Stimulated VUV Radiation from HD Excited
by a Picosecond ArF* Laser*

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Abstract

Stimulated emission on $E, F \rightarrow B$, $B \rightarrow X$, and $C \rightarrow X$ transitions of HD is observed following two-photon excitation in the E, F state with a picosecond ArF* laser. The efficiency of the strongest $B \rightarrow X$ line is $\sim 1\%$. Electron collisions are found to play an important role for $C \rightarrow X$ emissions.

1. Summary

Rare gas halogen lasers have been used extensively as a fundamental source for wavelength conversion by various nonlinear processes [1]. Multiphoton excitation of appropriate gain media has been shown to be a promising method for the production of coherent short wavelength radiation. Since these processes have a strongly nonlinear scaling, the efficiency of the wavelength conversion is strongly influenced by the intensity. However, the presence of photoionization of the excited state commonly limits the energy fluence for efficient conversion of pump radiation to short wavelength radiation [2]. Therefore, short pulse excimer lasers are ideal pump sources for multiphoton excited short wavelength lasers. In a previous experiment, a picosecond ArF* (193 nm) laser [3] has been used to two-photon excite the E, F state of molecular hydrogen [4]. The observation of subsequent stimulated emission in the vacuum ultraviolet (VUV) has confirmed the importance of the aforementioned factors. In this paper, it is shown that the same technique can be used to excite the E, F state of the related heteronuclear molecule HD. Strong stimulated emission in the VUV spectral range is observed following excitation of the E, F ($v = 6$) $J = 0, 1$ states of HD, with energy conversion efficiencies in one line approaching 1%.

The experimental apparatus consists of a 1-m vacuum monochromator (McPherson 225) with a PAR optical multichannel detector and analyser (OMA) at the exit port. A 3-m tube was attached to the entrance slit chamber. The tube has a fused silica entrance window and a LiF exit window and was filled with HD at pressures between a few Torr and 100 Torr. A 2 GW ArF* laser beam with a 10 ps pulse duration and 5 cm^{-1} bandwidth is focused with a 1.6 meter focal length lens to the middle of the tube and directed into the monochromator through the entrance slit. Because of the traveling wave character of the excitation, the amplified sponta-

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neous VUV emission (ASE) is expected to be mainly in the forward direction, and the use of a resonator does not relax the gain requirements. Therefore, no mirrors were used. For saturation of the gain medium by ASE, a small signal gain-length product $g_0 l$ of ~ 20 is necessary.

Several attempts to produce stimulated emission from E,F levels excited via S transitions ($\Delta J = -2$) were unsuccessful. However, the tuning range of the ArF* laser also allows selective population of the E,F $v = 6$, $J = 0$ and $J = 1$ states via Q(0) and Q(1) two-photon transitions. In this case, strong, directional radiation was observed in the infrared and vacuum ultraviolet spectral ranges, originating from E,F $\rightarrow$ B, B $\rightarrow$ X and C $\rightarrow$ X transitions. For identification of observed emissions, Refs. 5-8 have been used. The shortest wavelength transition seen was at 84039 cm^{-1} , on the C $\rightarrow$ X ($2 \rightarrow 6$) R(1) line. The most intense emission registered was at 73644 cm^{-1} , the B $\rightarrow$ X ($0 \rightarrow 5$) P(3) transition. The excitation and emission pathway is similar to that which applies to H_2 [4]. Using Frank-Condon factors, which are available for E,F $\rightarrow$ B and B $\rightarrow$ X transitions [9], and Hönl-London factors [10], the relative strengths of different rovibrational transitions can be predicted qualitatively and are found to agree with observed line strengths. Naturally, a quantitative comparison can only be made if the exponential character of a laser process, the effects of saturation, and population depletion are taken into account.

As in previous measurements [4,11] in H_2 , the optical Stark effect causes the optimum excitation frequency to be blue-shifted with respect to the unperturbed value, whereas no shift could be detected in the emission spectra, indicating that the stimulated emission occurs slightly later than the excitation. With the unperturbed transition frequencies for the Q(0) and Q(1) transition at 103219 cm^{-1} and 103171 cm^{-1} , respectively, the actual two-photon frequencies for optimum excitation are $\sim 103290 \text{ cm}^{-1}$ and $\sim 103200 \text{ cm}^{-1}$, respectively.

Besides the E,F $\rightarrow$ B $\rightarrow$ X cascade emissions, radiation in the 120 nm range is observed, originating from C $\rightarrow$ X transitions. As reported previously [4,11] in H_2 , electron collisions are very efficient in transferring population from the E,F to the C state. Since the E($2s\sigma$) and C($2p\pi$) states resemble their atomic counterparts, the atomic mechanism discussed by PURCELL [12] has been used to estimate a collisional rate constant of $k_e \sim 10^{-5} \text{ cm}^3 \text{ sec}^{-1}$, which depends only on electron temperature and energy difference between the E,F and C states. Since these two parameters and all the other experimental conditions do not change significantly for HD, the rate constant will be approximately the same as in the case of H_2 , and electron collisions are considered to be the most probable transfer mechanism. As shown in Fig. 1, this collisional transfer process correctly predicts the C $\rightarrow$ X transitions observed. Other possible processes which have been considered are (a) stimulated emission in the far infrared, which is excluded because, in some cases, an energy increase is required, and (b) in the case of HD, direct two-photon excitation of the C state, which is allowed because of the broken symmetry of the heterogeneous molecule. However, it is expected that the two-photon transition rate is not large enough to result in strong stimulated emission because the

C state wave function still has mainly a "u" character with only a rather small admixture of a "g" type wave function [7]. In addition, when krypton gas is mixed with HD in order to increase the electron density, improved performance is observed on the $C \rightarrow X$ transitions. This is a further indication that electron collisions play the dominant role in the $E, F \rightarrow C$ transfer.

Fig. 1. The excited states participating in the electron collisional transfer from the E,F state to the C state. For $Q(0)$ excitation one path $0(+) \rightarrow 1(-)$ is observed, as shown by the dashed arrow, while for $Q(1)$ excitation two paths, $1(-) \rightarrow 1(+)$ and $1(-) \rightarrow 2(+)$, are seen, as shown by the solid arrows. All the electron collisional amplitudes observed conform to dipole selection rules.

In conclusion, strong stimulated emission has been observed in HD following two-photon excitation of the E,F state. In the VUV spectral range, 28 lines were identified, which brings the total number of hydrogen laser lines (H_2 and HD) in the VUV to greater than 50. Peak efficiencies on individual lines approach 1%. The AC Stark effect has been observed in the excitation spectrum but not in emission, and electron collisions are found to play a dominant role in the population of the C state. The high power and short wavelengths available from the H_2 and HD sources make them attractive for the performance of a wide range of studies. The H_2 source, for example, is currently being used in photoemission studies of GaAs surfaces.

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