Laser induced amplified spontaneous emission from the f'0_g^+(^{1}D) ion-pair state of I_2

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Molecular iodine has a series of the excited states correlating with atomic ions $I^-(^1S) + I^+(^3P, ^1D, \text{and } ^1S)$ at the dissociation limit. Enormous efforts have been paid for understanding their electronic structures in connection with the development of laser excitation techniques, and a great volume of spectroscopic data exists in the literatures. Under these circumstances, renewed interest has arisen for these excited states in view of providing the benchmark for the study on their relaxation dynamics.

Recently, we have demonstrated that two kinds of relaxation processes occurred to populate the $D0_u^+(^3P_2)$ state when the $E0_g^+(^3P_2)$ state was prepared by optical-optical double resonance [1]: One is the collisional-induced energy transfer from $E0_g^+(^3P_2)$ to $D0_u^+(^3P_2)$. The other is the process involving amplified spontaneous emission (ASE) in the $E0_g^+(^3P_2)$ - $D0_u^+(^3P_2)$ radiative transfer. We distinguished two processes from the temporal profiles of fluorescence signals, and showed that the completely different vibrational distribution occurred in the $D0_u^+(^3P_2)$ state.

The present study aims to elucidate the f'0_g^+(1D) – F'0_u^+(^{1}D) ASE transfer reported by Ridley *et al*. [2]. We prepared the $0_g^+(^1D)$ state in the double resonance transition through the $B^3\Pi(0_u^+)$ state. The spectrum showed three electronic transitions from the F'0_u^+(^{1}D) state following the F'0_u^+(^{1}D)-f'0_g^+(^{1}D) transfer, other than the emission of the f'0_g^+(1D) - $B^3\Pi(0_u^+)$ system. They corresponded to the transitions terminating on $X^1\Sigma_g^+$, $0_g^+(ab)$, and $0_g^+(bb)$. These vibrational structures were well-resolved to show that the F'0_u^+(^{1}D) state were formed through the Franck-Condon windows favourable for the f'0_g^+(^{1}D)-F'0_u^+(^{1}D) transition. We have rotationally analysed the $0_g^+(bb)$ state correlating with the dissociation limit of I(1/2) + I(1/2), designated as (*bb*), and the $0_g^+(ab)$, in terms of the Franck-Condon factor calculations to locate their potential functions on the internuclear distance, and revised their molecular constants, ω_e, R_e, and D_e. Furthermore, we have observed the infrared emission due to ASE, which propagated in a limited range of solid angles. The radiative f'0_g^+(1D) – F'0_u^+(^{1}D) transfer by ASE will be discussed in view of temporal profiles of emission intensities and the spectral features.

[1] Y. Nakano, H. Fujiwara, M. Fukushima, T. Ishiwata, J. Chem. Phys. **2008**, 128, 164320.
[2] T. Ridley, K. P. Lawley, R. J. Donovan, V. E. Alekseev, Phys. Chem. Chem. Phys. **2007**, 9, 5885.