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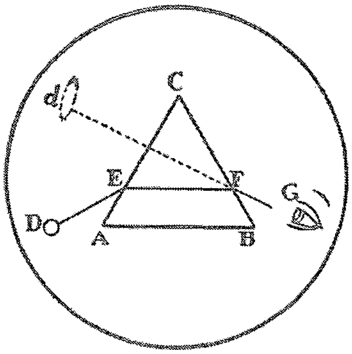

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Eleventh International Quantum Electronics Conference

Session A
Monday, June 23, 1980
8:30 A.M. **Independence Room**
Laser Spectroscopy I
Chairman: P. F. Liao

A.1. Doubly Excited Alkaline Earth Atoms* (Invited), T. F. GALLAGHER, K. A. SAFINYA, AND W. SANDNER, *SRI International, Menlo Park, CA,* AND W. E. COOKE, *University of Southern California, Los Angeles, California.* (30 min.)

We describe recent experiments to probe the properties of doubly excited autoionizing atoms using a laser spectroscopic technique. The physical basis of the excitation process as well as experiments to probe the autoionization process are described.

* This work supported by AFOSR, NSF, and DOE.

A.2. Rydberg Atom Masers (Invited), S. HAROCHE, C. FABRE, P. GOY, M. GROSS, AND B. MOI, *Laboratoire de Physique de l'Ecole Normale Supérieure, Paris, France.* (30 min.)

Using optically pumped alkali Rydberg atoms as the active medium, we have recently developed new types of pulsed maser sources with unusual characteristics. Due to the giant electric dipole matrix elements of the Rydberg states, these masers have extremely low inversion thresholds—several orders of magnitude smaller than those of conventional masers—and "microscopic" energy outputs. Very sensitive detection procedures have to be used in order to observe their emission. An indirect detection method consists in studying the field ionization characteristics of the atoms, which is strongly modified when maser action occurs. Direct detection of the tiny microwave bursts has also been achieved by a heterodyne technique with a very sensitive Schottky diode mixer. Using both methods, we have made a detailed study of the emission characteristics (typical number of inverted atoms at threshold $\sim 10^4$; pulse energy ~ 1 to 10 eV; peak emission power $\sim 10^{-12}$ W; pulse duration ~ 0.5 μs).

Owing to the large number of energy levels near the ionization limit, these new masers can be operated at many wavelengths ranging from the centimeter to the submillimeter range. In fact, the source should continuously evolve from the maser to the laser case when the binding energy of the levels involved in the emission is increased. These new coherent sources are bound to have very interesting applications in fundamental physics (study of superradiance of very small systems of atoms) and for the technology of millimeter and submillimeter wave detectors as well.

A.3. Singlet-Triplet Mixing by Hyperfine Interactions in ^{3}He, R. R. FREEMAN, P. F. LIAO, R. PANOCK, AND L. M. HUMPHREY, *Bell Telephone Laboratories, Holmdel, NJ 07733* (15 min.)

A complete determination of the hyperfine structure of the 2^3P and 3^3D states of ^{3}He through analysis of Doppler-free intermodulated fluorescence spectra is reported. We find the structure of the 3^3D state to be significantly modified by singlet-triplet mixing which is induced by hyperfine interactions. The hyperfine interaction is dominated by the Fermi contact interaction of the inner 1s open shell electron with the nucleus and therefore does not decrease for higher lying states. Hence, unlike the case of one electronlike spectra (e.g., alkali atoms) or multielectron atoms with zero spin (e.g., ^{4}He) the high Rydberg states of ^{3}He will have their electronic structures completely dominated by the hyperfine interaction. In particular, the hyperfine induced singlet-triplet mixing for ^{3}He will increase rapidly with increasing principal quantum number n. Our results are in good agreement with theoretical calculations of the hyperfine interaction.

In Fig. 1 we show a portion of our spectrum which contains transitions associated with the $2^3P_{1,2}$ levels to the $3^3D_{1,2,3}$ states of ^{3}He. These states were obtained in a dc discharge tube operated with 0.8 Torr of ^{3}He. The tube is probed with two counterpropagating tunable laser beams which are chopped at different frequencies. By monitoring fluorescence at the difference frequency we obtain the Doppler-free spectrum shown in Fig. 1. This spectrum is fit to a parametrized hyperfine Hamiltonian and the calculated resonance positions and line strength from the fit are shown in the figure. We find the majority of the interaction is due to the Fermi contact term of the 1s electron and that this term is nearly the same for the $2P$ and $3D$ states as expected. This term produces sizeable singlet-triplet mixing which must be included to correctly give the structure. The dotted lines show resonance positions if one neglects this mixing.

Because the hyperfine interaction is essentially constant, our results, along with published fine structure measurements allow a precise determination of the structure of all higher lying states in ^{3}He. We find, for example, we can reproduce the two-photon spectra recently obtained by Giacobino *et al.*[1] and also predict the hyperfine splittings of the n $^1D_2$ states observed in level crossing experiments.[2] In Table I we give our calculated values for these splittings and the measured experimental values. There is excellent agreement. As one of the simplest atoms, helium is amenable to accurate calculations. In Table I we also include the results of a theory based on hydrogenic electronic wave functions and good agreement with our calculations is again found.

In conclusion we have made a determination of the hyperfine interaction in ^{3}He. This determination shows important singlet-triplet mixing effects which will dominate the

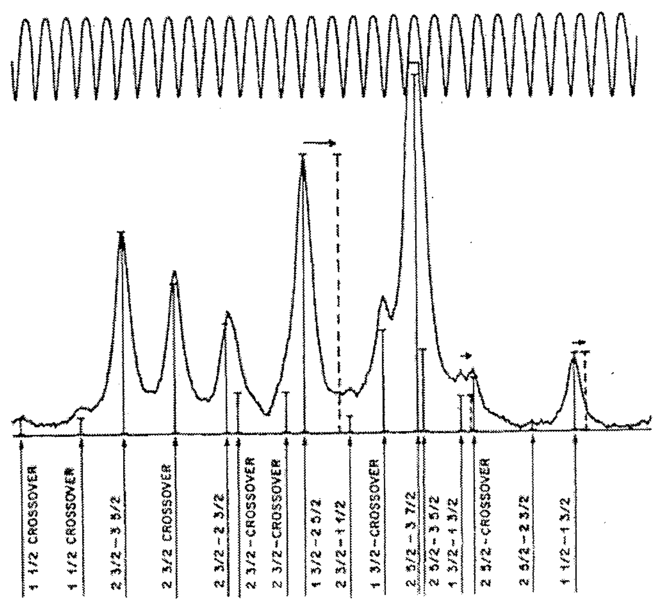

FIG. 1. Portion of Doppler-free spectrum of 2^3D–3^3D transitions in ^{3}He. The levels are marked by $(2^3P)F$–$(2^3D)F'$. The upper trace is the transmission of an interferometer having an FSR = 122.4 MHz. The solid lines show the calculated positions and intensities including singlet-triplet mixing. The dotted lines show the calculated positions if singlet-triplet mixing is ignored.

cm^{-1}. The magnitude of the cross section for collisional deexcitation by spontaneous emissions, σ_S, was determined by measuring the total number of signal photons integrated over the emission bandwidth and using, for the number of detected photons,

$$N[\mathrm{Ba}(5d\,^1D_2)]N[\mathrm{Tl}(6p\,^2P^0_{3/2})]\sigma_S\overline{V}\tau V_0\zeta,$$

where $N[\mathrm{Ba}(5d\,^1D_2)]$ and $N[\mathrm{Tl}(6p\,^2P^0_{3/2})]$ are the number densities for the initial storage levels, $\overline{V}$ is the mean velocity of collision, τ is the effective radiating time, V_0 is the effective radiating volume, and ζ is the ratio of detected to generated photons. In this manner we obtain a measured value for the cross section for dipole-quadrupole collisional deexcitation of $\sigma_S = 1.5 \times 10^{-22}$ cm^2, with an overall experimental uncertainty of approximately a factor of 7.

The results of this experiment have application to the construction of low-gain, high-energy storage media and to the spectroscopic study of the interaction potentials of colliding atoms.

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J.8. Sodium Plasmas Produced by Milliwatt cw Laser Irradiation,* M. E. KOCH, K. K. VERMA, AND W. C. STWALLEY, *Iowa Laser Facility and Departments of Chemistry and Physics, University of Iowa, Iowa City, IA 52242.* (15 min.)

There are a variety of reports of significant laser-produced ionization of alkali metal vapors using pulsed lasers (in Li[1] and in Na[2]) and using cw lasers (in Cs[3] and in Na[4]). (See also Ref. 5 for additional background.) While all of this work is quite interesting, much of it involved resonance lines[1,2,4] and so is perhaps not terribly surprising. The other work,[3] on the other hand, involves transitions between a radiatively trapped upper level of an alkali-metal resonance line and a more highly excited level which can associatively ionize to form M_2^+. In principle, then, this cw plasma may contain a concentration of M_2^+ which is quite nonequilibrium. Moreover, in contrast to a discharge where M_2^+ may be rapidly destroyed by photodissociation, the M_2^+ may be stable with respect to laser and other light (e.g., near-resonance lines) found in the laser-produced plasma.

With this in mind, we have irradiated a sodium heat pipe (typically at 10 Torr), using a focused cw dye laser at 5688.2 or 5682.6 Å ($3p \rightarrow 4d$), and also reproduced the Cs result[3] at 6010 Å. Unlike Cs,[3] where a strong atomic ion-electron radiative recombination continuum is seen, we see no significant spectroscopic evidence for *atomic* ions in our Na plasma. We feel this is because in Cs, at the upper level of transition studied by Tam and Happer, the channel of ion pair formations ($\mathrm{Cs}^{**} + \mathrm{Cs} \rightarrow \mathrm{Cs}^+ + \mathrm{Cs}^-$) is available in addition to associative ionization ($\mathrm{Cs}^{**} + \mathrm{Cs} \rightarrow \mathrm{Cs}_2^+ + e^-$). However, for $\mathrm{Na}^{**} = \mathrm{Na}(4d)$, only associative ionization can occur energetically, so we have produced essentially a *molecular* ion plasma. Also we note that this plasma can be produced at quite low power (~2 mW focused broadband laser light at 5688.2 Å!) and we are currently examining the energy balance in detail.

We have obtained spectra of this plasma in the 2000–9000 Å region. The interpretation of this spectrum is still not completely clear. The various atomic lines seen can be understood in terms of Na($4d$)–Na and Na($3p$)–Na($3p$) collisions[6] and the process (dissociative recombination): $\mathrm{Na}_2^+ + e^- \rightarrow \mathrm{Na}^{**} + \mathrm{Na}$, where Na^{**} is a highly excited Na atom (e.g., $4d$ or $5s$). The structure seen near the exciting line and to the red is presumably molecular fluorescence and D line absorption. We see five broad features at ~3650, 3780, 4350, 4520, and 8000 Å which remain to be explained.

We have examined the 4200–4700 Å region under high resolution and find the structure in that region to be a continuum, not densely spaced lines. A possible explanation is that these continua represent the processes $\mathrm{Na}_2^+ + e^- \rightarrow \mathrm{Na}_2^* + h\nu$, where Na_2^* is an excited state of Na_2. The occurrence of such molecular ion-electron radiative recombination has never been previously established, although the atomic form is well known. The features we see peaking at 4350 and 4520 Å have been observed in other ways, e.g., in discharges,[7–9] in Ar^+ laser UV-line irradiation of the Na_2 $C \leftarrow X$ bands,[10] in two-photon Na_2 excitation,[11] and in cw and pulsed dye-laser excitation at the Na D lines.[12–14] Similar features occur in K, Rb, and Cs.[8,15,16] Several explanations have been proposed involving free-free, free-bound, or bound-free processes. Note that the radiative recombination discussed above can be cast in "bound-free" form when a high molecular Rydberg state Na_2^{**} is formed as a resonance in e^-–Na_2^+ scattering. Since many of the potential energy curves of Na_2 are fairly well known, e.g., from high quality *ab initio* calculations and a variety of recent experiments, we are carrying out explicit calculations of a number of these alternatives. We also have and will continue to carry out simultaneous ionization detection to attempt to resolve the origin of the 4350 and 4520 Å continua. Finally we note that some mechanisms suggest these bands might be made into a powerful violet laser with limited tunability.

The 8000 Å feature almost certainly corresponds to the

$$A\,^1\Sigma_u^+ - X\,^1\Sigma_g^+$$

satellite band[17,18] and has previously been observed in a Na (or other alkali) discharge.[8,18,19] There is a continuum overlaid with many discrete lines. The discrete lines, however, extend through all parts of the laser path while the continuum is concentrated in the central "white" region near the focus where the 4350 and 4520 Å features appear.

The 3650 and 3780 Å features have apparently not been previously reported. They also appear continuous and possible explanations for them are similar to those mentioned above for the 4350 and 4520 Å bands.

We are currently examining these spectra and extending them in a variety of ways, with emphasis on obtaining microscopic understanding of the plasma formation process.

*Supported by the National Aeronautics and Space Administration and the National Science Foundation.

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J.9. Laser-Induced Penning/Associative Ionization in Crossed Atomic Beams, P. POLAK-DINGELS, J.-F. DELPECH, AND J. WEINER, *Department of Chemistry, University of Maryland, College Park, MD 20742.* (15 min.)

Laser-switched or laser-modified collisions are the object of intensive theoretical and experimental study because they offer the possibility of controlling the relative probabilities of competing inelastic and reactive exit channels. The influence of the laser field is to modify the electronic states of the system during a collisional encounter. Laser-induced collisions are characterized by atomic-field interactions which are nonresonant with respect to dipole-allowed transitions of the separated collision partners. We discuss here new results on Penning/associative ionization of Na/Na collisions in the presence of optical field power densities of $\simeq 10^7$ W/cm^2.

The experimental set-up is as follows. Two alkali atomic-beam sources are mounted on a multiported vacuum chamber at right angles in the horizontal plane. Two laser beams enter from opposite ports and overlap at the interaction region with an angle of nearly 180°. The light sources are flash-lamp pumped tunable dye lasers synchronized together and with a box car integrator/amplifier used to record the ion signal. A quadrupole mass filter, mounted above the