V. ATOMIC RESONANCE AND SCATTERING

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1. FIELD IONIZATION AND PHOTOIONIZATION

U.S. Energy Research and Development Administration (Contract EG-77-S-02-4370)

Daniel Kleppner, Michael G. Littman, Michael M. Kash, Harold J. Metcalf

We have constructed a new tunable dye laser system for the study of field ionization in highly excited atoms. Pump power is provided by a Quanta Ray Nd:YAG laser with second, third, and fourth harmonic generators. A new type of dye laser has been devised which has important advantages of simplicity and economy. Work has begun on a study of field ionization of lithium. The atom is prepared in a Rydberg state by a three-step excitation process, using two fixed-frequency and one tunable dye lasers.

2. HIGHLY EXCITED ATOMS

U.S. Air Force – Office of Scientific Research (Contract F44620-72-C-0057)

Daniel Kleppner, Theodore W. Ducas, Michael G. Littman, Myron L. Zimmerman

We have carried out a study of the Stark structure of barium. The aim of this work was to study a two-electron atom in a region where a valence state (a low-lying state of the excited core) interacts with a Rydberg progression. The interactions of the valence state with the Rydberg manifolds were clearly displayed. The system was analyzed by combining a simple configuration-interaction picture with a calculational method

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previously developed for analyzing one-electron Rydberg Stark structure. A paper on the work[1] will appear in J. Phys. B.

References

1. M. L. Zimmerman, T. W. Ducas, M. G. Littman, and D. Kleppner, "Stark Structure of Barium Rydberg States."

3. SUBMILLIMETER PHOTON COUNTING

Joint Services Electronics Program (Contract DAAB07-76-C-1400)

Daniel Kleppner, Theodore W. Ducas, William P. Spencer,
A. Ganesh Vaidyanathan

We have constructed a FIR laser and have observed resonance transitions in Rydberg atoms at 118 μm and 496 μm. A number of transitions at CO_2 wavelengths near 10 μm have also been observed. Work has begun on a study of noise sources and the resonance line shape of the proposed detection. A millimeter wave phase-locked klystron system is under construction; it will be used for the study of millimeter wave resonance transitions.

4. STUDIES IN OPTICAL PHYSICS

Joint Services Electronics Program (Contract DAAB07-76-C-1400)

Daniel Kleppner, Michael G. Littman, William P. Spencer,
A. Ganesh Vaidyanathan

We have developed methods for producing highly excited states of a variety of atoms, including all the alkalis and several of the alkaline earths. Both cw and pulsed laser techniques have been used. A new apparatus has been constructed for high-resolution spectroscopic studies, and techniques have been developed for introducing infrared and millimeter-wave radiation to the atomic beam interaction region.

5. STUDIES IN ATOMIC PHYSICS

National Science Foundation (Grant PHY75-15421-A01)

Daniel Kleppner, Myron L. Zimmerman, Jarbas C. Castro Neto

We have obtained initial results on a study of a highly excited one-electron atom in a high magnetic field. The problem is of interest because the system is fundamentally

simple, yet no comprehensive theory exists. An atomic beam of sodium was excited by tunable dye lasers in the center of a superconducting solenoid, and the structure of levels with principal quantum numbers in the vicinity of n = 25 was studied in magnetic fields up to 65 kG. The diamagnetic interaction, normally an extremely small perturbation, was large enough to mix several terms. We have analyzed the data using a perturbative approach based on all terms in the range $22 \leq n \leq 31$. Quantitative agreement is good. The work is being extended to larger values of n, where the structure starts to resemble Landau levels.

6. SPECTROSCOPY OF VAN DER WAALS MOLECULES

National Science Foundation (Grant PHY77-09155)

Riad N. Ahmad, Walter P. Lapatovich, William P. McGrath,
Philip E. Moskowitz, David E. Pritchard

The sodium neon (NaNe) data obtained from our seeded molecular beams machine[1] has been analyzed in some detail. The ground state well depth of this weakly bound molecule is found to be $D_{0X} = 10^{-3}$ eV, and the equilibrium separation of Na and Ne atoms is $r_e = 5.3 \times 10^{-8}$ cm; for the first excited state we find $D_{0A} = 1.74 \times 10^{-2}$ eV and $R_e - 2.7 \times 10^{-8}$. This means that NaNe is, by almost an order of magnitude, the weakest diatomic molecule to be studied in an isolated condition. Experimental determination of these interatomic potentials shows several theoretical calculations[2, 3] to be in error and provides a method of determining the applicability of certain techniques used to analyze earlier rare gas-alkali scattering data.[4] Our apparatus is now undergoing substantial modification so that we may conclude our study of the NaNe complex and continue on to investigate other systems.

References

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2. W. E. Baylis, J. Chem. Phys. 66, 3778 (1977).
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4. G. M. Carter, D. E. Pritchard, M. Kaplan, and T. W. Ducas, Phys. Rev. Letters 35, 1144 (1975).

7. COLLISIONAL BROADENING OF THE Na D LINES

National Science Foundation (Grant PHY77-09155)

Douglas J. Ely, William D. Phillips, Robert Walkup, David E. Pritchard

Collisional broadening of the Na D lines has long been a subject of intensive theoretical and experimental investigation, both because of interest in the fundamental process and for application to the study of such lines in stellar atmospheres. We have begun experiments which, in contrast to most previous work on the subjects, employ techniques to study sub-Doppler collision broadening. One is saturated absorption spectroscopy, which can be used selectively to observe only those atoms having a specific velocity along a given axis. Thus the velocity dependence of line broadening processes can be studied with increased resolution over current methods which use temperature dependence to infer velocity dependence. Four-wave mixing, a technique in which three laser beams create a polarization in Na vapor which radiates a fourth coherent beam, can also be used to study line broadening and has the advantage of being able to select more than one component of the Na velocity. Finally, using photon counting techniques to observe laser-induced fluorescence in Na vapor, one can detect the small Lorentzian absorption wings of the collisionally broadened Na line at frequencies well beyond the Doppler width. These various techniques all yield information about the collisional linewidth for widths small compared with the Doppler width. The last technique can also easily be used for linewidths greater than the Doppler width, so we can make contact between the two regimes of collisional broadening. Work is in the preliminary stages. We have observed saturated absorption and fluorescent signals, as well as four-wave mixing signals.

8. DIFFERENTIAL CROSS-SECTION MEASUREMENTS USING DOPPLER SELECTIVE LASER FLUORESCENCE ANALYSIS

National Science Foundation (Grant CHE76-81750)

David E. Pritchard, Douglas J. Ely, William D. Phillips, John A. Serri, Kermit R. Way, James L. Kinsey
[James L. Kinsey is Professor in the Department of Chemistry, M.I.T.]

By placing a cw narrow-band dye laser along the relative velocity axis of two atomic beams in a crossed-beam experiment, we are able to detect the scattered velocity distribution of one of the beams by Doppler-tuning the atoms into resonance and detecting their subsequent fluorescence. Atoms are excited into resonance when the projection of their velocity along the direction of the laser beam, $v_\ell = v \cos \theta_{cm}$, is equal to

$c(\nu-\nu_o)/\nu_o$. The frequency ν_o represents the rest frame resonance frequency and ν selects a certain velocity component. By recording the fluorescence signal as a function of ν, we directly obtain the center of mass differential cross section versus cos θ_{cm}. Transformation from laboratory angles to center-of-mass angles is not required in the analysis. The collision we studied was

$$Na^*(3P_{1/2}) + Ar \rightarrow Na^*(3P_{3/2}) + Ar - .002 \text{ eV}.$$

The first excited state of sodium was produced by a velocity selective laser beam, Doppler-tuned to excite the $3S_{1/2}$ to $3P_{1/2}$ transition. Another laser, tuned to the $3P_{3/2}$ to 4D transition, detected $Na^*(3P_{1/2})$ that underwent the fine structure changing collision. The detection signal resulted from the 4P → 3S transition after the atom decayed from the 4D → 4P state. Our measured cross sections yielded oscillatory structure which seems to be in approximate agreement with recent theoretical calculations. Construction of a new beams machine is now under way to use this technique (coined VADS, Velocity Analysis using the Doppler Shift) on molecular systems. We plan to measure the angular differential cross section for state-to-state, vibrational, and rotational changing collisions of Na_2 upon collision with a rare gas atom. Supersonic oven sources will be utilized to provide a cold Na_2 beam of high density, and to provide a narrow velocity distribution for both beams.

9. STUDIES OF ROTATIONAL ENERGY TRANSFER

U.S. Air Force – Office of Scientific Research (Grant AFOSR-76-2972A)

Ibrahim Al-Agil, Timothy Brunner, Richard D. Driver, David E. Pritchard, Neil Smith, Mark D. Wainger

We have studied the rotational energy transfer process

$$Na_2^*(vj) + Xe = Na_2^*(v, j+\Delta j) + Xe - \Delta E$$

where a Na_2 molecule, in the excited A electronic state and with vibrational and rotational quantum numbers v and j, respectively, collides with a xenon atom and changes its rotational quantum number to $j + \Delta j$ with a corresponding loss ΔE in translational energy. We have used the velocity selection by Doppler shift technique[1] to obtain the velocity dependence of the rotational changing rate constants. The experimental rate constants obtained using the VSDS technique have some thermal averaging present from the velocity components which are unspecified by the exciting laser. We have developed a Fourier transform deconvolution technique which permits us to obtain the dependence of the rate constants on relative velocity from the experimental data. An example of this experimental data, together with the deconvoluted rate constant, is shown in

Fig. V-1 for the v = 18, j = 38 state for a collision with $\Delta j = -2$. A paper describing this work is in preparation.

We have studied the dependence of the cross sections for a given initial j on Δj and noted how our data change with different initial j within a given vibrational state. These

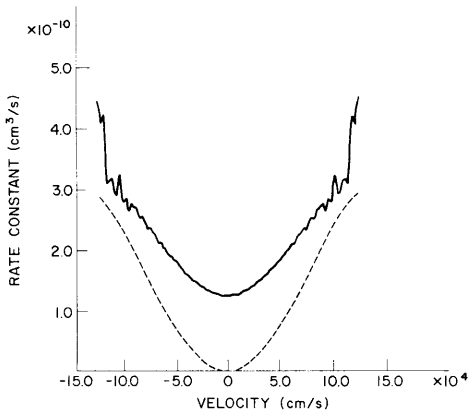

Fig. V-1.

Plots of the experimental and deconvoluted rate constants with collision velocity for $j_{initial} = 38$ and $\Delta j = -2$.

— — — Deconvoluted data vs v_z.

——— Experimental data vs v_{rel}.

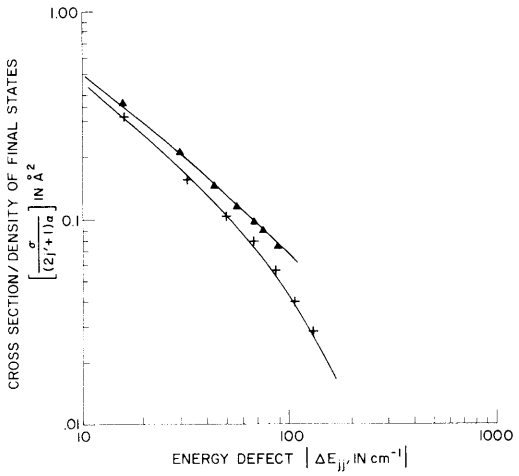

Fig. V-2. Plot of the cross section/density of final states against the energy defect of the collision. The density of the final states is the product of the rotational final state density (2j'+1) and the momentum state density $\alpha = [E-E_{j'}/E-E_j]^{1/2}$.

▲ Experimental data $\Delta j < 0$ collisions.

\+ Experimental data $\Delta j > 0$ collisions.

——— Parametrized fits of the form $+\ln[\sigma_{jj'}/(2j'+1)\alpha] = -a \ln|\Delta E_{jj'}| - b|\Delta E_{jj'}| - c$.

For $\Delta j < 0$ the parameters are a = -0.789, b = -0.00219, c = 1.142. For $\Delta j > 0$ the parameters are a = -0.642, b = -0.0095, c = 0.754.

data are extremely valuable in the light of recent theories of rotational energy transfer using the information-theoretic-based prediction of surprisal theory.[2] We have proved conclusively that the linear surprisal model is not adequate to describe our experimental data, and we will shortly be in a position to differentiate between a number of the alternative surprisal theories recently proposed (see Fig. V-2). Our experiment represents the most definitive experimental study of atom-diatom energy transfer so far undertaken: selections of the initial and final vibrational-rotational levels of the diatomic have been made and we have studied the velocity dependence of the collision cross section.

References

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