

Intracavity C atom absorption in the tuning range of the ArF excimer laser

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The photodissociation of carbon monoxide at 193 nm has received great experimental interest in recent years.¹⁻⁶ The resonant excitation of the spin-forbidden $a^3\Pi(v'=2) \leftarrow X^1\Sigma^+(v''=0)$ transition of CO by the ArF excimer laser at 193 nm, followed by absorption of at least two additional 193 nm photons, leads to the dissociative production of C(3^1P) atoms, which fluoresce at 247.9 nm to the C(2^1S) state. There are two possible dissociation channels leading to the production of 3^1P carbon atoms: (a) by a one-photon dissociation of the $a^3\Pi$ state, forming atomic carbon C(2^1D), followed by the resonant excitation to the C(3^1P) state at 193.1 nm or (b) by a two-photon dissociation of the $a^3\Pi$ state whereafter C(3^1P) atoms are directly formed. Many studies^{1,2,4,6} follow the pathway according to case (a), where the coincident overlap of the atomic carbon transition with the ArF gain profile is needed to produce 3^1P carbon atoms.

In this note we want to draw the reader's attention to the possible occurrence of an intracavity absorption in the discharge cavity of the ArF excimer laser at exactly the C: $3^1P \leftarrow 2^1D$ transition frequency. In our tunable ArF excimer laser (Lambda Physik EMG 150 MSCT) an intracavity absorption prevents laser action around $51\,790\text{ cm}^{-1}$. The absorption line is as broad as 2.5 to 10 cm^{-1} , depending on the quality of the laser filling. Lasing could never be achieved at the center frequency. By calibrating the laser against NO,⁷ the center frequency of the absorption was determined to be $51\,789.2 \pm 0.2\text{ cm}^{-1}$. The absorption is located near two well-known oxygen resonances [O_2 : $B^3\Sigma_u^-(v'=4) \leftarrow X^3\Sigma_g^-(v''=0)$ P(15) and R(17) at $51\,783.6$ and $51\,788.3\text{ cm}^{-1}$,⁸ respectively], and has previously been confused with these. The absorption is clearly seen only when the oscillator output is monitored directly, e.g., when the laser is used in the so-called amplified oscillator mode. In this mode the unstable resonator optics of the amplifier are removed and the oscillator beam is directed in a single pass through the amplifier cavity.⁷ When the laser is used in the standard mode the cessation of laser action in the oscillator does not appear in the total output energy of the laser, because the amplifier itself delivers 50 mJ of broadband energy when not injected by sufficient oscillator power. In the amplified oscillator mode the power drops to zero when no oscillator power is available.

We believe that the absorption is due to the strong $3^1P \leftarrow 2^1D$ transition of the metastable carbon atom at $51\,789.18\text{ cm}^{-1}$. This absorption line has been reported before under near lasing conditions of a broadband ArF

excimer laser.⁹ The $3^1P \leftarrow 2^1D$ transition is one of the strongest C atom transitions, with a transition probability of $3.7 \times 10^8\text{ s}^{-1}$.¹⁰ Several ppb of metastable 1D carbon atoms in the discharge cavity are therefore enough to defeat laser action at this frequency.

The output power of the tunable ArF excimer laser (bandwidth 0.4 cm^{-1}) near the C atom absorption line is shown in the upper part of Fig. 1. For this the beam path to the detector and the intracavity and extracavity components of the laser are flushed with nitrogen to decrease the intensity of the oxygen absorption lines. The C atom absorption is clearly seen in the figure. The O_2 : $B^3\Sigma_u^-(v'=4) \leftarrow X^3\Sigma_g^-(v''=0)$ P(15) and R(17) transitions are indicated in the upper part of the figure. This plot is taken with a fresh gas filling. After 3×10^4 laser shots at 10 Hz the absorption line appears much broader as is seen in the lower part of Fig. 1. The carbon atoms may be formed in

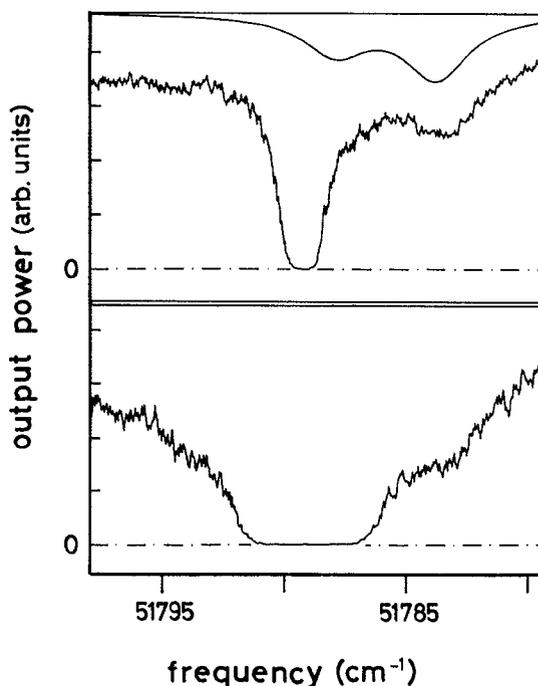


FIG. 1. Upper: The output power of the tunable ArF laser near $51\,790\text{ cm}^{-1}$ with a fresh gas filling. The strongest absorption, due to the carbon atom $3^1P \leftarrow 2^1D$ transition, has a linewidth of 2.6 cm^{-1} . The calculated line profile of the O_2 : $B^3\Sigma_u^-(v'=4) \leftarrow X^3\Sigma_g^-(v''=0)$ P(15) and R(17) absorption lines are indicated in the upper part of the figure. The linewidth of 3.9 cm^{-1} is determined by the predissociation lifetime of the $B^3\Sigma_u^-(v'=4)$ state. The linewidth of the ArF laser is 0.4 cm^{-1} . Lower: The output power after 3×10^4 laser shots. The absorption linewidth has increased to 8.2 cm^{-1} , due to an increased number of carbon atoms.

the discharge from impurities in the filling gas or from other materials in the discharge tube. The filling gas is composed of 90% helium (gas purity 99.99%), which contains several ppm of CO, CO₂, and hydrocarbons. In the discharge these molecules can dissociate to produce C(2^1D) atoms which give rise to the absorption line. Contamination by oil, originating from the rotary pump used to evacuate the laser tubes, may be an additional source of the metastable C atoms.

Although not always recognized as such, the strong C atom absorption is reflected in excitation spectra measured with the tunable excimer laser.^{3,11} It should be explicitly noted that resonant $3^1P \leftarrow 2^1D$ excitation of metastable C(2^1D) atoms is not possible with our tunable excimer laser, since there are no ArF laser photons available at the transition frequency.

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