GENERATION OF COHERENT VUV (VACUUM ULTRAVIOLET) AND SOFT X-RAYS

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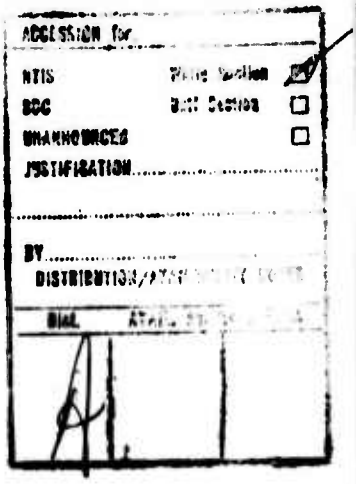

I. TECHNICAL SUMMARY

The purpose of this program is the development of techniques for the generation of coherent vacuum ultraviolet and soft x-ray radiation. During this reporting period three projects have been active. The first project is a study of three frequency summing in inert gases for the generation of short wavelength radiation. The conversion efficiency of the mixing process is enhanced by choosing two of the input frequencies so that they sum to a non-allowed atomic transition. To date we have studied both helium and neon systems; we have concluded that the helium experiment is impractical because of the technical difficulties involved in generating the required wavelength. These difficulties are much less severe for the neon system, because of a natural near-coincidence of a neon non-allowed transition and the 16^{th} harmonic of the Nd:YAG laser frequency. This fact greatly simplifies the experimental apparatus, and substantially improves the signal-to-noise ratio. The results of our calculations, and our experimental plan to generate 626 Å are described in Section II.

Our second project resulted in the first observation of a laser induced inelastic collision. This new type of atomic collision process was first proposed and studied under this program, and appears to be a very promising technique for the construction of short wavelength lasers. The process involves the efficient exchange of stored energy from one atomic specie to

another in the presence of an intense optical field, while in the absence of the field the exchange does not take place. In our experiment, energy was switched from the resonant level of strontium to an excited state level of calcium by the application of a short visible laser pulse. Further details of the experiment are given in Section III, and in a paper entitled "Inelastic Collision Induced by Intense Optical Radiation," attached as Appendix B.

Section IV describes our studies of thermal velocity charge exchange collisions as a method of selectively exciting strongly forbidden levels of singly ionized column IIA elements, leaving the ground state empty. Such a reaction results in a large inversion to the ground state and the inverted system could be used as a stimulated anti-Stokes laser or as a two-photon laser. We have successfully demonstrated a charge exchange collisional energy transfer from Mg^+ to the $Sr^+(4d^2D)$ metastable level; however, low population densities have so far prevented us from firmly establishing the selective nature of the process. Demonstrating the selective nature of the Mg^+-Sr collision remains a major goal of this project. We plan to show the selectivity of the process by creating higher metastable densities, transferring them to an allowed Sr^+ level, and thus creating measurable laser gain. Future work on this project will be solely supported by an ERDA contract.

The work reported here was jointly supported by the Office of Naval Research under Contract N0001$\frac{4}{}$-75-C-0576.

II. HARMONIC GENERATION OF EXTREME UV RADIATION

(K. S. Hsu, L. J. Zych, J. F. Young, and S. E. Harris)

This project is directed towards the generation of coherent extreme UV and soft x-ray radiation using three frequency summing in inert gases. The conversion efficiency of the mixing process is enhanced by choosing two of the input frequencies so that they sum to a non-allowed atomic transition. The inert gases are the simplest atomic species having such suitable energy levels in the extreme UV spectral region; to date we have studied both helium and neon systems. We have concluded that the helium experiment is impractical because of the technical difficulties involved in generating the required wavelengths. These difficulties are much less severe for the neon system, and we are presently constructing an experiment which will generate 626 Å radiation.

Sum Generation in Helium

Helium has the highest energy levels of any of the inert gases, and thus is most suitable for harmonic generation of extremely short wavelengths. We generated one-half the non-allowed He (1s-2s) transition, 1908.8 Å, using a parametrically amplified dye laser followed by frequency mixing in mercury. Details of this work are described in the paper attached as Appendix A. Because of the technical complexity of the tunable sources that have to be

used, only 300 watts of 1202.8 Å radiation could be generated. Unfortunately, our calculations indicate that this will not provide sufficient signal-to-noise ratio for reliable detection of the resultant 569 Å signal, and this approach is no longer being pursued.

Sum Generation in Neon

Resonant two-photon-pumped sum generation in neon is simplified by a natural near-coincidence of a Ne 2p-3p non-allowed transition and the 16^{th} harmonic of the Nd:YAG laser frequency. Thus, tunable sources are not required to resonantly enhance the nonlinear susceptibility; instead, one may use any two Nd:YAG harmonics which sum to the 16^{th} harmonic, e.g., the 8^{th} + 8^{th}, the 7^{th} + 9^{th}, etc. This fact greatly simplifies the experimental apparatus, and the higher powers generally available at these fixed wavelengths, relative to tunable sources, substantially improves the signal-to-noise ratio. Therefore, our efforts during the last six months have been primarily devoted to this approach.

The simplest pumping scheme uses two photons of the 8^{th} harmonic of the Nd:YAG laser: 1330 Å. Unfortunately, our attempts to generate 1330 Å in Hg, and in Cd, both failed to produce usable signal levels. We believe the sum generation efficiency in Hg was reduced by cancelling contributions to the susceptibility from the many higher transitions involved; the magnitude and sign of these matrix elements are not known and exact calculations are not possible. In Cd multi-photon ionization limited the power density and reduced the output power. Thus the simplest pumping scheme does not appear practical.

Resonant pumping with a combination of the 9^{th} and 7^{th} harmonics, however, now appears quite feasible. Over the past several years we have developed techniques for the efficient generation of 1182 Å, the 9^{th} harmonic of Nd:YAG, using third harmonic generation in phase matched mixtures of Xe and Ar. Typically, useful peak powers of 30 kW are available. In addition, we have recently produced 10 kW peak power pulses of 1520 Å, the 7^{th} harmonic, using sum generation in Cd vapor. These pumping levels are 100 times greater than those available for the He experiment, and further improvements are probable.

Figure 1 is an energy level diagram of our proposed experiment: input wavelengths of 1182 Å, 1520 Å, and 10,640 Å are summed to 626 Å. The third input wavelength of 10,640 Å was chosen for several reasons; it is naturally present in the optical path following the generation of 1182 Å and 1520 Å, and high peak powers are available. In addition, because the sum wavelength lies just above the Ne (4s) levels we will be able to phasematch the process, thus improving the conversion efficiency by perhaps 100. Based on the dispersion of the Ne levels we estimate a coherence length $L_c = 4.8 \times 10^{17}/N$ cm , and a Ne:He phasematching ratio of 1:2.3. The calculation of the nonlinear susceptibility involves the unknown Ne (3s-3p) oscillator strength. Using a conservative value of 0.01, however, we estimate a single coherence length conversion efficiency of 10^{-4} from 1182 Å to 626 Å, yielding signal-to-noise ratios of greater than 100.

The spatial and temporal combination of three laser beams with widely differing frequencies is a difficult technical problem. A schematic of our proposed experiment is shown in Fig. 2. A mode-locked Nd:YAG oscillator-amplifier system, plus KDP crystals, provides two separated but synchronous

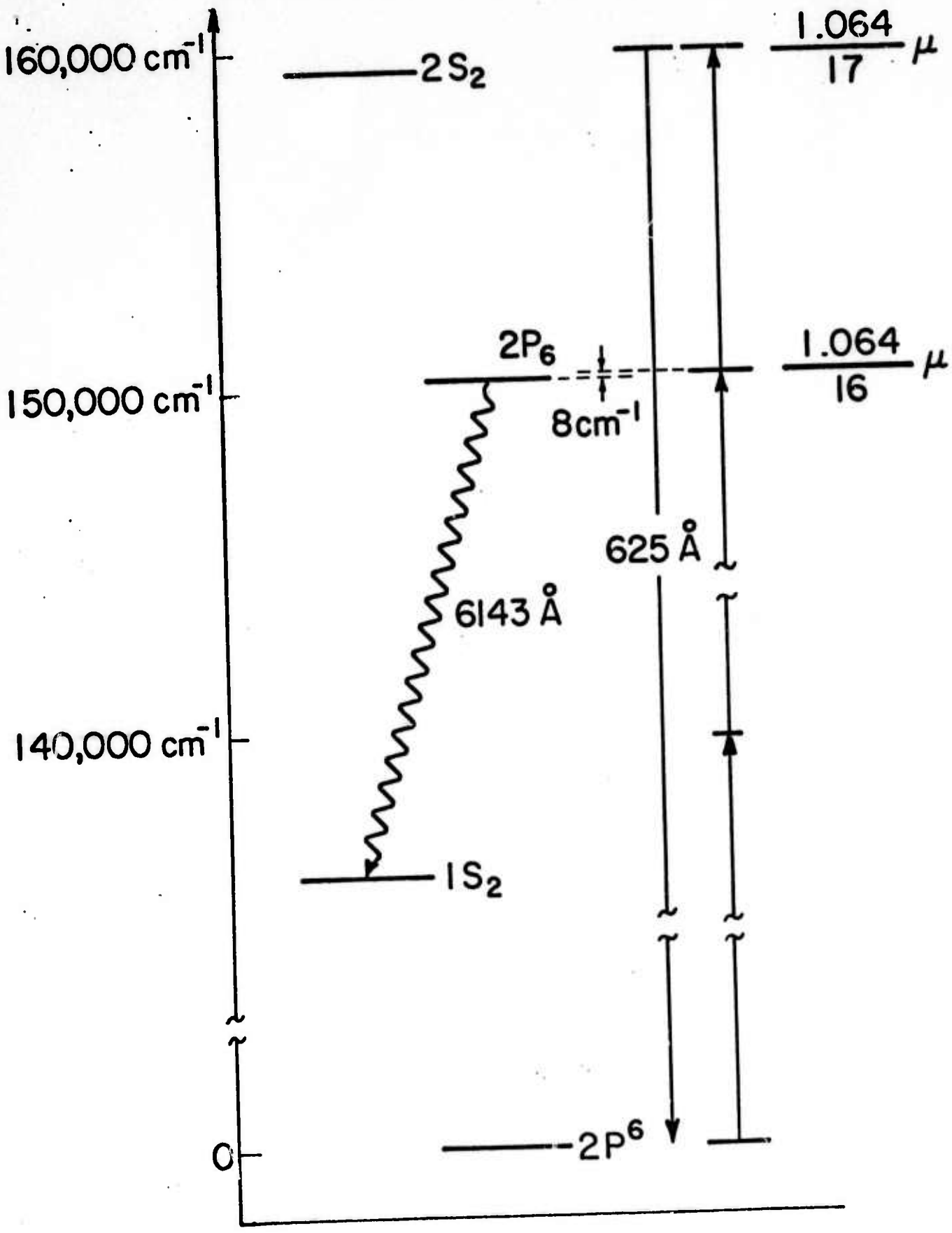

FIGURE 1

Energy level diagram of neon (Paschen notation).

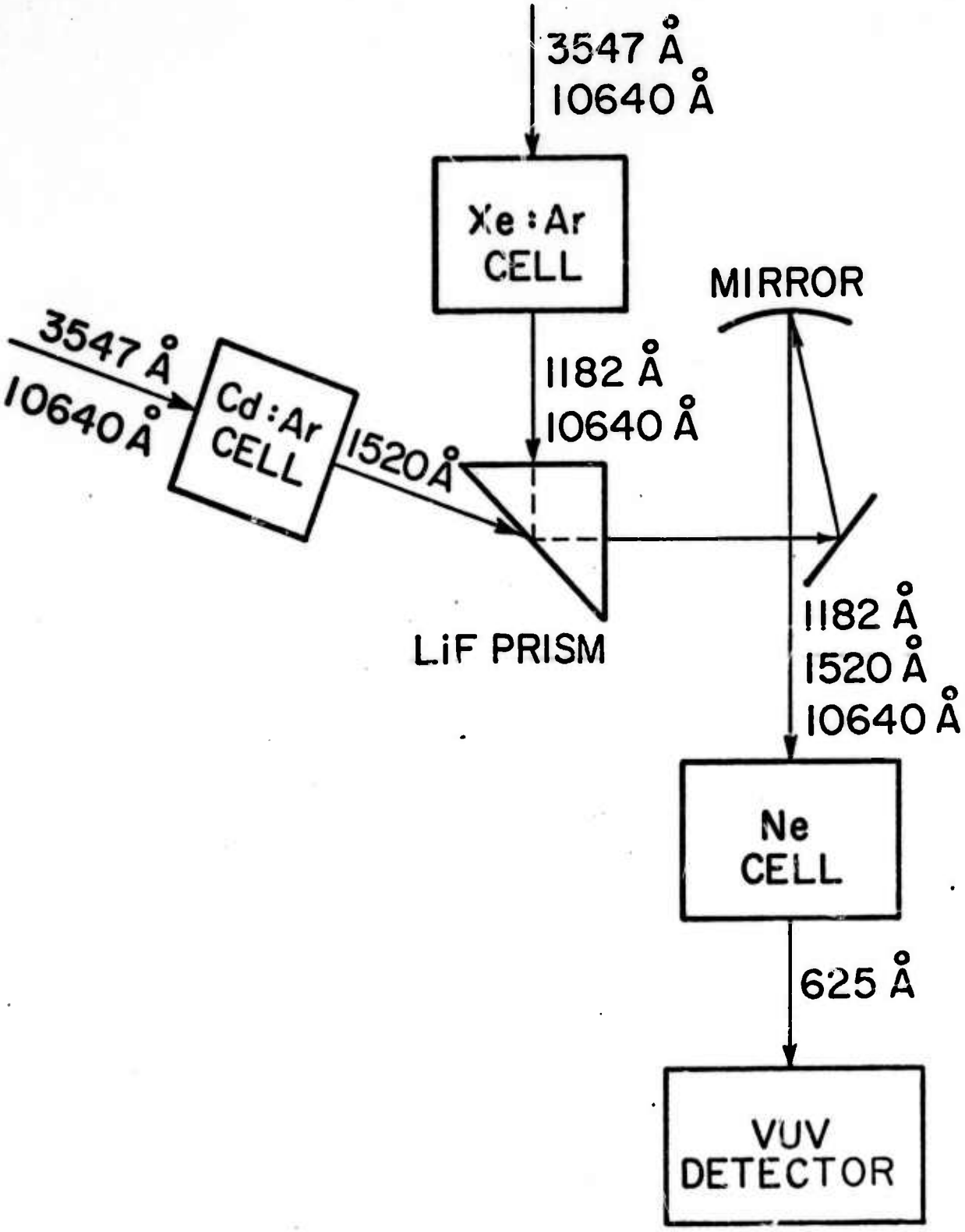

FIGURE 1

Schematic of extreme UV up-conversion in neon.

30 ps pulses of 3547 Å radiation; the two beam paths also contain residual 10,640 Å and 5320 Å light. Careful measurements of the oscillator wavelength indicate it will have to be tuned ~ 0.5 cm^{-1} in order to exactly match the Ne two-photon transition. We plan to do this by controlling the laser rod temperature; a 25° temperature change produces a ~ 1 cm^{-1} frequency shift. An internal etalon could also be used.

A Xe:Ar cell, and a Cd:Ar cell will generate 1182 Å and 1520 Å, respectively, as shown in Fig. 2. Both wavelengths are strongly absorbed in air and the remaining beam paths are in vacuum. Since no suitable dielectric coatings exist, the two beams are combined in a LiF prism using a combination of refraction (1520 Å) and total internal reflection (1182 Å and 10,640 Å). All subsequent beam steering and focusing is done with mirrors in order to eliminate chromatic aberration.

Temporal synchronization will be adjusted using two separate beam delays. A standard corner cube delay in the 1520 Å channel will be used to compensate for gross length differences between the two beam paths. However, the large group velocity dispersion between 1182 Å and 10,640 Å in the LiF combining prism will produce an unacceptable 66 ps temporal walk-off. A unique structure, invented by Mr. Hsu and detailed in Fig. 3, will compensate for this walk-off by delaying the input 10,640 Å radiation relative to 1182 Å. Both delays are located before the sum generation cells, simplifying optics requirements, and reducing losses. Using these two delays the relative timing of the three wavelengths can be arbitrarily adjusted.

Given the uncertainties of space, time, and wavelength adjustments, a preliminary diagnostic experiment that monitors these variables is essential.

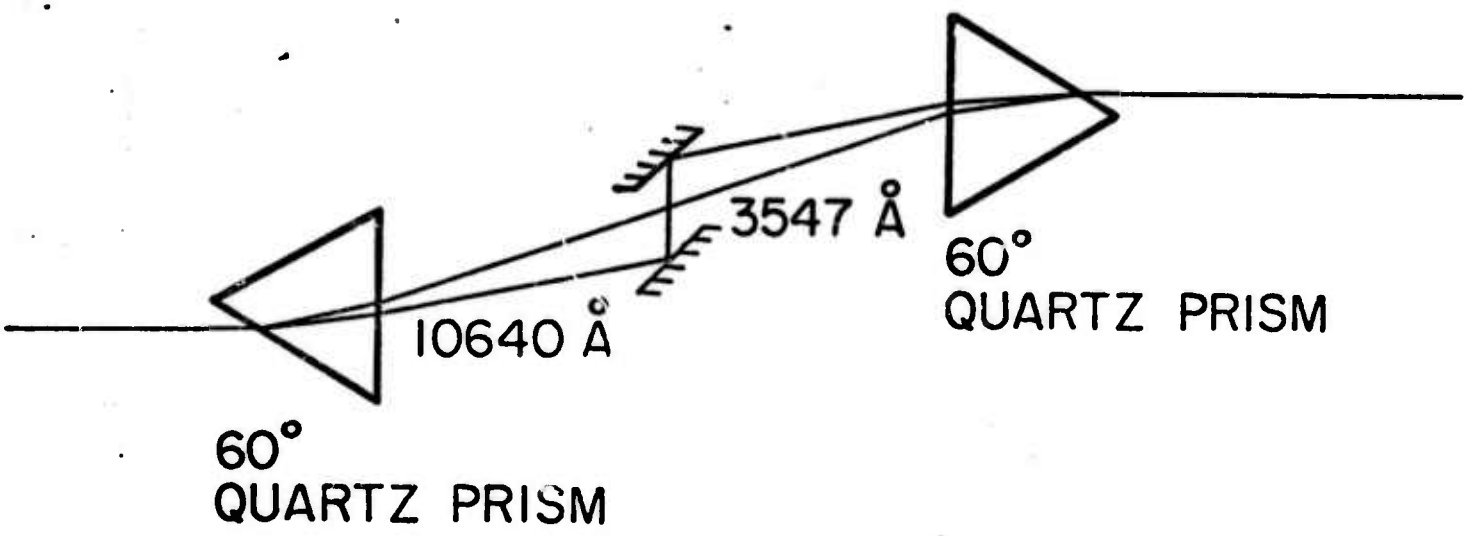

FIGURE 3

Picosecond delay of 1.064 μ relative to 3547 Å.

We believe that two-photon absorption in high pressure Ne, followed by relaxation fluorescence provides such a monitor. At 1 atm. of Ne, two-photon absorption of 1520 Å plus 1182 Å will produce a population of 2×10^8 in the Ne (3p) state, 1/3 of which will decay into the 3s state emitting 6143 Å fluorescence. The strength of this easily detectable visible radiation will be a sensitive measure of the 1520 Å and 1182 Å spatial and temporal overlap, as well as the fundamental Nd:YAG frequency tuning. Thus, using this technique, we will be able to adjust and to monitor the critical experimental parameters without relying on the 626 Å generation process itself. Note that although the two-photon absorption signal will be easily observable, the absolute absorption will be very small and will not affect the sum generation experiment.

Following the Ne generation cell, a 500 Å thick Aℓ filter will block all the input wavelengths while transmitting 1% of the generated 626 Å signal. The 626 Å radiation will strike a sodium salicitate film and a sensitive, low noise photomultiplier will measure the resulting 4200 Å fluorescence. Based on our calculated conversion efficiency and available input powers we estimate a signal-to-noise ratio of over 100 using this detection technique. A number of techniques are available to verify that an observed signal is indeed due to 626 Å radiation. The Aℓ filter is highly frequency selective; an additional filter can be inserted and the signal attenuation measured. The time or spatial overlap of the input beams can be varied, and the Nd:YAG frequency can be tuned; any of these will reduce the 626 Å signal, without affecting spurious signals caused by scattering, etc. But the most definitive verification will be the observation of the predicted phase matching

by He: no conceivable spurious effects could produce this characteristic behavior.

All major experimental components have been received or built. During the next period we will proceed with final construction, alignment, and measurements.

III. INELASTIC COLLISIONS IN THE PRESENCE OF AN ELECTROMAGNETIC FIELD

(D. B. Lidow, J. C. White, J. F. Young, and S. E. Harris)

We have observed the first laser induced inelastic collision;[1] inelastic collision processes of this type have recently been predicted by Gudzenko and Yakovlenko[2] and Harris and Lidow.[3] Details of the experiment are given in a paper entitled "Inelastic Collision Induced by Intense Optical Radiation," to be published in Physical Review Letters and attached as Appendix B.

We have studied the collision reaction:

$$Sr(5p\,^1P^0) + Ca(4s^2\,^1S) + \hbar\omega(6409\ \text{Å}) \rightarrow Sr(5s^2\,^1S) + Ca(4d\,^1D)$$

Population was first stored in the radiatively trapped $5p\,^1P^0$ level of SrI. Collisions of the excited strontium atoms with the ground state calcium atoms produced sizeable population in the calcium $4d\,^1D$ state in the presence of the 6409 Å radiation. No population transfer was observed in the absence of the 6409 Å switching signal. This process demonstrates a convenient and useful way of influencing collision reaction kinetics, and we believe it will lead to a number of exciting applications.

At an incident power density of 8.6×10^6 W/cm^2 11% of the energy stored in the strontium was transferred to the calcium, representing an induced cross

section of $\sigma_c = 3 \times 10^{-16}$ cm^2 . Our theory predicts that the induced collision cross section will increase linearly with incident 6409 Å power density until $\sigma_c \sim 8.8 \times 10^{-14}$ cm^2 . Realization of such very large cross sections in the Sr-Ca system is currently being studied.

We believe the reverse reaction may also find a number of applications. In this case population stored in one atomic species is transferred to another species with the emission of a photon. For example, the reaction

$$Mg(3p\,^1P^0) + Tl(6p\,^2P^0) \rightarrow Mg(3s^2\,^1S) + Tl(6^2P^0_{3/2}) + \hbar\omega(3368\ \text{Å})$$

might yield a high powered UV laser at 3368 Å. Assuming densities of $N_{Tl} = 3 \times 10^{18}$ cm^{-3} and $N_{Mg} = 1 \times 10^{18}$ cm^{-3} , we estimate a gain cross section of 2×10^{-19} cm^2 and a gain coefficient of $\alpha = 0.7$ cm^{-1} . Systems of this type have a unique advantage for the construction of high energy lasers: the gain cross section may be easily varied by adjusting the densities of the colliding species. These possibilities will be studied further during the next months.

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IV. SELECTIVE CHARGE EXCHANGE COLLISIONS FOR METASTABLE STORAGE

(R. W. Falcone, J. F. Young, and S. E. Harris)

Thermal velocity charge exchange collisions have been studied as a method of selectively exciting strongly forbidden levels of singly ionized column IIA elements (Ca^+, Sr^+, Ba^+) leaving the ground state empty. Thus a large inversion to ground should result and the inverted system could be used as a stimulated anti-Stokes laser or as a two-photon laser. We have successfully demonstrated charge exchange collisional excitation of the $Sr^+(4d^2D)$ metastable level. However, low population densities have prevented us from firmly establishing the selective nature of the process; we have no direct evidence that an inversion has been produced.

We have considered the charge exchange reaction

$$Mg^+ + Sr \rightarrow Mg + Sr^{+*}(4d^2D) + \Delta E$$

where $\Delta E = 1182\ cm^{-1}$ (see Fig. 1). Recent theoretical and experimental work[1] shows that thermal velocity charge exchange reactions have maximum cross sections when the energy defect is on the order of 0.1 eV. Based on this we expect the Mg^+-Sr collision to have a large cross section (approximately $10^{-15}\ cm^2$) for selective excitation of the metastable $4d^2D$ level in ionic strontium.

CHARGE EXCHANGE
STIMULATED ANTISTOKES LASER

$$Mg^{+} + Sr = Mg + Sr^{+*} + \Delta E$$

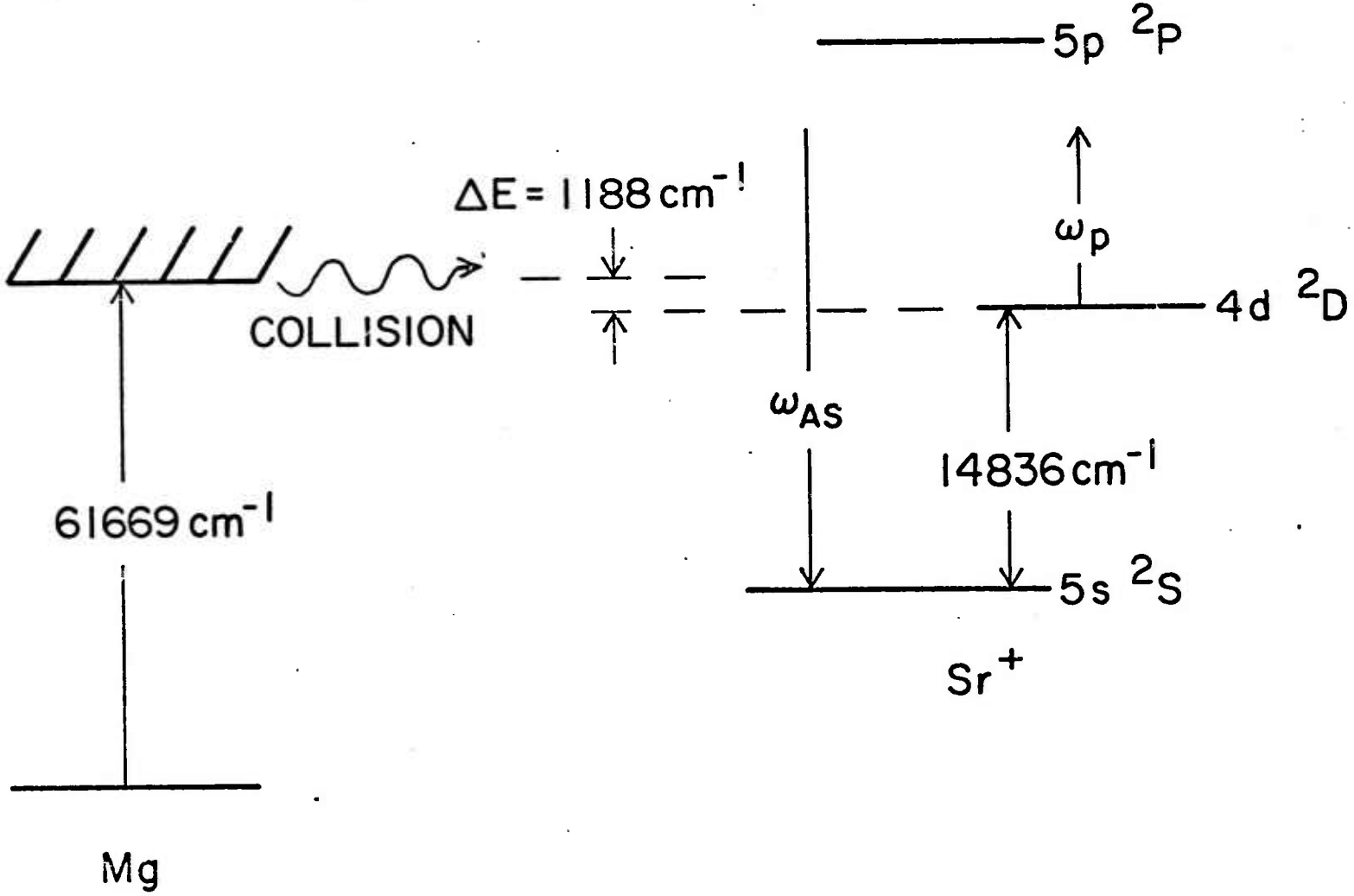

FIGURE 1

The reaction has been studied experimentally using a heat pipe type cell containing Mg and Sr vapor at densities of about 10^{16} atoms/cc. The Mg atoms are selectively ionized using two different processes. In the first process resonance radiation at 2852 Å creates a high density of excited Mg atoms which are quickly ionized by a second photon of the same wavelength. While this process seems to create large numbers of ions, the exact density is unknown since the cross section for this process has not been measured. A second process uses a recently identified autoionization level in Mg having the same parity as the ground state. Two photons of 2929 Å coincide with this level and they create a well defined volume of Mg^+ at a density of about 5×10^{13} ions/cc.

The metastable level in Sr^+ which is excited by charge exchange collisions with Mg^+ is detected by tuning a 1 μ laser to the $4d^2D - 5p^2P$ resonance in Sr^+ and observing fluorescence to the ground state. Strontium ion densities of about 10^{13} ions/cc have been observed.

Demonstrating the selective nature of the Mg^+-Sr collision remains a major goal of this project. We hope to show the selectivity of the process by creating high metastable densities, transferring them to the $5p^2P$ level by absorption of 1 μ radiation, and thus creating measurable laser gain. The magnitude of the gain is a direct measure of the degree of inversion and therefore selectivity. Currently we are restricted to working with a $Sr^+(4d^2D)$ density of about 10^{13} ions/cc because of an unidentified noise source populating the $Sr^+(5p^2P)$ level in the presence of Mg^+ ions at higher densities. Although this noise source appears to be collisional excitation of the $Sr^+(5p^2P)$ due to highly excited neutral atoms

formed through recombination of Mg^+ ions, the exact source of the noise has yet to be determined and the ultimate ion density limit has not been established. Electron cooling, three-body recombination rates and electron excitation to higher levels will probably limit the metastable storage in high density Sr^+ to 10^{17} ions/cc.

The next stage of the experiment will be to understand this "noise" source (which destroys the inversion ionic levels), to determine the $Sr^+(4d^2D)$ density limit, and to measure laser gain.

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V. PUBLICATIONS

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APPENDIX A

"1202.8 Å Generation in Hg Using a Parametrically Amplified Dye Laser"

IEEE J. Quant. Elect. QE-12, 60 (January 1976)

1202.8-Å Generation in Hg Using a Parametrically Amplified Dye Laser

K. S. HSU, A. H. KUNG, L. J. ZYCH, J. F. YOUNG, AND S. E. HARRIS

Abstract—We have generated 1202.8-A radiation, one-half the frequency of the $1s^1S_0$-$2s^1S_0$ forbidden transition in He, using resonantly enhanced four-wave optical mixing in Hg vapor. Peak powers of 300 W in a ~ 15-ps pulse have been achieved using a tunable narrow-band source consisting of a parametrically amplified commercial dye laser. The dependence of the conversion efficiency on the spectral properties of this source is described. Hg is shown to be negatively dispersive for this process, and thus phase matchable.

We report the generation of 1202.8-Å radiation by resonantly enhanced four-wave optical mixing [1]-[3] in Hg vapor. 1202.8-Å radiation is particularly interesting because it is the half frequency of the $1s^1S_0$-$2s^1S_0$ forbidden transition in He. Such a source could be used for the creation of high densities of metastable $2s^1S_0$ He by two-photon absorption, as well as for the generation of 400-Å radiation using resonantly enhanced harmonic generation in He. A peak power of 300 W in a ~15-ps pulse has been obtained at 1202.8 Å. Resonant optical mixing in Hg is achieved using a tunable picosecond-duration narrow-band source consisting of a commercial flashlamp pumped dye laser parametrically amplified using the frequency quadrupled output of a passively mode-locked Nd:YAG laser. The amplified tunable pulse is then frequency doubled and mixed with the output from a parametric generator-amplifier [4] to yield 1202.8-Å radiation.

Following the first demonstration of harmonic generation of UV and VUV radiation in metal vapors [5], Hodgson *et al.* [1] and Bloom *et al.* [3] demonstrated the advantages of using tunable sources to obtain resonant enhancement of the mixing process. The degree of enhancement achieved depends critically on the linewidth and stability of the source used. In addition, the efficiency of the mixing process depends on the ability to accurately control and adjust the interacting beams with respect to both time and space. In all of these respects, a source consisting of a parametrically amplified, stable, well-controlled laser has a number of attractive advantages, as recently described by Massey *et al.* [6], [7].

A schematic of our experimental arrangement is shown in Fig. 1. The output of a passively mode-locked Nd:YAG oscillator amplifier chain is frequency quadrupled to provide single mode-locked pulses of high-power 2660-Å radiation at 3 pulses per second. In addition to the usual system components, an electrooptic Pockel's cell is used in the oscillator cavity to determine a fixed time for the start of the laser buildup from noise. We found that by opening the Pockel's cell about 200 μs after triggering the flashlamp, the normal temporal jitter of the laser is reduced from $\pm$ 10 μs to less than $\pm$ 0.25 μs. When operating in this fashion the quality of the mode locking is not noticeably affected. It is possible, however, to destroy the mode locking by delaying the shutter opening too long.

The low temporal jitter of the 2660-Å pulse permits us to synchronize it reliably to a commercial (Chromatix CMX-4) flashlamp pumped dye laser of 1-μs pulsewidth. The bandwidth of the laser is narrowed to less than 1 cm^{-1} with an in-

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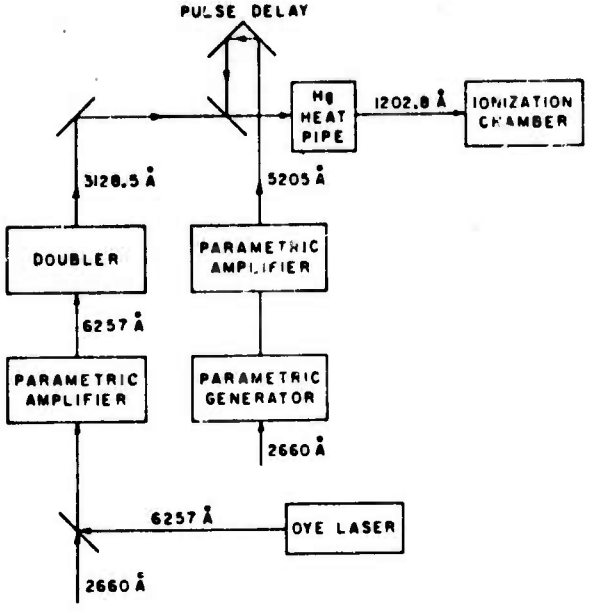

Fig. 1.

ternal etalon. A small portion of the dye laser output, ~ 100 W, and the 2660-Å pulse are incident collinearly into a parametric amplifier consisting of a 5 mm × 5 mm × 50 mm long $NH_4H_2PO_4$ crystal in a temperature controlled oven. For these experiments the laser is tuned to 6257 Å, four times the wavelength of the $6s^1S_0$–$7s^1S_0$ transition of Hg, and the temperature of the crystal oven is adjusted for maximum gain at this wavelength. For a power of 20 MW at 2660 Å focused to a density of 200 MW/cm^2, the theoretical small signal gain is 10^5. We measured peak output powers in excess of 1 MW each for the signal and idler waves, representing ~ 10-percent power conversion of the pump into tunable radiation.

The amplified output is frequency doubled in an angle-tuned crystal of KH_2PO_4 to 3128.5 Å and combined spatially and temporally with a second tunable, synchronized, picosecond pulse from a parametric generator-amplifier [4]. When tuned to 5205 Å (such that the sum wavelength is 1208.8 Å) the bandwidth of this source is ~ 30 Å due to its proximity to degeneracy of the parametric generator. However, because the efficiency of the mixing process is not affected by the spectral width of this (nonresonant) source, this somewhat simpler, more conventional approach is adequate. The collinear beams are focused at the center of a 12-cm-long stainless-steel Hg heat pipe cell. The VUV output signal passes through LiF windows into an ionization chamber used for energy measurements. The space between the Hg cell and the ionization chamber is purged with Ar to avoid absorption of the VUV signal by oxygen. The ionization chamber is filled with research grade p-xylene at 0.1 torr and is sensitive from the short wavelength cutoff of the LiF window at 1150 Å to 1400 Å, the long wavelength limit of the photoionization of p-xylene. We found the quantum yield at 1202.8 Å to be 0.275 based on an absolute calibration using a CW hydrogen discharge lamp through a McPherson 225 spectrometer and a calibrated CsSb photodiode.

For incident power densities of 6 GW/cm^2 at 3128.5 Å, 650 MW/cm^2 at 5205 Å, and an Hg pressure of 20 torr, we measured an output energy of 4.5 nJ, or 400-W peak in an estimated 15-ps-long pulse. This corresponds to a conversion efficiency of 0.5 percent from 5205 Å to 1202.8 Å in the overlapped beam areas. As expected, the output is very sensitive to the dye laser bandwidth and tuning. Tunings of a few angstroms off resonance results in signal decreases of greater than 100. Similarly, increasing the bandwidth from less than 1 cm^{-1} to 3 cm^{-1} by removal of the etalon, decreases the signal by about 10. This is in good qualitative agreement with the results of Stappaerts *et al.* [8]. We did not measure the bandwidth of the 1202.8-Å output, but based on the input bandwidths we expect it to be ~ 16 Å. A second parametrically amplified dye laser for the 5205-Å wavelength would probably be required to reduce the bandwidth to acceptable values for subsequent resonantly enhanced harmonic generation stages.

In these experiments the input radiation was tightly focused such that the effective focal region was short compared to the length of the nonlinear media. In this case the amplitude of the sum wavelength as a function of wave vector mismatch is no longer described by the familiar $\sin x/x$ function associated with plane-wave conditions [9]. Using the theory of [9] it is possible to determine the sign of the mismatch by adding a gas with known dispersion such as Ar. As Ar was introduced into the Hg cell the signal decreased monotonically by two orders of magnitude to below noise level. Since the signal from a positively dispersive vapor would have exhibited an oscillating signal amplitude with comparable peak heights as the Ar pressure was increased, while that from a negatively dispersive media would show no such oscillation, this indicates that Hg is negatively dispersive for the process. Thus the conversion efficiency could be increased using higher power sources to maintain the power density over a longer length and phase matching the process by adding the proper ratio of a positively dispersive vapor.

In summary, we have demonstrated the use of parametric amplification of a flashlamp pumped dye laser to obtain tunable, picosecond duration, high-power visible pulses. We have demonstrated the application of these pulses for efficient resonantly enhanced sum generation into the VUV region at 1202.8 Å. By tuning the second source from 4000 Å to 7000 Å, it should be possible to obtain tunable signals from 1125 Å to 1280 Å. This falls within the gain profiles of Ar excimer and hydrogen amplifiers, thus creating the possibility of sources of very high brightness in this region of the spectrum.

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ACKNOWLEDGMENT

The authors wish to thank B. Yoshizumi for his expert technical assistance.

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APPENDIX B

"Inelastic Collision Induced by Intense Optical Radiation"

Phys. Rev. Lett. 36, 462 (March 1976)

INELASTIC COLLISION INDUCED BY INTENSE OPTICAL RADIATION*

by

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ABSTRACT

A large cross section for inelastic collision is induced by an incident laser tuned to the frequency of the inter-atomic energy defect. We study energy transfer from the Sr ($5p\,^1P^0$) level to the Ca ($4d\,^1D$) level and measure a cross section for inelastic collision of 3×10^{-16} cm^2 at a laser power density of 8.0×10^6 W/cm^2 and a wavelength of 4409.0 Å.

INELASTIC COLLISION INDUCED BY INTENSE OPTICAL RADIATION

by

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The cross section for inelastic collision between atoms is infinitesimally small if the energy defect ΔE is large with respect to kT. In this Letter we report the first observation of a process where a large cross section for inelastic collision is created by applying optical radiation at a frequency $\omega = \Delta E$. Energy transfer is initiated or "switched" by the presence of the optical radiation. Inelastic collision processes of this type have recently been predicted by Gudzenko and Yakovlenko[1] and by Harris and Lidow.[2]

We have observed this process in the system (Fig. 1):

$$Sr(5p\,^1P^0) + Ca(4s^2\,^1S) + h\nu(\text{about } 8\,\text{Å}) \rightarrow Sr(5s^2\,^1S) + Ca(4d\,^1D) \qquad (1)$$

Energy was first stored in the radiatively trapped $5p\,^1P^0$ level of Sr I. This level was populated by two photon pumping of the $5d\,^1D$ Sr level, followed by radiative decay. Inelastic collision to the $4d\,^1D$ level of Ca I was produced by a second laser beam at about 8 Å.

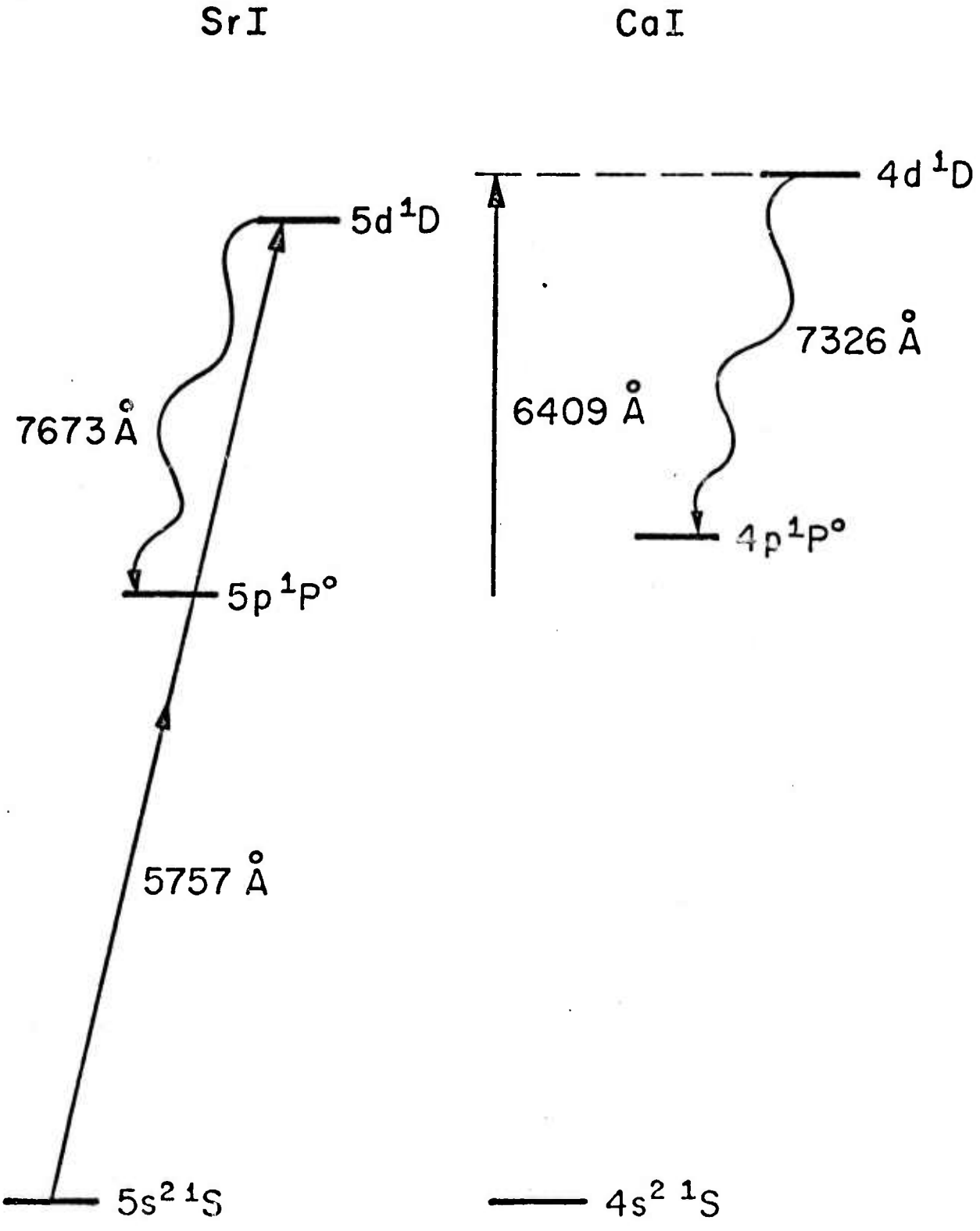

FIG. 1--Energy level diagram for Sr-Ca induced collision experiment.

During collision of an excited Sr ($5p\,^1P^o$) atom and a ground state Ca ($4s^2\,^1S$) atom the strong dipole-dipole coupling of the (5p-5s) Sr transition and the (4p-4s) Ca transition causes a virtual transition of the Ca atom. Absorption of a 6409 Å photon completes the Ca excitation. The process may thus be viewed as a virtual collisional excitation, followed by a real absorption.[2] Alternately, and for the case described here equivalently, the process can be viewed as a free-free photon absorption of the Sr-Ca quasi-molecule.[1] From either viewpoint, for dipole-dipole coupling, maximum collision cross section is predicted when $\hbar\omega$ is equal to the inter-atomic transition frequency of the infinitely separated atoms ($R = \infty$). For incident radiation at the $R = \infty$ frequency, the predicted cross section for collision σ_c is given by[2]

$$\sigma_c = \frac{4}{3}\left(\frac{\pi}{\hbar}\right)^2\left[\frac{\mu_{Sr}\mu_{Ca}\mu_{Ca^*}E}{\bar{V}\hbar\omega_0}\right]^2 \qquad (2)$$

where μ_{Sr}, μ_{Ca}, and μ_{Ca^*} are the magnitudes of the dipole matrix elements of the Sr (5s-5p), Ca (4s-4p), and Ca (4p-4d) transitions respectively; E is the strength of the applied optical field at 6409 Å; $\hbar\Delta\omega$ is the energy difference between the Sr ($5p\,^1P^o$) and Ca ($4p\,^1P^o$) levels; and $\bar{V}$ is the average velocity. The quantity ρ_0 is the minimum impact parameter such that the integrated relative phase shift during transit is 1 radian, i.e.,

$$\frac{1}{\hbar}\int_{-\infty}^{+\infty}\frac{C_6}{R^6(t)}\,dt = \frac{3\pi\mu_{Sr}^2\mu_{Ca}^2}{4\hbar^2\bar{V}\rho_0^5} = 1$$

where C_6 is the energy shift constant of the Sr $(5p\,^1P^0)$ level. We neglect the comparatively small shift of the Ca $(4d\,^1D)$ level. Equation (2) includes only the dominant path of the perturbation calculation. For our experiments, $\mu_{Sr} = 3.0$ a.u. , $\mu_{Ca} = 2.8$ a.u. , $\mu_{Ca^*} = 0.95$ a.u. ,[3] $\Delta\nu_0 = 1954$ cm^{-1} , $\bar{V} = 6.6 \times 10^4$ cm/sec , $\rho_0 = 16.7$ Å , and $\sigma_c = 2.9 \times 10^{-23} \frac{P}{A}$ (W/cm^2) cm^2 .

A schematic of the experimental apparatus is shown in Fig. 2. The Sr-Ca heat pipe type cell was operated at a temperature of approximately 875°C and a zone length of 2 cm. The number density of the Sr and Ca ground state atoms were determined by measuring the linewidth of their resonance absorption lines at 4608 Å and 4228 Å. These were $N_{Sr} = 8.0 \times 10^{16}$ atoms/cm^3 and $N_{Ca} = 3.8 \times 10^{16}$ atoms/cm^3 .

A Chromatix CMX-4 flashlamp pumped dye laser produced the 5757 Å radiation for two photon pumping of the Sr $(5d\,^1D)$ level. An incident power of about 1 kW was focussed to a power density of 1.4×10^7 W/cm^2. Emitted radiation on the (5d-5p) Sr transition was measured and used to estimate the population of the radiatively trapped Sr $(5p\,^1P^0)$ level. Typically this density was about $N_{Sr}(5p\,^1P^0) \approx 6 \times 10^{15}$ atoms/cm^3 . Two photon pumping and radiative decay was used as the populating mechanism of the Sr $(5p\,^1P^0)$ storage level in order to avoid the necessity of directly applying radiation at the (5s-5p) Sr frequency and thus possibly masking the desired collision process by a two photon sum process to the Ca $(4d\,^1D)$ level.

Tunable radiation for inducing the collision process was provided by a Chromatix Model 1050 dye laser (kiton red) pumped with the second harmonic of the 1.1? μ line of a Q-switched Nd:YAG laser. A peak power of 175 W in a 150 nsec pulse was focussed to a power density of about 8.6×10^6 W/cm^2

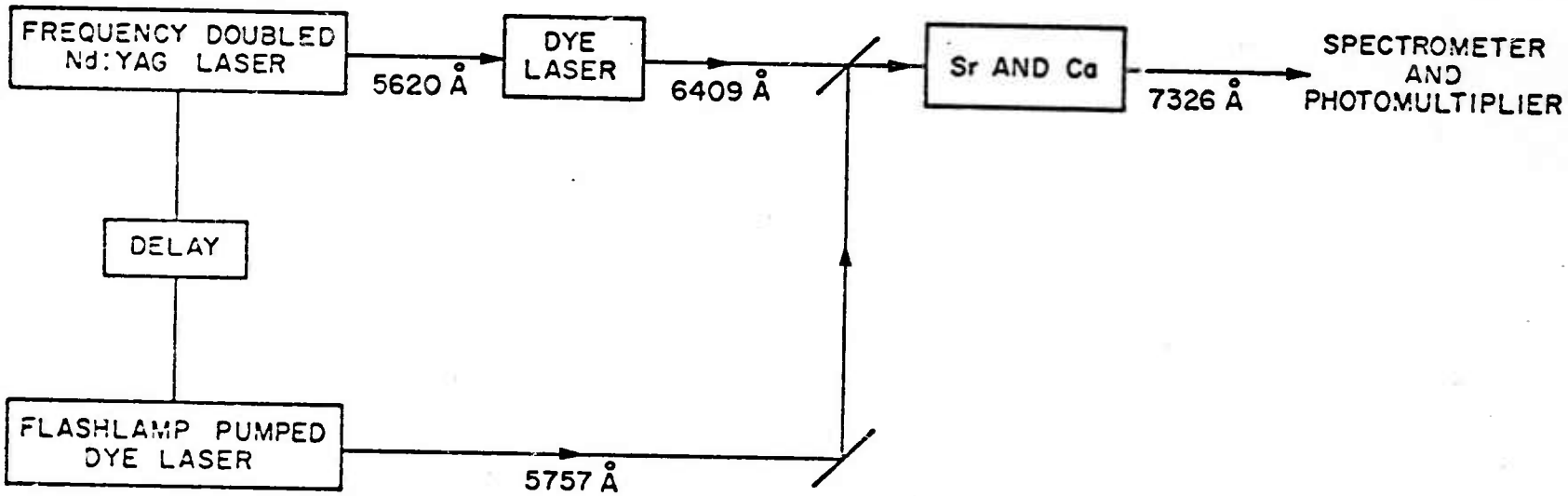

FIG. 2--Schematic of experimental apparatus.

and spatially overlapped with the 5757 Å pump beam. The relative timing of the two lasers could be adjusted over a range of several microseconds using a variable delay. Fluorescence of the Ca ($4d$-$4p$) transition was measured and used to monitor the population of the Ca ($4d\,^1D$) level.

The 5757 Å laser was first tuned to maximize the Sr 7673 Å radiation and thus the population of the Sr ($5p\,^1P^0$) level. With both lasers on, a maximum signal was measured with the transfer laser tuned to 6409.0 Å, as compared to the predicted ($R = \infty$) value of 6408.6 Å. (All wavelengths are given in air.) This is within the ± 0.7 Å uncertainty of our wavelength calibration. The half-power linewidth for transfer was 1.0 ± 0.2 Å. The linewidth of the Ca 7326 Å fluorescence signal was resolution limited. The 5757 Å laser, by itself, produced some 7326 Å fluorescence. A signal-to-noise ratio of about 50 was obtained by integrating over five pulses.

The ratio of the fluorescence output at the 7326 Å Ca line to that of the 7673 Å Sr line was 1:36. Allowing for the somewhat tighter focussing (4×4) of the 6409 Å laser, as compared to the 5757 Å laser, this fluorescence ratio indicates a collisional transfer of 11% of the excited Sr ($5p\,^1P^0$) atoms to the Ca ($4d\,^1D$) level. This transfer takes place during the 150 nsec pulsewidth of the 6409 Å laser, indicating an induced cross section of 3×10^{-16} cm^2. This cross section was linear in the incident power density. We estimate an experimental uncertainty of a factor of 3. For our power density of 8.6×10^6 W/cm^2, Eq. (2) predicts a cross section of 2.5×10^{-16} cm^2. The 6409 Å transfer pulse could be delayed by about 0.5 μsec after the end of the 5757 Å pulse before the 7326 Å signal was significantly reduced. This is consistent with the estimated 0.5 μsec radiative trapping time of the Sr ($5p\,^1P^0$) level.

We note that there are several Sr ($5p\ ^1P$ levels a few Å from coincidence with the Ca ($4d\ ^1D$ level. By tuning the transfer laser exactly to these levels, we ascertained that no direct collisional transfer was taking place.

Theory predicts that the optically induced collision cross section should continue to increase linearly with power density until $\sigma_c = \pi\rho_0^2$ and as the square root of power density thereafter. For the Sr-Ca system studied here $\pi\rho_0^2 = 8.8 \times 10^{-14}$ cm^2. Large energy transfer rates should thus be possible using this process.

The collisional process demonstrated here should be applicable to the direct measurement of inter-atomic potentials. Additional transfer peaks are expected at frequencies where the difference of the atomic potentials has zero slope, and at frequencies corresponding to turning points of the classical motion. Application to the construction of short wavelength lasers is likely. Energy could be stored in a metastable atomic level, and using a short tunable optical pulse, collisionally switched to a radiative state of a second specie. The inverse radiative process where lasing takes place between an excited state of one specie and a lower state of a second specie may be applicable to the construction of low gain, high energy lasers.

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