V. ATOMIC RESONANCE AND SCATTERING

Academic and Research Staff

Prof. Daniel Kleppner
Prof. David E. Pritchard
Dr. Christopher H. Becker
Dr. Stuart S. B. Crampton*
Dr. Richard D. Driver
Dr. Theodore W. Ducas†
Dr. Richard A. Gottscho
Dr. Mark D. Havey
Dr. Michael G. Littman
Dr. Harold J. Metcalf‡
Dr. Alejandro Morales-Mori
Dr. William D. Phillips
Dr. Kermit R. Way
Dr. Haider R. Zaidi
William M. Hamilton
Annie Spielfiedel**

Graduate Students

Ibrahim Al-Agil
Lawrence R. Brewer
Timothy A. Brunner
Jarbas C. Castro Neto
Randall G. Hulet
Allan W. Karp
Michael M. Kash
Walter P. Lapatovich
Alan L. Migdall
Philip E. Moskowitz
John A. Serri
David A. Smith
Neil Smith
William P. Spencer
A. Ganesh Vaidyanathan
Robert E. Walkup
Myron L. Zimmerman

1. ATOMS IN STRONG MAGNETIC FIELDS

National Science Foundation (Grant PHY77-09155)

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

Magnetic interactions of Rydberg atoms are qualitatively different from those of tightly bound atoms. The diamagnetic interaction can not only exceed the paramagnetic interaction, it can exceed the term separation, and even the total electrostatic interaction.

We have studied the diamagnetic structure of sodium in an intermediate region where it exceeds the term separation and starts to display free-electron (Landau-like) behavior. The atoms are stepwise-excited to Rydberg states within a superconducting solenoid and are detected by field ionization. An atomic beam is used. The atoms move parallel to the field. Motional Stark effects are greatly reduced, permitting the study of states unperturbed by parity mixing or Stark shifts.

We have observed completely resolved levels in the vicinity of n = 28 at fields up to 60 kG. We have been able to account quantitatively for the diamagnetic structure by solving the secular equation for levels in the range of n = 25 to n = 31. The work has also been extended to n = 33 at 60 kG, where the energy starts to vary linearly with field, instead of quadratically, a characteristic of Landau-like behavior. We can account for

---

*Visiting Faculty Fellow from Williams College.

†Assistant Professor at Wellesley College.

‡Visiting Associate Professor from State University of New York at Stony Brook.

**Visiting Scientist from Observatoire de Paris, France.

the structure using our perturbative method based on a low-field representation, although it is apparent that this approach is reaching the limit of usefulness.

Our most recent efforts have been centered on the construction of a new apparatus which should permit us to achieve 100 kG. In addition, plans are under way for employing a swept cw dye laser instead of a pulsed dye laser, which will provide a hundred-fold increase in resolution.

Publications

Myron L. Zimmerman, Jarbas C. Castro Neto, and Daniel Kleppner, Phys. Rev. Lett. 40, 1083 (1978).

2. FAR INFRARED DETECTION WITH RYDBERG ATOMS

Joint Services Electronics Program (Contract DAAG29-78-C-0020)

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

We have detected radiation at 496 μ using Rydberg atoms and the technique of selective field ionization. Sodium atoms are prepared in the 26s state, and the incident IR radiation induces transition to the 26p state. The radiation is supplied by a methyl fluoride FIR laser. The Rydberg transition is brought into coincidence with the laser by applying a field of 27 V/cm. The transition tunes at the rate of approximately 250 MHz/(V/cm). The radiation is detected in a pulsed mode at a rate of 10 pps. The interaction time is 1.3 μsec, and the resonance linewidth is typically 1 MHz. The FIR power was measured directly by using a calibrated power meter followed by attenuators, and by observing the broadening of the resonance. The two methods agreed to within a factor of two.

The detector combines features of incoherent and coherent detection. It is fundamentally a photon-counting device and resembles an incoherent detector in having no inherent limitation on the detection area or angular aperture. On the other hand, it is narrow-banded and tunable, features usually associated with a heterodyne system.

The marginal sensitivity of the detector was taken to be the quotient of the power needed to drive the resonance and the observed signal-to-noise ratio. In our initial study the sensitivity was 3×10^{-15} watt/$\sqrt{\text{Hz}}$. The quantum efficiency was 0.1%, although this can be substantially increased. It is believed that the observed noise can be greatly reduced, and that much higher sensitivity is possible.

We have also observed resonance IR absorption at 10.8 μ and 118 μ, although we have not studied the systematics in detail. Our method should be applicable throughout the IR spectrum, and in the millimeter-wave region.

3. FIELD IONIZATION AND PHOTOIONIZATION

U.S. Department of Energy (Grant EG-77-S-02-4370)

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

We have undertaken a study of the role of nonadiabatic effects on field-ionization processes. Briefly, if a Rydberg atom is subjected to a rapidly increasing electric field, it can make transitions to other states as the electric field sweeps the energy levels through successive level anti-crossings. The ionization characteristics of the system are then dominated by the properties of these levels, rather than the initial state.

The process by which an atom "jumps" to another state as the energy levels are swept through an anti-crossing is called the Landau-Zener effect. We are studying the Landau-Zener effect in a two-part program. The first part comprises the accurate mapping of an anti-crossing in order to determine the parameters which enter the Landau-Zener theory. This is essentially completed. We have mapped a group of crossings between the n = 18 and n = 19 levels of lithium, using a cw dye laser with 30-MHz resolution. A number of the crossings appear to be good candidates for studying the Landau-Zener effect.

We have also studied the anti-crossings theoretically. Our calculations are in good agreement with the observation, and give us confidence that anti-crossings can be reliably calculated for many Rydberg systems.

We have also undertaken photoionization measurements of Rydberg atoms. Photoionization from Rydberg states is important to energy transfer in stellar atmospheres and in plasmas. Relatively little experimental work has so far been carried out on photoionization from excited species. Our initial studies are on photoionization near the continuum edge, where the cross sections vary most rapidly. We have employed a CO_2 laser to photoionize Rydberg states of sodium, and have observed both s and d states on the range n = 12 to n = 20. The experimental results are still preliminary. Nevertheless, they are in qualitative agreement with calculations that we have carried out based on the Coulomb approximations.

Publications

Michael G. Littman, Michael M. Kash, and Daniel Kleppner, Phys. Rev. Lett. 41, 103 (1978).

(V. ATOMIC RESONANCE AND SCATTERING)

4. SPIN-POLARIZED HYDROGEN

National Science Foundation (Grant DMR 77-10084)
National Aeronautics and Space Administration (Grant NSG-1551)

Daniel Kleppner, Thomas J. Greytak, Stuart S. B. Crampton,
William D. Phillips, David A. Smith, Abel Weinrib

We have developed a source of atomic hydrogen at liquid-helium temperature, and have studied the properties of H on frozen H_2 using spin resonance. The zero-field hyperfine transition has been observed with atoms stored in a bulb coated with solid H_2. The frequency shift due to wall collisions and the transverse and longitudinal relaxation times have been measured.

The cold-hydrogen source should have a number of applications in atomic and surface physics, as well as in the production of spin-polorized hydrogen.

5. EFFICIENT ENERGY-TRANSFER PROCESSES

Joint Services Electronics Program (Contract DAAG29-78-C-0020)

Neil Smith, David E. Pritchard

Using a technique for selecting velocity based on the Doppler shift,[1,2] we have completed study of the velocity dependence of the total cross section for collisions which change the fine-structure level of Na atoms,[3]

$$Na(3P_{1/2}) + X \rightleftarrows Na(3P_{3/2}) + X,$$

where X can be an atom or a molecule. These cross sections are typically 100 $\mathring{A}^2$, and we found that the target determined whether they increased or decreased with velocity. For some of the target atoms we studied, the velocity dependence has subsequently been measured in a crossed-beams machine – this has enabled us to show that our technique gives reliable results within 5%.

We have also completed a theoretical paper[4] in which we have been able to give a procedure based on Fourier transforms which can remove the thermal averaging inherent in this technique, as well as in several other classes of collision experiment.

References

1. W. D. Phillips and D. E. Pritchard, Phys. Rev. Lett. 33, 1254 (1974).
2. J. Apt and D. E. Pritchard, Phys. Rev. Lett. 37, 91 (1976).

3. J. Apt and D. E. Pritchard, J. Phys. B, to be published.
4. N. Smith, T. A. Brunner, R. D. Driver, and D. E. Pritchard, J. Chem. Phys. 69, 1498 (1978).

6. DETAILED STUDY OF EXCITED-STATE ENERGY-TRANSFER PROCESSES

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

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

One of the most probable inelastic collisions involving molecules is Rotational Energy Transfer (RET). We have measured rate constants for the RET process

$$Na_2^*(v, j_o) + X \rightarrow Na_2^*(v, j_o + \Delta) + X - \Delta E_r,$$

where v is the vibrational quantum number, j_o is the initial rotational quantum number, X is a rare-gas atom, and ΔE_r is the increase in rotational energy. We use a tunable dye laser to populate the desired initial level, and observe the resulting fluorescence with a monochromator to monitor the populations of the levels.

The results of our experiment with Na_2^* in collision with Xe are shown in Fig. V-1 where we plot the rate constant divided by a translational (R) and a spin (N_o) density of final states versus energy transfer. The positive ΔE_r data fall on the same line as those with negative ΔE_r due to our use of a novel spin-space factor N_o which assumes conservation of the magnetic quantum number. Furthermore, the data follow a power law[1,2] rather than the previously accepted exponential gap law.[3] We have applied this power-scaling law to other experimental and theoretical data[4] and found it to be superior to the exponential gap law in every case.

We are currently measuring the velocity dependence of the above RET process by using the Doppler shift to vary the velocity component of the excited molecule along the laser beam. To reduce the averaging due to unselected translation degrees of freedom we have developed a new deconvolution procedure.[5] We are currently measuring the velocity dependence of RET in Na_2^*-Xe collisions.

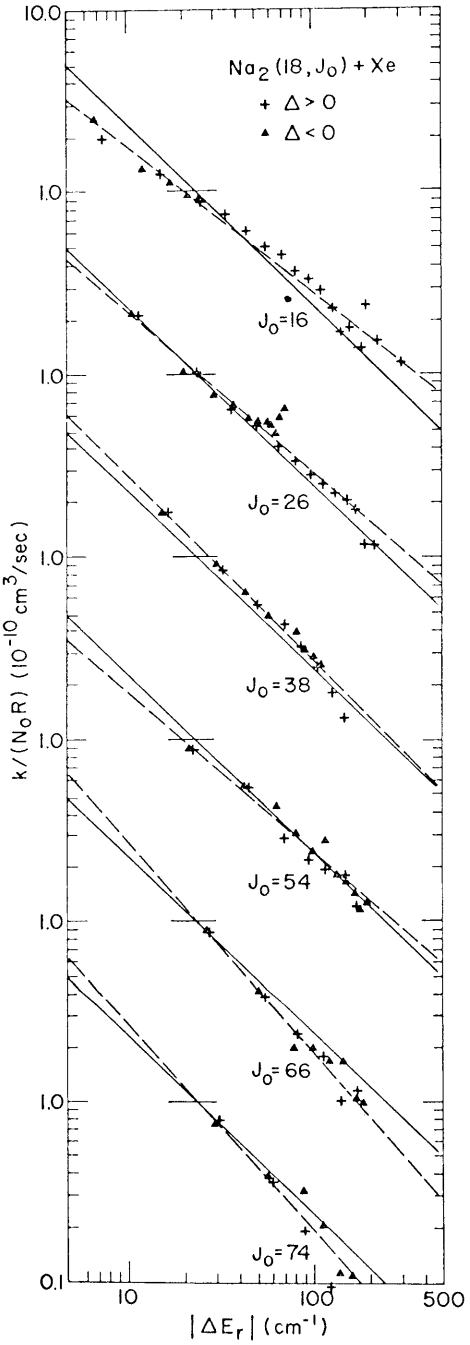

Fig. V-1. $k/(N_0R)$ versus $|\Delta E_r|$. Dashed lines are power-law fits to data for individual j_0. Solid lines are all a single power-law function which fits all data well.

References

1. T. A. Brunner, R. D. Driver, N. Smith, and D. E. Pritchard, Phys. Rev. Lett. 41, 856 (1978).
2. T. A. Brunner, R. D. Driver, N. Smith, and D. E. Pritchard, J. Chem. Phys. 70, 2115 (1979).
3. R. D. Levine, R. B. Bernstein, P. Kahana, I. Procaccia, and E. T. Upchurch, J. Chem. Phys. 64, 796 (1976).
4. D. E. Pritchard, N. Smith, R. D. Driver, and T. A. Brunner, J. Chem. Phys. 70, 4155 (1979).
5. N. Smith, T. A. Brunner, R. D. Driver, and D. E. Pritchard, J. Chem. Phys. 69, 1498 (1978).

7. STATE-RESOLVED DIFFERENTIAL CROSS-SECTION MEASUREMENTS USING DOPPLER VELOCITY ANALYSIS

National Science Foundation (Grant CHE76-81750)

John A. Serri, Richard Mittleman, Alejandro Morales-Mori,
David E. Pritchard, Christopher H. Becker, James L. Kinsey
[James L. Kinsey is Professor in the Department of Chemistry, M. I. T.]

We have demonstrated a new technique for measuring inelastic differential cross sections. This method, named Angular Distribution using the Doppler Shift (ADDS) is based on the idea that atoms are excited into resonance by a laser at frequency ν only when the projection of their velocity along 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. By tuning a laser beam which travels along the relative velocity axis of two intersecting atomic beams and recording the subsequent fluorescence signal as a function of ν from one of the beams, we directly obtain the center of mass differential cross section versus $\cos \theta_{cm}$. The collision process studied was[1]

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

The first excited state of sodium ($3P_{1/2}$) was produced by a laser beam tuned to the $3S_{1/2}$ to $3P_{1/2}$ transition. A second laser beam, placed on the relative velocity axis and tuned across the $3P_{3/2}$ to 4D sodium transiton, analyzed the scattering angle of the $Na^*(P_{3/2})$ produced by the structure state-changing collisions. Figure V-2 compares our experimental results (points) with an ab initio calculation convoluted with the experimental resolution profile and the appropriate hyperfine structure.

The ADDS method is now being applied to rotational energy transfer in diatomic

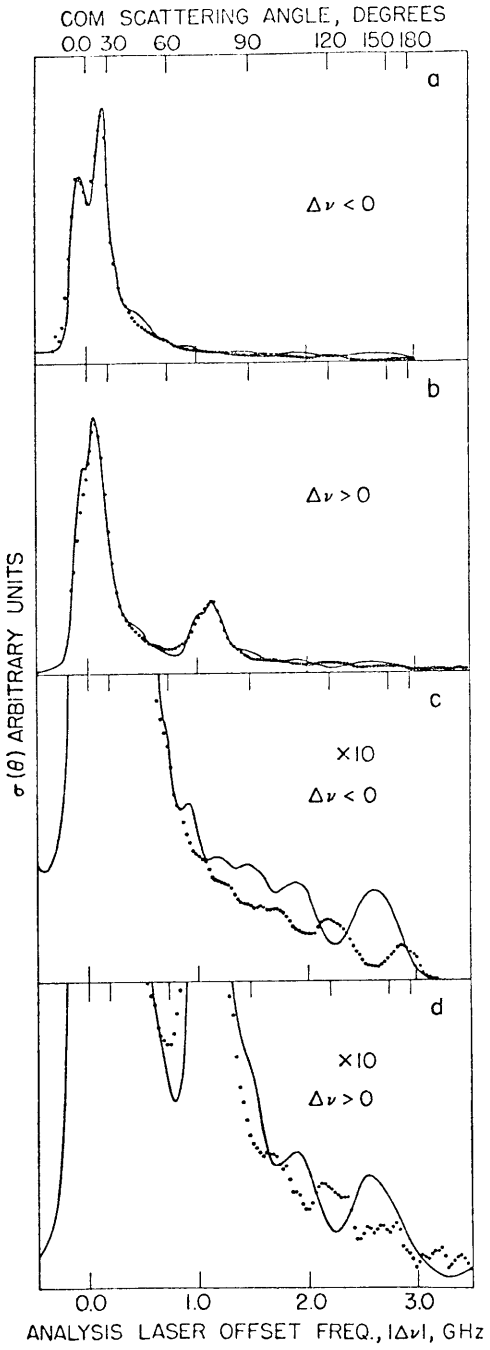

Fig. V-2. Straight lines – theoretical fit.
Points – experimental data.

sodium,

$$Na_2(V, J) + Ar \rightarrow Na_2(V, J') + Ar + \Delta E,$$

where V is the vibrational quantum number. The experiment uses two lasers, one to modulate the dimer population of the initial state (V, J) and the other to analyze the final state (V, J'). The angular distribution of the final state (V, J') resulting from inelastic collisions with the argon will be measured by fluorescence from the Doppler-tuned transition $X^1\Sigma(v, j) \rightarrow A^1\Pi(v', j \pm 1)$. Collisions which connect states (v, j) and (v, j') will then be isolated by phase detecting at the modulation frequency of the laser tuned to the (V, J) level.

These measurements will be conducted in a new crossed-beams machine which utilizes Campargue nozzle sources[3] to produce high-intensity molecular beams with narrow velocity distribution and low internal temperature. Construction is complete, and at the present time we are performing beam diagnostics on the dimers using laser-induced fluorescence. Our immediate objective is to optimize the production of rotationally cold Na_2 and to further reduce the velocity spread, currently 10% FWHM.

References

1. W. D. Phillips, J. A. Serri, D. J. Ely, D. E. Pritchard, K. R. Way, and J. L. Kinsey, Phys. Rev. Lett. 41, 937 (1978).

2. R. P. Saxon, R. E. Olson, and B. Liu, J. Chem. Phys. 67, 2692 (1977).

3. R. Campargue and J. P. Breton, Entropie 42, 18 (1971).

8. MOLECULAR SPECTROSCOPY

National Science Foundation (Grant PHY77-09155)

Mark D. Havey, Walter P. Lapatovich, Philip E. Moskowitz,
David E. Pritchard

We are studying weakly bound diatomic molecules. The van der Waals molecule NaAr has been produced in a supersonic expansion, and laser-induced bound-bound transitions have been observed. The spectra obtained extends earlier data of Smalley and his co-workers,[1] and shows much hitherto unseen structure, particularly in the region where the excited state is close to dissociation. A long-range analysis,[2] utilizing the new data, is currently in progress. Analysis of the spectra attributable to the highest vibrational levels of the $A^2\Pi$ excited state (and comparison to spectra from NaNe) should yield information about the elusive $B^2\Sigma$ state. This would complete the experimental determination of all potentials necessary to calculate line-broadening and

atomic-collision processes for this system. This will provide a sharp test of theories for these processes.

Future plans include studying the free-bound transitions in NaHe.

References

1. R. E. Smalley et al., J. Chem. Phys. 66, 3778 (1977).
2. R. J. Le Roy, Molecular Spectroscopy, The Chemical Society (Burlington House, London), Vol. 1.
3. R. Ahmad-Bitar et al., Phys. Rev. Lett. 39, 1657 (1977).