



4.4 Vacuum Ultraviolet Ar₂* Laser Pumped by a High-Intensity Laser

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We observed a small-signal gain of Ar₂* emission at 126 nm by use of an Ar-filled hollow fiber to guide the ultrashort-pulse high-intensity laser propagation. The small signal gain coefficient was measured to be 0.05 cm⁻¹ at 126 nm. Kinetic analysis revealed that the electrons produced by the high-intensity laser through an optical-field ionization process initiated the Ar₂* production process. This laser scheme could be combined with high harmonic radiation of the pump laser in the vacuum ultraviolet (VUV), leading to the production of amplified ultrashort VUV pulses.

Keywords: Vacuum ultraviolet laser, Ultrashort pulse laser, Rare gas excimers,
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1. Introduction

Practical vacuum ultraviolet (VUV) lasers have been desired in various industrial and scientific application fields. Such VUV lasers are applicable for photochemistry, biological science, and new materials processing, since they produce high-energy photons with high photon flux. Currently available practical VUV lasers are ArF and F₂ lasers at 193 and 157 nm, respectively. In addition to these lasers, rare gas excimers have been one of the practical VUV laser media since 1970s. Their center wavelengths are 126, 147, and 172 nm for Ar₂*, Kr₂*, and Xe₂*, respectively. These wavelengths are short enough to be applied to the above application fields, and are long enough for utilization of transmission optics such as MgF₂ or LiF. Among these rare-gas excimers, the Ar₂* laser produces radiation with the highest photon energy of 9.8 eV. A high-peak-power laser operation pumped by an electron-beam excitation method has been developed [1]. This method, however, may not be appropriate to produce "practical lasers" because of its large facility

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with a limited repetition rate. An alternative excitation method, therefore, has long been desired.

We proposed a new excitation method to demonstrate a practical Ar₂* laser using a high-intensity ultrashort pulse laser [2]. A high-intensity laser produced electrons are utilized to initiate the Ar₂* production kinetics. Production of the initial electrons by an optical-field ionization (OFI) process will be adequate for the Ar₂* production since the electron parameters can be controlled by appropriate choice of the high-intensity laser parameters [3]. The calculated initial average electron energies of Ar plasma at the intensity of 10¹⁵ Wcm⁻² are 80 and 5 eV, respectively, for circularly and linearly polarized laser pulses. Based on the laser energy that is dissipated in the fiber, a maximum electron density of 10¹⁷ cm⁻³ is estimated at the Ar pressure of 10 atm. These electron characteristics are, therefore, very suitable to initiate the excimer formation kinetics. An ultrashort-pulse pump laser can also provide coherent harmonic radiation in the relevant VUV spectral region in rare gases, which could be utilized as an ultrashort VUV seed pulse. An OFI produced Ar₂* excimer laser would be applied as an amplifier and could be combined with the ultrashort VUV seed pulse, resulting in the production of ultrashort pulse high intensity VUV radiation.

2. Experiment

Detail of the experimental setup is found elsewhere [4,5]. The setup consists of three elements: a high-intensity laser to produce an OFI plasma, a high-pressure Ar cell (~10 atm) where a hollow fiber is installed, and a VUV detection system. A Ti:sapphire laser at 780 nm produced the maximum output energy of 1.5 mJ with a pulse width of 150 fs at a repetition rate of 10 Hz. The laser beam was focused at a hollow core of the hollow fiber with a diameter of 250 μm installed a high-pressure Ar cell. The focused laser intensity was measured to be 10¹⁵ Wcm⁻². According to the Ammosov-Delone-Krainov theory [6], this laser intensity was high enough to selectively produce singly ionized Ar ions that lead to Ar*, a precursor of Ar₂* excimer. Different lengths of 15, 30, and 50 cm were chosen to guide the focused laser pulse. Time-integrated emission spectra from a fiber were detected by a VUV microchannel plate coupled to a VUV spectrometer. Solid angle correction of the VUV emission caused by three different fibers was performed by measurement of the beam pattern of the visible laser radiation at the fiber exit. The temporal behavior of the Ar₂* emission at 126 nm was measured by use of a fast VUV photomultiplier tube connected to a digital oscilloscope. All equipment was connected to a personal computer for signal collection and processing.

3. Ar₂* excimer production

We observed Ar₂* emission centered at 126 nm, with a spectral bandwidth of 9 nm (FWHM). This bandwidth has a typical value similar to those observed in different excitation methods [7,8]. The Ar₂* emission intensity at 126 nm quadratically increased with the Ar pressure. This pressure dependence of Ar₂* emission intensity indicated that the excimer production kinetics of the high-intensity laser was mainly governed by the three-body association process ($\text{Ar}^* + 2\text{Ar} \rightarrow \text{Ar}_2^* + \text{Ar}$) as expected.

Figure 1 shows the temporal behavior of Ar₂* fluorescence emission at 126 nm. The Ar

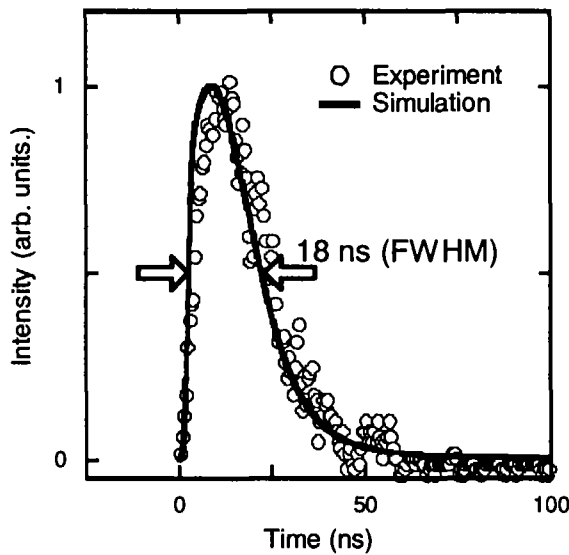


Figure 1 Temporal behavior of Ar_2^* emission. Calculated temporal result reproduces the experiment.

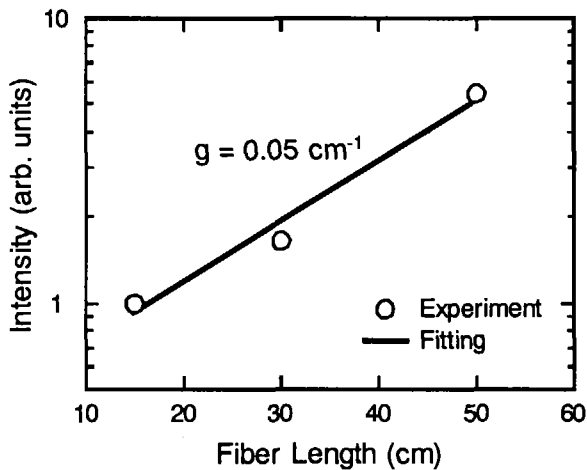


Figure 2 Exponential increase of the emission intensity as a function of the fiber length.

pressure was 10 atm. The pulse duration was 18 ns (FWHM). This pulse width would be long enough for one to construct an optical cavity for laser operation and eventually an amplifier of a seed beam. The calculation was performed based on a kinetics code developed for an electron-beam-excited Ar_2^* excimer laser. Initial electron energy and density were assumed to be 5 eV and 10^{17} cm^{-3} , respectively. Electron conductive cooling process analyzed elsewhere [9] was also included in the calculation. The agreement of the experiment verifies that the electrons produced by the high-intensity laser through an OFI process initiated the excimer formation kinetics with the assumed initial plasma conditions. Note that no excimer emission was observed when a self-breakdown plasma was produced by a nanosecond Q-switched Nd:YAG laser [10]. Inverse bremsstrahlung process and the following electron avalanche in a nanosecond laser-produced plasma produced highly uncontrolled plasma conditions, which was in contrast to those observed using a high-intensity laser.

Figure 2 represents the exponential increase of the Ar_2^* emission intensity as a function of a fiber length. Three fibers with lengths of 15, 30, and 50 cm were used. The Ar pressure was 4 atm. The laser intensity was 10^{15} Wcm^{-2} , so the laser beam was focused more tightly than the diameter of the hollow fiber. This tight focusing was necessary to yield the excimer emission. Since the pulse

broadening of the excitation pulse was calculated to be negligible under our experimental conditions, the excitation intensity was kept constant during pulse propagation inside a fiber. Based on this fiber-length dependence of the emission intensity, a small-signal gain of 0.05 cm^{-1} was evaluated [4,5]. The fiber length was considered to be same as the excitation length, since visible plasma emissions were observed over the fiber length when the laser pulse propagated. Note that this is to our knowledge the first observation of a

reasonable gain coefficient at 126 nm other than the electron-beam-pumped Ar₂* laser. An optical cavity construction is under way to produce laser oscillation at 126 nm. The maximum laser output energy of the order of 1 mJ would be plausible when an optimum cavity at 126 nm is assumed. Present technology to produce high quality optics at 126 nm may limit the output energy of this laser system. Since this laser scheme is realized by an ultrashort-pulse laser, the same plasma-initiating laser could be utilized to produce an ultrashort seed pulse at 126 nm, or other excimer wavelengths by use of high harmonic radiation. Demonstration of this ultrashort VUV amplification scheme is currently in progress.

4. Conclusions

We have observed a small-signal gain of Ar₂* emission at 126 nm by use of an Ar-filled hollow fiber to guide the high-intensity laser propagation. Kinetic analysis revealed that the electrons produced by the high-intensity laser through an OFI process initiated the Ar₂* production processes. The observation of a small-signal gain would lead to laser oscillation at 126 nm by use of an appropriate optical cavity. Furthermore, by combining this scheme with high harmonics, an ultrashort VUV amplifier would be plausible after careful analysis.

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