhood of an excited atomic Xe state that is allowed via two photon transitions from the atomic ground state. Thus, the empirical observation of single photon absorption studies that the most intense van der Waals dimer transitions are to molecular states derived from dipole allowed atomic dissociation limits also appears to apply to two photon transitions. In the wavelength regions near the two photon allowed atomic Xe transitions, the NeXe, ArXe, and KrXe spectra display a number of similarities, and the interpretation of these spectra is consistent with the prediction that the

trend in the dissociation energies for the excited neutral dimer states will follow the trend in the dissociation energies of the corresponding dimer ion states, i.e., $D_0(\text{NeXe}^*) < D_0(\text{ArXe}^*) < D_0(\text{KrXe}^*)$.

9. Potential Energy Curves of the Ionic States of Xe_2 . Several years ago, we determined photoelectron spectra of the homonuclear rare gas dimers Ar_2 , Kr_2 , and Xe_2 using conventional HeI photoelectron spectroscopy. However, since the rare gas dimer is present in concentrations of only a few percent in the supersonic molecular beam, the HeI photoelectron spectrum is dominated by photoelectrons from photoionization of the free atoms. In the case of Xe_2 , these atomic photoelectron peaks completely or partially obscure 3 of the 6 molecular photoelectron bands. However, the REMPI-PES of Xe_2 taken in the region of the $\text{Xe}^* 5d$ and $6p$ excited states, provide new information on the ionic states. In one example, the REMPI-PES of Xe_2 showed all four molecular bands in the region of the $\text{Xe}^+ 2p_{3/2}^0$ dissociation limit (corresponding to the $\text{Xe}_2^+ A 2\Gamma_u^+$, $B 2\Pi_{3/2g}$, $B 2\Pi_{1/2g}$, and $C 2\Pi_{3/2u}$ ionic states) with no evidence of atomic photoionization. This is in sharp contrast to the HeI-PES obtained by us earlier, in which the atomic bands were about a factor of 100 more intense than the molecular bands. Franck-Condon factor calculations are now being performed on the present data to obtain detailed information on the potential energy curves of the ionic states.

10. Vibrational Effects in the Spin-Orbit Autoionization of HBr. Recently, a multichannel quantum defect theory of spin-orbit autoionization in molecules with 2Π ion cores was developed and applied to the photoionization spectrum of HI. It was found that the calculated spectrum between the $2\Pi_{3/2}$ and $2\Pi_{1/2}$ ionization thresholds was in good qualitative agreement with the observed spectrum. The present work is both an experimental and theoretical extension of this work to HBr. The high resolution relative photoionization cross section for HBr determined in the energy region between the $2\Pi_{3/2}$ and $2\Pi_{3/2}$ ionic limits was compared to new theoretical calculations. The theory for HBr is more complex than that for HI since the spin-orbit splitting of the $X 2\Pi$ state in HBr^+ (2651 cm^{-1}) is nearly equal to the vibrational spacing of each 2Π substate in HBr^+ ($\Delta G = 2348 \text{ cm}^{-1}$). Consequently, Rydberg states with low principal quantum numbers that converge to the $2\Pi_{3/2}$, $v^+ = 1$ limit appear in the energy region between the $2\Pi_{3/2}$, $v^+ = 0$ and $2\Pi_{1/2}$, $v^+ = 0$ thresholds. It is therefore necessary to generalize the theory used for HI to

include additional vibrational channels. The vibrational couplings due to the R-variation of both the electrostatic and the spin-orbit matrix elements were introduced and were shown to be necessary in order to reproduce the qualitative features of the experimental data.

IV. PUBLICATIONS

The papers, conference abstracts, and invited talks resulting from this work are appended. New items during the current contract period are papers 26-33, abstracts 24-28, and invited talks 37-42.

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PAPERS - Continued

14. P. M. Dehmer, P. J. Miller, and W. A. Chupka, "Photoionization of $N_2^+ X^1 \Sigma_g^+, v''=0$ and 1 Near Threshold -- Preionization of the Worley-Jenkins Rydberg Series," J. Chem. Phys. 80, 1030 (1984).
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