



Hartmut Gruhl was born in Gnaschwitz, East Germany, on December 23, 1954. He received the Vordiplom degree from the Georg-August-Universität, Göttingen, West Germany, in 1977 and the Diplom degree in physics from the Ludwig-Maximilians-Universität, München, West Germany, in 1981.

His diploma work (upon which this paper is based) was carried out at the Max-Planck-Institut für Quantenoptik, Garching, West Germany. He is currently working towards the Ph.D. degree at the University of Göttingen.



Richard Sigel was born in Heilbronn am Neckar, West Germany, on October 22, 1939. He studied physics at the Universities of Tübingen and München and received the Ph.D. degree from the Technical University, München, West Germany, in 1969.

He is Group Leader for laser-plasma interaction at the Max-Planck-Institut für Quantenoptik, Garching, West Germany. Besides laser-plasma interaction, his research interests include hydrodynamics, high-speed photography, lasers, and nonlinear optics.

Continuously Tunable Sum-Frequency Generation Involving Sodium Rydberg States

ROBERT W. BOYD, DANIEL J. GAUTHIER, JERZY KRASINSKI, AND MICHELLE S. MALCUIT

Abstract—Broadly tunable sum-frequency generation has been observed in a vapor of atomic sodium in the presence of a dc electric field. This field induces a $\chi^{(2)}$ nonlinearity which is resonantly enhanced when the sum frequency corresponds to the energy separation between the ground state and an atomic Rydberg state. In a vapor of number density $4 \times 10^{14} \text{ cm}^{-3}$, we obtain an energy conversion efficiency as large as 3×10^{-4} and a $\chi^{(2)}$ as large as 1.2×10^{-8} ESU. We have also observed sum-frequency generation in the absence of an applied dc field, and we relate these observations to mechanisms that have been proposed to explain this effect.

INTRODUCTION

THERE is currently great interest in sum-frequency generation as a means of producing tunable ultraviolet radiation [1]–[10]. Most of the mixing processes that have been demonstrated thus far make use of a third-order or $\chi^{(3)}$ optical nonlinearity, in which case three input fields combine to produce the output frequency. To avoid the inconvenience of using three separate laser sources, such mixing experiments usually derive at least two of the input fields from a single laser. In such cases, at least one of the intermediate steps of the mixing process is usually nonresonant, which limits the efficiency with which the ultraviolet frequency can be produced.

In this paper, we describe a nonlinear mixing process utilizing two input lasers in which each intermediate step is resonantly enhanced [11], [12]. As shown in Fig. 1, an incident laser field at frequency ω_1 connects the sodium $3^2S_{1/2}$ ground

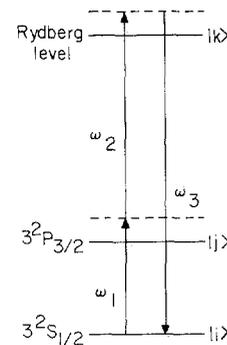


Fig. 1. Optical waves at frequencies ω_1 , ω_2 , and $\omega_3 = \omega_1 + \omega_2$ interact by means of the nonlinear response of an atom with energy eigenstates $|i\rangle$, $|j\rangle$, and $|k\rangle$. In the experimental work, these states correspond to the $3^2S_{1/2}$, $3^2P_{3/2}$, and a Rydberg level, respectively.

state to the $3^2P_{3/2}$ excited state while a second laser field at frequency ω_2 completes a two-photon resonance with a sodium Rydberg level. In addition, a dc electric field is applied to the atomic system. This field breaks the inversion symmetry of the sodium atom, and thus allows the existence of an electric dipole moment oscillating at the sum frequency $\omega_3 = \omega_1 + \omega_2$ [13]. The resulting nonlinear polarization is related to the applied field strengths $E(\omega_1)$ and $E(\omega_2)$ through the relation

$$P(\omega_3) = 2\chi^{(2)}(\omega_3 = \omega_1 + \omega_2)E(\omega_1)E(\omega_2) \quad (1)$$

where the nonlinear susceptibility describing the mixing process is given by [11]

$$\chi^{(2)}(\omega_3 = \omega_1 + \omega_2) = \sum_{jk} \frac{\mu_{ik}\mu_k\mu_{ji}}{2\hbar^2(\omega_{ki} - \omega_2 - \omega_1)(\omega_{ji} - \omega_1)} \quad (2)$$

Manuscript received January 16, 1984; revised April 24, 1984. This work was supported by the U.S. Army Research Office and the Air Force Office of Scientific Research.

R. W. Boyd, D. J. Gauthier, and M. S. Malcuit are with the Institute of Optics, University of Rochester, Rochester, NY 14627.

J. Krasinski is with Allied Corporation, Mt. Bethel, NJ 07060.

Here N denotes the number density of atoms, and μ_{lm} denotes the electric dipole matrix element connecting levels l and m . In these equations we have suppressed the tensor aspect of the nonlinear susceptibility since in our experiment all of the electric fields were polarized in the same direction. In the absence of the dc electric field, at least one of the three matrix elements appearing in (2) would vanish as a consequence of the parity selection rule for electric dipole transitions. However, dc field strengths of the order of 1 kV/cm are sufficient to mix nearly completely opposite-parity sodium Rydberg states with $n \geq 10$ [14], thus inducing a large nonlinear susceptibility [15]. To good approximation, the wave functions describing the sodium Rydberg states can be considered hydrogenic in character. Within the hydrogenic approximation, closed form expressions can be derived for the electric-field induced dipole moments that appear in (2) [11]. An approximate value of the nonlinear susceptibility can be obtained from these expressions. Let us consider an atomic vapor of density $4 \times 10^{14} \text{ cm}^{-3}$ with ω_1 detuned 1 cm^{-1} from the intermediate resonance and $\omega_1 + \omega_2$ detuned 5 GHz (i.e., several linewidths) from the Rydberg state resonance. Under these conditions, the numerical example given in [11] predicts a value of $\chi^{(2)}$ as large as 8×10^{-8} ESU.

In addition to modifying the electric dipole selection rules, the dc electric field results in a splitting and shifting of the Rydberg levels through the Stark effect. This phenomenon leads to regions of broad tunability of the sum-frequency generation process. The positions of the Stark-shifted Rydberg levels can be predicted by diagonalizing the Hamiltonian matrix describing the sodium Rydberg states in the presence of a dc electric field. Such a calculation has been performed by Zimmerman *et al.* [16]; the results for energies near that of the $n = 15$ manifold are shown in Fig. 2. For field strengths $< 1500 \text{ V/cm}$, the $l \geq 2$, $m = 0$ sublevels display a linear Stark effect and fan out from these zero-field positions. At higher field strengths, the various levels are strongly mixed and cover nearly completely the region between successive values of the principal quantum number.

EXPERIMENTAL SETUP

The setup used in our experimental study is shown in Fig. 3. An excimer laser (Lumonics Model 861S-2) operating at 308 nm with XeCl was used to pump two dye lasers. Each laser consisted of a two-grating Littman-style [17] oscillator followed by a power amplifier. One laser (hereafter called the yellow laser) used rhodamine 6G as the laser dye and was tuned to the vicinity of the $D_2(3^2S_{1/2}-3^2P_{3/2})$ resonance line. The other laser (the blue laser) used stilbene 1, stilbene 3, POPOP, or DPS as the laser dye and could be tuned over the range 3900–4300 Å to access all Rydberg levels with $n > 11$. These lasers produced output energies of typically 0.5 mJ with linewidths of $\sim 5 \text{ GHz}$ and durations of $\sim 4 \text{ ns}$. The output beams from these lasers were combined using a dichroic beam splitter and were loosely focused to obtain a beam diameter of several millimeters inside the sodium vapor cell. This cell presented an interaction length of 25 mm and was constructed of commercially available stainless steel vacuum fittings. The cell windows are 25 mm diameter sapphire

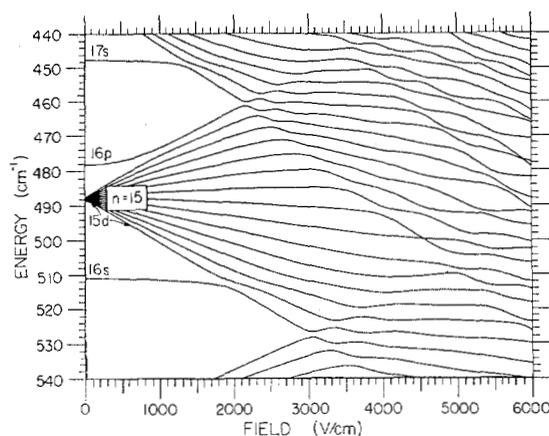


Fig. 2. Energy level structure of the $m = 0$ Rydberg levels of the sodium atom in the presence of a dc electric field. (Reproduced with permission from M. L. Zimmerman, M. G. Littman, M. M. Kash, and D. Kleppner, *Phys. Rev. A*, vol. 20, p. 2251, 1979.)

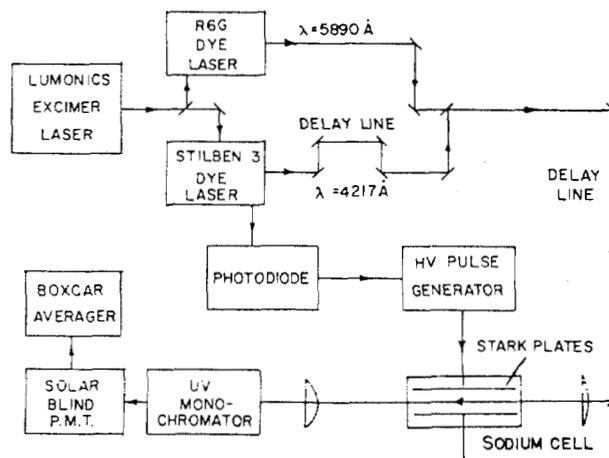


Fig. 3. Experimental setup.

flats permanently attached to standard vacuum flanges. Plane parallel Stark plates were mounted inside the cell at a separation of 5 mm. A stainless steel sidearm, high voltage feed-through, and pump-out line were welded to the body of the cell. The pump-out line consists of a 1 m long stainless steel capillary tube that is spiraled about the body of the cell. This design prevents the rapid diffusion of sodium out of the interaction region. Typically, the cell was operated with no buffer gas and with the cell body at a temperature of 410°C and the sidearm at a temperature of 330°C , thus producing a sodium number density of $4 \times 10^{14} \text{ cm}^{-3}$.

High voltage pulses were applied to the Stark plates simultaneously with the arrival of the laser pulses. The high voltage pulses were produced by an avalanche transition chain (2N5551) that was triggered by a portion of the laser output beam using a fast photodiode. The risetime of the electrical pulse was $\sim 1 \text{ ns}$, and a short pulse duration of $\sim 10 \text{ ns}$ was used to avoid establishing a breakdown in the sodium vapor.

The beam exiting the sodium cell was focused onto the entrance slit of a quartz-prism monochromator. Additional spectral discrimination was provided by UV-transmitting interference filters. The ultraviolet signal was detected using a solar blind photomultiplier tube (Hamamatsu R166 UH) and was processed using a boxcar averager.

EXPERIMENTAL RESULTS

Resonantly enhanced sum-frequency generation is found to occur whenever the yellow laser is tuned near (to within ~ 10 Å of) the sodium D lines and the blue laser is tuned to complete a two-photon resonance with a Rydberg state. The UV signal is emitted in the forward direction in a beam whose divergence angle is comparable to that of the incident lasers (several milliradians). The magnitude of the UV signal has been found to depend critically on the temporal overlap of the laser and high voltage pulses [12]. Furthermore, the pulse length of the sum frequency signal is found to be comparable to that of the incident lasers to within the 6 ns risetime of our photomultiplier. We have also verified that the wavelength of the UV signal tunes as the wavelength of the blue laser is varied. These observations indicate that a true parametric process is occurring and rule out the possibility that the observed signal is due to fluorescence excited by the lasers or by a high voltage discharge.

We have conducted a systematic study of the tuning characteristics of the sum-frequency generation process. A typical tuning curve is shown in Fig. 4. Here the intensity of the UV output signal is plotted as a function of the wavelength of the blue laser, with the yellow laser detuned 1.1 Å to the short wavelength side of the D_2 resonance line. Resonance enhancement of the output signal is observed whenever the sum frequency is coincident with a Stark-split resonance of the sodium atom. The $l \geq 2$ levels are seen to mix and spread out in a fan as predicted by the theoretical curves shown in Fig. 2.

A series of high resolution scans through the region near the $12p$ level is shown in Fig. 5 for several different values of the dc field strength. Here the yellow laser is tuned close to the D_2 resonance line. The centroid of the resonance is seen to shift to higher energies with increasing dc field strength, in agreement with the theoretically predicted behavior shown in Fig. 2. Especially at the lower field strength, a pronounced dip is observed in the tuning curve. This dip is presumed to be a phase matching effect and not due to absorption of the generated UV radiation because the calculated value of the line center absorption coefficient based on the tabulated value [18] of the oscillator strength of the $3s \rightarrow 12p$ transition is only 0.02 cm^{-1} .

Scans covering a much broader spectral interval are shown in Fig. 6 for several different values of the dc field strength. In these scans a large portion of the Rydberg series of the sodium atom is clearly visible. In the absence of the dc electric field, narrow resonances are observed whenever the blue laser wavelength has a value such that $\omega_1 + \omega_2$ is resonant with a Rydberg s or d state. The existence of sum-frequency generation in atomic vapors in the absence of applied fields is well documented [19]–[25], although the origin of this effect is not completely understood. Several different mechanisms have been proposed to explain the origin of this effect, and it is likely that any of these mechanisms can become important in specific cases. Our results are most consistent with the mechanism proposed by Mossberg *et al.* [22] and Miyazaki *et al.* [23] which assumes that the second-order nonlinearity is induced by an electric field generated by photo-ionization of

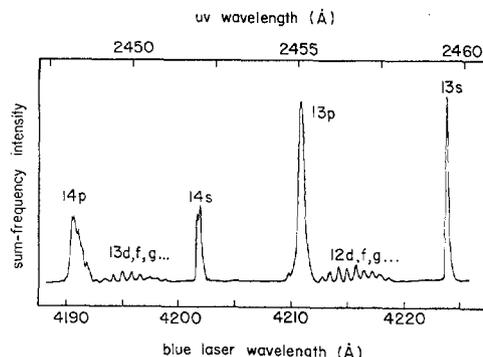


Fig. 4. The measured intensity of the sum-frequency signal is shown as a function of the wavelength of the blue laser. The wavelength of emitted UV radiation is shown on the upper abscissa. The yellow laser was detuned 1.1 Å to the high frequency side of the D_2 resonance line and the dc electric field strength was 1200 V/cm.

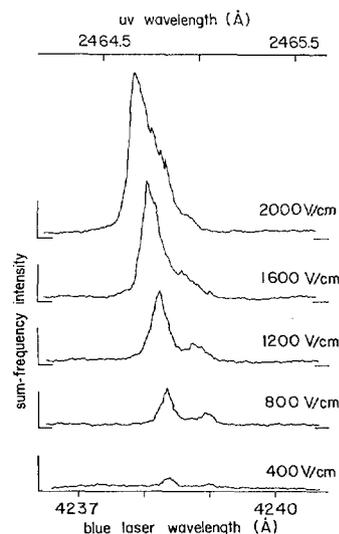


Fig. 5. Tuning characteristics of the sum-frequency generation process in the region near the $3s$ – $12p$ transition for several different values of the dc electric field. The yellow laser was tuned to the D_2 line.

the atomic species followed by radial charge separation. This photo-ionization rate is enhanced when the applied fields are tuned to a two-photon allowed resonance with an s or d level, leading to the peaks observed in our data. Another proposed mechanism that can produce a second-order nonlinearity in an atomic vapor is an electric quadrupole or magnetic dipole coupling between two of the interacting levels [19]–[21], [24]. This model cannot explain our observation of sum frequency generation at the $3s$ – ns frequency since no multipole moment connects two s states. At an applied dc field strength of 400 V/cm, resonances appear at the energies of the s , p , and d levels, as shown in Fig. 6. As the field strength is increased still further, the position of the p -state resonance shifts to higher energy, that of the s -state resonance remains approximately fixed, and the breadth of the d -state resonance increases.

In Fig. 7 we show a series of scans obtained for several different values of the detuning of the yellow laser and with the dc electric field fixed at its maximum value. We see that the positions of the resonances excited by the blue laser depend on the detuning of the yellow laser, as expected for a two-photon resonantly enhanced parametric mixing process. These

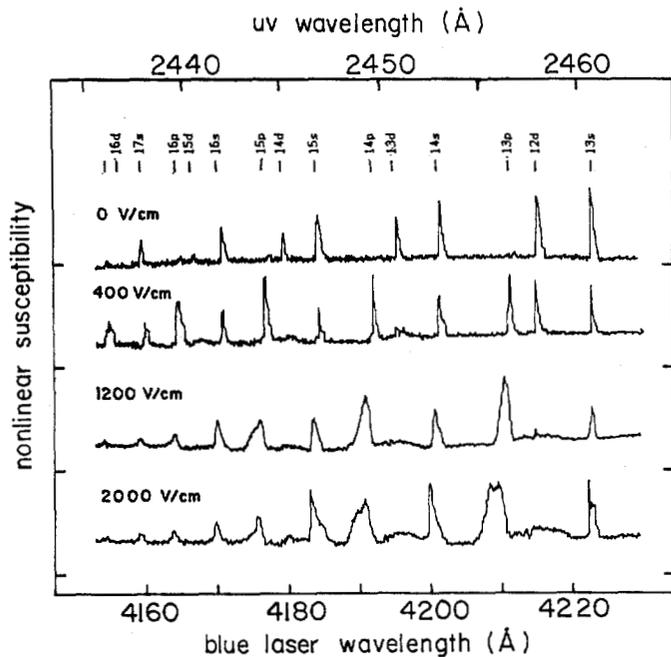


Fig. 6. Tuning characteristics of the sum-frequency generation process for several values of the dc electric field, with the yellow laser detuned 1.3 Å to the low-frequency side of D_2 . The positions of the two-photon resonances in the limit of zero field strength are indicated.

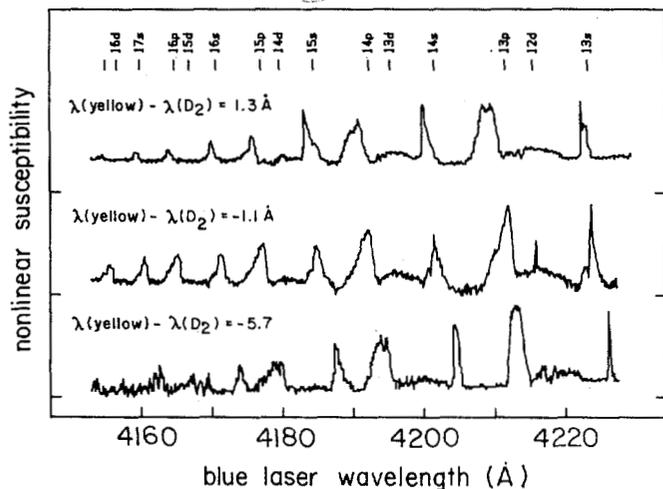


Fig. 7. Tuning characteristics of the sum-frequency generation process are shown for several different values of the yellow laser detuning. The dc field strength is 2000 V/cm. The positions of the two-photon resonances for an on-resonance yellow laser are shown. The experimental curves are shifted from these positions due to the detuning of the yellow laser.

curves also show that at the largest field strengths the various resonances have broadened into one another, leading to broad regions of continuous tunability.

We have performed another measurement to determine the maximum achievable value of the nonlinear susceptibility under our experimental conditions. The maximum output signal is obtained with the yellow laser detuned 1 cm^{-1} to the high-frequency side of D_2 and with the blue laser tuned to complete a two-photon resonance with the $13s$ level, with a dc field strength of 2000 V/cm. Under these conditions, the measured conversion efficiency (defined as the ratio of the UV output energy to the total energy of the two input

pulses) was found to be 3×10^{-4} . The nonlinear susceptibility is calculated using the equation relating the input and output intensities to the length of the interaction region and to the k -vector mismatch Δk [26]. For our conditions, the k vector mismatch is due primarily to the dispersion of the D_2 resonance line and has the value 58 cm^{-1} . Taking the input energies as 0.08 mJ, the pulse length as 4 ns, and the laser beam radius as 1.3 mm, we calculate the value $\chi^{(2)} = 1.2 \times 10^{-8}$ ESU. Due to uncertainties regarding our beam profile within the interaction region, this value is probably accurate only to within a factor of 3. This measured value is in good qualitative agreement with the predicted value given above.

CONCLUSIONS

In conclusion, we have demonstrated that extremely large values of the second-order nonlinear susceptibility can be obtained through use of Rydberg states perturbed by an external electric field. Values of $\chi^{(2)}$ as large as 1.2×10^{-8} ESU have been obtained in a sodium vapor of density $4 \times 10^{14} \text{ cm}^{-3}$. This value is approximately ten times larger than that of KDP. The splitting of the highly excited levels due to the external electric field creates a broad region of overlapping levels, leading to continuous tunability. This technique can be applied to any system displaying a Rydberg series. When applied to a system with a large ionization potential such as hydrogen or a noble gas, it should be capable of producing coherent tunable radiation in the extreme ultraviolet.

ACKNOWLEDGMENT

The authors gratefully acknowledge useful discussions with M. G. Littman, D. G. Steel, and C. R. Stroud, Jr. We thank M. A. Kramer for his assistance in the construction of the tunable dye lasers.

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Robert W. Boyd received the B.S. degree in physics in 1969 from the Massachusetts Institute of Technology, Cambridge, and the Ph.D. degree in physics in 1977 from the University of California, Berkeley. His thesis work involved the use of nonlinear optical techniques for infrared detection for astronomy.

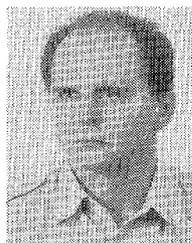
In 1977 he became Assistant Professor and in 1982 Associate Professor at the Institute of Optics, University of Rochester, Rochester, NY. His research interests include studies of

the nonlinear optical properties of materials, nonlinear optical interactions involving atomic Rydberg states, and the development of new laser systems.



Daniel J. Gauthier was born in Peterborough, NH, on October 29, 1960. He received the B.S. degree in 1982 and the M.S. degree in 1983, both in optics from the University of Rochester, Rochester, NY.

As an undergraduate, he worked on subsystem optimization for the Laboratory for Laser Energetics' OMEGA fusion laser. His current research involves the study of nonlinear frequency mixing and optical phase conjugation.



Jerzy Krasinski received the M.S. degree in physics in 1966 and the Ph.D. degree in 1973 from the University of Warsaw, Warsaw, Poland. His thesis work was concerned with photon statistics and nonlinear absorption.

In 1973 he became an Adjunkt (equivalent to Assistant Professor) and in 1977 a Docent (equivalent to tenured Associate Professor) and head of the Department of Optics of the University of Warsaw. He has also had visiting positions at Indiana University, Bloomington,

Max-Planck-Institut, Garching, West Germany, Colgate University, Hamilton, NY, and the University of Rochester, Rochester, NY. Since 1982 he has lived in the United States and in 1984 he became Senior Research Physicist at Allied Corporation. His research has involved nonlinear optics, laser spectroscopy, new laser media, and development of new laser systems.



Michelle S. Malcuit was born in Akron, OH, on June 22, 1957. She received the B.A. degree in physics and chemistry from the State University of New York College at Potsdam in 1979.

Currently, she is at the Institute of Optics, University of Rochester, Rochester, NY, where her research involves the development of tunable solid-state lasers and nonlinear optics.