Generation of 1182-Å radiation in phase-matched mixtures of inert gases*

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Coherent radiation at 1182 Å is obtained by third-harmonic generation in a phase-matched mixture of Xe and Ar. For generation from 3547 to 1182 Å, Xe is negatively dispersive, and phase matching is obtained at a ratio of Xe : Ar = 1 : 430. A conversion efficiency of 2.8% is obtained at an input power of 13 MW. As predicted by theory the conversion efficiency increases linearly to the limit of our available input power.

Third-harmonic generation in phase-matched mixtures of metal vapors and inert gases has recently been described both theoretically and experimentally.[1-3] In the first experiments, 1.064-μ radiation was tripled to yield 3547-Å radiation in a mixture of rubidium and xenon. Recently, the technique was extended to the vacuum ultraviolet where 1773-, 1520-, and 1182-Å radiation was generated in a mixture of Cd and Ar. Though the metal-vapor—inert-gas system is attractive theoretically,[4] a number of practical problems concerned with the homogeneous mixing of the metal vapor and inert gas have thus far limited the conversion efficiency to about 10^{-4}.

In the present letter, we report the generation of 1182-Å radiation in a phase-matched mixture of Xe and Ar. No metal vapor is employed. The large nonlinearity of Xe and the homogeneous mixing of the inert gases allows a conversion efficiency of 2.8% at an input power of 13 MW. This efficiency increases linearly to the limit of our available input power at 3547 Å.

To obtain efficient third-harmonic generation it is essential that one of the inert gases be negatively dispersive (have a refractive index at the third-harmonic frequency less than its refractive index at the fundamental frequency). This allows the use of a positively dispersive gas to phase match, or of equal importance, the use of very tight focusing to the center of a gas cell.[4,5] In the case of the metal vapors, the negative dispersion is obtained by allowing the lower or fundamental frequency to be less than and relatively close to the resonance frequency of the metal vapor. In the case of Xe, the negative dispersion is obtained by allowing the third-harmonic frequency to be greater than and relatively close to a transition of high oscillator strength. (In these experiments, the 1192-Å $5p$-$5d$ transition of Xe is probably the dominant transition involved.) The lowest energy level of any inert gas is the 1469.5-Å line of xenon; thus inert-gas—inert-gas generation is possible over the spectral region of 1469 to at least 500 Å, where the continuum of helium begins.

We note that third-harmonic generation in inert gases has been demonstrated much earlier by Ward and New.[5] In these important early experiments, the third harmonic of ruby at 2314 Å was generated in each of the inert gases. However, in this spectral region, the inert gases are not negatively dispersive and their nonlinearity is much lower ($\chi^{(3)} = 9.8\times10^{-37}$ esu for xenon). The conversion efficiency reported by Ward is about eleven orders of magnitude lower than that reported here.

Our experimental setup is similar to that reported in Ref. 3 and is shown in Fig. 1. A single pulse from a mode-locked 1.064-μ Nd:YAG laser is amplified by a Nd:YAG amplifier to yield an estimated peak power of 3×10^8 W and a pulse length of 25 psec at 1.064 μ. The pulse is frequency doubled in an ADP crystal to 5320 Å, and mixed with remaining 1.064-μ radiation to yield 3547 Å. A maximum peak power of 1.3×10^7 W is obtained at 3547 Å. Two Xe:Ar gas cells were employed in these experiments. Each had a quartz input window and a lithium fluoride output window. The generated 1182-Å radiation was directed into a helium-purged lithium fluoride prism spectrometer. Detection was accomplished with a solar blind model EMR 542G photomultiplier with a cesium iodide photocathode. A sensitive lithium tantalate pyroelectric detector[6] was used for absolute intensity measurements at 1182 Å.

In the first set of experiments, the laser was focused to a spot with 70-μ diameter (confocal parameter, 2.1 cm), and a 0.95-cm-long cell was placed at the center of the focus. The xenon pressure was fixed at 1 Torr. Generated third-harmonic power at 1182 Å was monitored as the argon pressure was gradually increased. Experimental results are shown in Fig. 2. Peak third-harmonic power was obtained at an Ar:Xe ratio of 430:1. The Xe pressure was then increased to 5.7 Torr, and the experiment was repeated. Peak third-harmonic power was again obtained at a ratio of 430:1, and was 2500 times greater than that obtained with pure Xe. The conversion efficiency for these experimental conditions

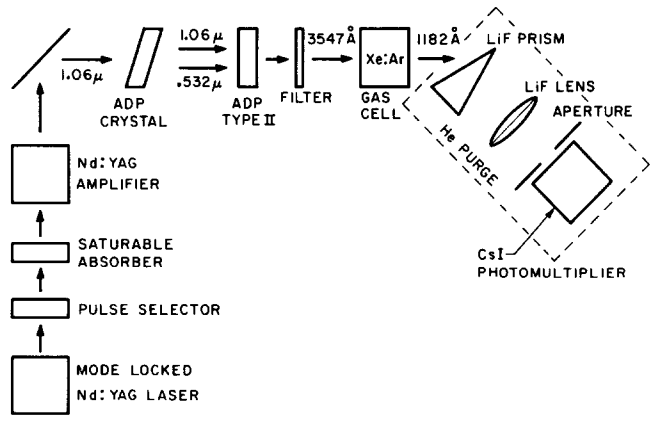

FIG. 1. Schematic of experimental apparatus for 1182-Å generation.

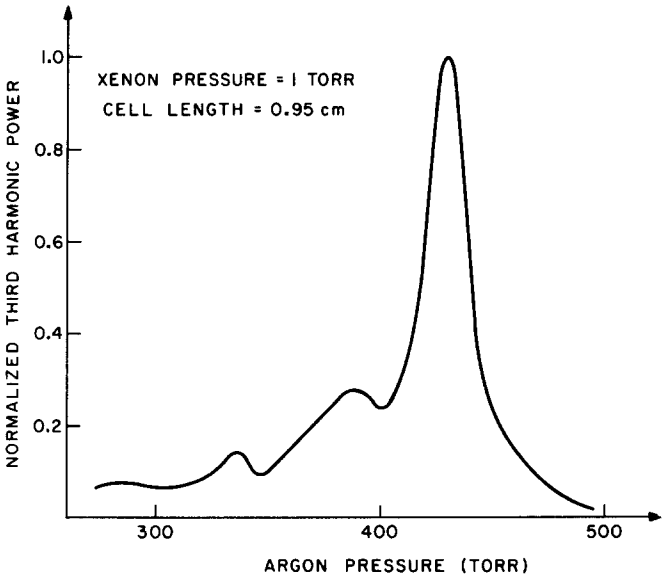

FIG. 2. Normalized 1182-Å output power vs argon pressure. Confocal parameter of input beam is 2.1 cm.

was 0.13%. A further increase in Xe pressure did not yield significantly higher output powers. The ratio of third-harmonic power outputs with argon present to that with argon absent, yields a coherence length for pure xenon of -0.033 cm at 10^{17} atoms/cm^3. The measured conversion efficiency, cell length, and coherence length yield a nonlinear susceptibility $\chi^{(3)} \simeq 2.5\times10^{-34}$ esu.

To obtain higher conversion efficiency, the 3547-Å radiation was focused to a confocal parameter of 0.25 cm in the center of a 9.5-cm cell. At an input power of 13 MW the power density on the cell windows was still reasonable, while the density at the focus was about 6.3×10^{12} W/cm^2. For these tight focusing conditions, the ratio of Ar to Xe which was necessary to achieve phase matching was reduced to about 50:1. This reduction in ratio is a result of the tighter focusing employed.[4] At an input power of 13 MW and an optimized Xe pressure of 3 Torr, an energy conversion efficiency of 2.8% from 3547 to 1182 Å is obtained. For these tight focusing conditions, even pure Xe at a pressure of 3 Torr yields a conversion efficiency of 0.9%.

In general, to obtain maximum conversion efficiency, it is desirable to work at the highest power density allowed by either breakdown or multiphoton ionization. For the 25-psec pulses employed in our experiment, we found that the third-harmonic power output varied as the cube of the incident power up to an incident power density of 7×10^{12} W/cm^2. This may be compared with a four-photon ionization density of 1.7×10^{12} W/cm^2 estimated by Morton,[7] and 1×10^{13} W/cm^2 estimated by Bebb and Gold.[8]

A different type of saturation of the third-harmonic output power results due to absorption at the third-harmonic frequency causing a change in the refractive index and a breaking of the phase-matching condition. If the pressure of the Xe is reduced as the square root of the incident energy density, theory[4] predicts that conversion efficiency should increase linearly with input power. This was found to be the case to the limit of our available power. On the basis of the measured susceptibility and with the assumption that theory continues to hold, 20% conversion efficiency should be obtained at an input power of about 9.3×10^7 W. For this input power the laser should be focused to a confocal parameter of 1.8 cm ($P/A = 5.8\times10^{12}$ W/cm^2), and 1.1 Torr of Xe and 28 Torr of Ar should be used.

The general technique of phase-matched harmonic generation in mixtures of inert gases should be applicable to the spectral region from 1469 to at least 500 Å. Tripling of the second harmonic of a mode-locked ruby laser to yield radiation at 1157 Å should be obtained at a ratio slightly less than that reported here, and with a nonlinearity which is approximately the same. By tripling the radiation obtained from dye lasers and frequency-doubled dye lasers,[9,10] high-power tunable radiation over much of the vacuum ultraviolet should also be obtainable.

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