Theoretical scheme for lasing without population inversion in the H_2 molecule

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We have shown that $(1+1')$-photon lasing without population inversion (LWOPI) can be achieved in a H_2 molecule for emission from the lowest doubly excited autoionizing (AI) state of ${}^1\Sigma_g$ symmetry to the ground $X\ {}^1\Sigma_g$ $(v=0,j=1)$ state in the presence of the resonant $B\ {}^1\Sigma_u$ $(v=4,j=2)$ level. We have considered both coherent and incoherent pumping of the AI state, which is the upper lasing level in this scheme. Coherent pumping of the AI state can be done by two-photon excitation from the ground to the AI state. Incoherent pumping of the AI state can be done by charge-transfer collision of a $H_2{}^+$ ion with atomic and molecular species. For a weak coherent pumping field of intensity 6.4×10^7 W/cm^2, a lower probe field [for the $X\ {}^1\Sigma_g$ $(v=0,j=1)\rightleftharpoons B\ {}^1\Sigma_u$ $(v=4,j=2)$ transition] of intensity 9.6×10^6 W/cm^2 is required to obtain an appreciable gain. However, for stronger coherent fields of intensity 6.4×10^9 W/cm^2, the lower probe intensity can be lowered by orders of magnitude without significant loss in gain. In this case, using a lower probe field of intensity 9.6×10^4 W/cm^2, we have calculated the gain by the perturbative method. However, for a lower value of coherent field strength, 6.4×10^7 W/cm^2, and a higher intensity of the lower probe field, 9.6×10^6 W/cm^2, we have done nonperturbative calculations and have shown that LWOPI can be achieved only in a short range of probe pulse duration less than or equal to 3.45 ps. In all these cases, the optimum intensity for the upper probe field [for the $B\ {}^1\Sigma_u$ $(v=4,j=2)\rightleftharpoons{}^1\Sigma_g$ transition] is 9.6×10^7 W/cm^2. We have also compared the evolution of gain for the $\mathrm{sech}^2(t/T)$-shaped pulse with that for the square pulse, the pulse duration $T=3.45$ ps being the same in both cases.

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INTRODUCTION

Recent theoretical studies have proposed several models [1–7] to show that either lasing or amplification on a probe field is achieved in the absence of population inversion. Some experimental [8–10] investigations have confirmed a few of the proposed models in atomic systems. In the case of a two- or three-level system of which one or two levels are autoionizing (AI), it has been shown that the gain is obtained around the absorption minimum and persists for a much longer time when the two AI states are coupled via a continuum. Recently, it has been shown [7–11] that two-photon lasing or amplification from an excited state (it may be a single AI state or two AI states coupled via a continuum) is possible in the presence of an intermediate resonance. The effects of near-resonant channels on these amplification processes have also been shown to be important [12]. All these studies are based on parameters that should be fitted with physical systems. A recent study on the amplification of the third-harmonic radiation field in the Ca atom has shown [13] that the presence of a Fano minimum in an AI line shape facilitates the amplification process. In a previous work [14] we have shown that for $(1+1)$-photon resonance enhanced autoionization, the absorption profile differs significantly from the Fano profile obtained for single-photon autoionization. Therefore, the condition for a gain in such a system differs from that of the two-level single-photon autoionizing system. If this single-photon autoionizing transition is replaced by an n-photon term, the above-mentioned features remain unchanged in the weak-field limit. In this work we have chosen such a $(1+1)$-photon configuration in a H_2 molecule and have shown that the lasing from the lowest AI state of H_2 is possible without population inversion in the presence of (i) both coherent and incoherent pumping and (ii) coherent pumping alone. However, in the absence of coherent pumping, the amplification of the probe fields may be obtained in the weak-field limit when incoherent pumping is done from the outside. To our knowledge, this is the first theoretical study of lasing without population inversion in a real molecular system.

The transition scheme considered here is shown in Fig. 1. The upper and lower lasing levels for $(1+1)$-photon emission in the H_2 molecule have been chosen to be the doubly excited lowest AI state (${}^1\Sigma_g$ symmetry) and the ground $X\ {}^1\Sigma_g$ $(v=0,j=1)$ state, respectively. The intermediate $B\ {}^1\Sigma_u$ $(v=4,j=2)$ state has been chosen to be the resonant level. Incoherent pumping of the AI state can be done from the outside by the process of electron capture in this doubly excited state in collision of the H_2^+ ion with H_2 or other atomic or molecular species. The coherent pumping of this AI level from the ground state can be achieved by a two-photon transition or by a resonant transition via the $B'\ {}^1\Sigma_u$ state or the $B''\ {}^1\Sigma_u$ state of H_2. Since the Fano q parameter for these transitions [15,16] can be very large (much greater than 10^2), one can neglect the direct photoionization loss to the continuum caused by this coherent pumping field when the laser frequency is tuned around the peak value of autoionization line shape. The two probe fields considered here

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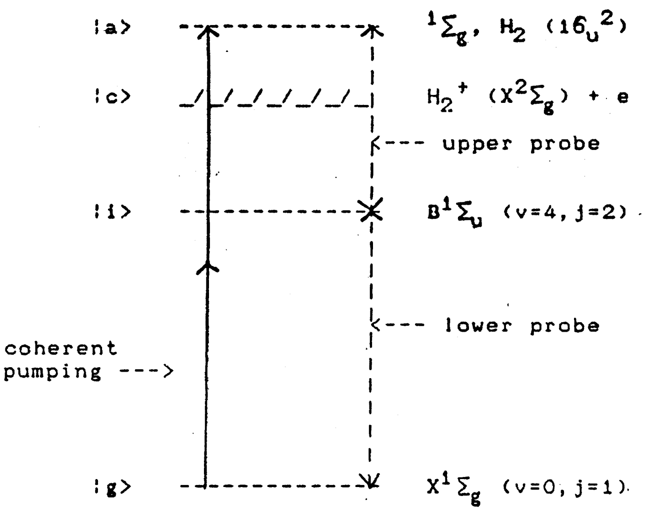

FIG. 1. Schematic diagram for the transitions considered in the H_2 molecule.

are of two different wave lengths, one around 105 nm (the lower step transition $X\ ^1\Sigma_g \rightleftharpoons B\ ^1\Sigma_u$) and the other around 315 nm (the upper step transition $B\ ^1\Sigma_u \rightleftharpoons {}^1\Sigma_g$). We have studied the dependence of two-photon gain on the variation of the strength of coherent pumping and the relative intensities of the two probe fields.

THEORY

To obtain the emission and absorption probabilities we use the resolvent operator technique [11,12]. Starting from the resolvent operator equation

$$(Z-H)G(z)=1, \tag{1}$$

where $G(z)$ is the resolvent operator and H is the total Hamiltonian, one can obtain the set of equations for the matrix elements of resolvent operators $G_{pq}(z)=\langle p|G(z)|q\rangle$, where $|p\rangle$ and $|q\rangle$ are the product of bare atomic states $|r\rangle$ and the photon number states $|n\rangle$, i.e., $|p\rangle=|r\rangle|n\rangle$. Here the product state representations for the ground, intermediate, autoionizing, and continuum states are given as $|g\rangle|n\rangle$, $|i\rangle|n-1\rangle$, $|a\rangle|n-2\rangle$, and $|c\rangle|n-2\rangle$, respectively. The set of equations can be written as

$$\begin{aligned}
&(Z-E_g)G_{gg}-D_{gi}G_{ig}-D_{ga}^{(2)}G_{ag}=A,\\
&(Z-E_i)G_{ig}-D_{ig}G_{gg}-\int D_{ic}G_{cg}dE_c-D_{ia}G_{ag}=0,\\
&(Z-E_a)G_{ag}-D_{ai}G_{ig}-D_{ag}^{(2)}G_{gg}-\int V_{ac}G_{cg}dE_c=B,\\
&(Z-E_c)G_{cg}-D_{ci}G_{ig}-V_{ca}G_{ag}=0,
\end{aligned} \tag{2}$$

where the D_{pq}'s are the dipole transition moments due to the probe fields between product states $|p\rangle$ and $|q\rangle$. The coherent pumping amplitude $D_{ga}^{(2)}=I\mu_{ga}^{(2)}$ is a two-photon term between the states $|g\rangle|n\rangle$ and $|a\rangle|n-2\rangle$ due to the coherent pumping field of intensity I. The direct ionization width $\gamma_i=2\pi|D_{ic}|^2$ and the autoionization width $\Gamma=2\pi|V_{ac}|^2$, where V_{ac} is the configuration interaction between the AI states $|a\rangle|n-2\rangle$ and the continuum $|c\rangle|n-2\rangle$. In the presence of incoherent pumping we solve these equations twice for two different boundary conditions, i.e., $A=1$ and $B=0$ for absorption and $A=0$ and $B=1$ for emission. Hence one can get two sets of matrix elements of the resolvent operator formally as $G_{pq}(z)=f_p(z)/F(z)$, where $F(z)$ is a polynomial in z. The roots of the equation $F(z)=0$ correspond to poles of $G_{pq}(z)$ and hence the complex energies of the dressed states. By doing the inverse Laplace transformation of the matrix elements of the resolvent operator one can get the corresponding matrix elements of the evolution operator as $U_{pq}(t)$. Hence the emission, absorption, and gain probabilities can be obtained as

$$P^{(a)}(t)=1-|U_g^{(a)}(t)|^2-|U_i^{(a)}(t)|^2-|U_a^{(a)}(t)|^2, \tag{3a}$$

$$P^{(e)}(t)=|U_g^{(e)}(t)|^2+|U_i^{(e)}(t)|^2, \tag{3b}$$

$$\mathscr{G}(t)=P^e(t)-P^a(t). \tag{3c}$$

The positive value of $\mathscr{G}(t)$ as a function of time corresponds to lasing or amplification. Here we have omitted the second subscript on the evolution operator and put superscripts (a) and (e) to denote that these matrix elements have been obtained for absorption and emission, respectively. In the presence of incoherent pumping, initially the total population is considered to be equally distributed between states $|g\rangle$ and $|a\rangle$. In the absence of incoherent pumping the population in state $|a\rangle$ will grow by coherent pumping and we have solved the equations once for the boundary condition $A=1$ and $B=0$, i.e., for absorption. Since formally $|U_{ga}^{(e)}(t)|^2=|U_{ag}^{(a)}(t)|^2$, the emission probability can be written as $P^e(t)=|U_{ag}^{(a)}(t)|^2$, the fractional population retained in state $|i\rangle$ during emission being negligible. The absorption probability is given by Eq. (3a) as before. Hence gain can be obtained as the difference of the absorption probability from the emission probability.

In the presence of strong radiation fields where a Rabi oscillation between different bound levels leads to oscillating transition probabilities and the strong field causes an appreciable splitting of the degenerate dressed states, one will have to do nonperturbative calculations as mentioned above. However, in the weak-field limit, one can do perturbative calculations to obtain the rate of absorption, emission, and gain. Actually, for the perturbative calculation to be valid, the Rabi oscillation between different bound states should be weak enough so as not to impose any oscillation on the ionization yield as a function of time. In this limit one can neglect the splitting of degenerate product states due to radiation dressing and the matrix element of the resolvent operator can be expressed as $G_{pq}(z)=f_p(z)/(z-z_0)$, z_0 being the pole of $G_{pq}(z)$. Hence the corresponding matrix elements of the evolution operator can be obtained by an inverse Laplace transform to express the rate of absorption $dp^{(a)}/dt$ and emission $dp^{(e)}/dt$ in the limit $t\to 0$ and ∞, respectively. Hence the rate of gain can be written as

$$d\mathscr{G}/dt=dp^{(e)}/dt-dp^{(a)}/dt. \tag{4}$$

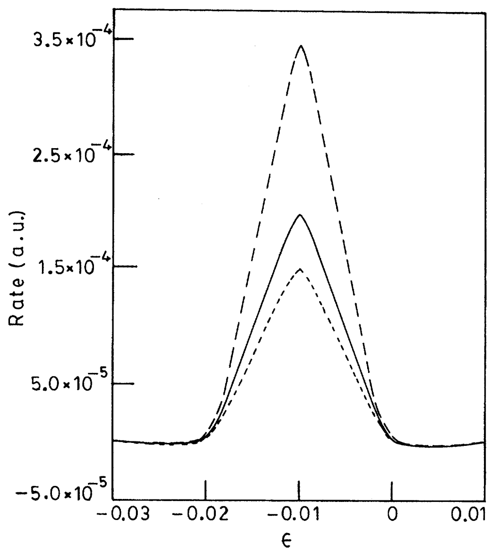

FIG. 2. Rate of absorption, emission, and gain as a function of dimensionless detuning $\epsilon = \delta/(\Gamma/2)$, δ being the detuning from the AI state. Here $\Gamma = 7.36 \times 10^{-4}$ a.u. ------, absorption rate; –––, emission rate; —, gain rate $d\mathcal{G}/dt$.

CALCULATION

The dipole transition moments and the autoionization and ionization widths used for this calculation have been obtained from a previous work [17]. Here the frequency of the lower probe field has been considered to be fixed at the resonant frequency between $B\,^1\Sigma_u$ $(v=4, j=2)$ and $X\,^1\Sigma_g$ $(v=0, j=1)$ levels, while that for the upper probe field can be varied around the resonance between the $B\,^1\Sigma_u$ $(v=4, j=2)$ level and the AI state. The molecule is excited to the continuum by this upper probe field just above the $v^+=1$ level of the H_2^+ ion. The photoelectron energies for the branching of the H_2^+ ion in $v^+=0$ and 1 levels are 0.32 and 0.05 eV, respectively. It should be mentioned here that while calculating the dipole transition moments for bound-bound and bound-continuum transitions, the explicit dependence of these moments on the internuclear separation and photoelectron energies has been taken into consideration. For this transition the turning point for the repulsive AI level is away from the Franck-Condon region; hence the direct photoionization dominates over autoionization leading to fractional values of the Fano q parameter.

To obtain an estimate of the strength of coherent pumping one can consider the two-photon matrix element of the order of 10 a.u. [16] and hence for a typical laser intensity 6.4×10^7 W/cm^2, the estimated value of $D_{ga}^{(2)}$, is approximately 1×10^{-7} a.u. For $D_{ga}^{(2)} \ll \Gamma$ and $D_{gi} \ll D_{ia}, D_{ic}, \Gamma$ one can do a perturbative calculation, whereas for larger values of D_{gi} one will have to do a nonperturbative calculation. We have shown here the results of both types of calculations, perturbative and nonperturbative.

RESULTS AND DISCUSSION

In Fig. 2 we have shown the rate of the gain $(d\mathcal{G}/dt)$ profile as a function of detuning from the AI state, in the weak-field limit. The intensity of the coherent field is $I = 6.4 \times 10^9$ W/cm^2 and the intensities for the lower and upper probes are 9.6×10^4 and 9.6×10^7 W/cm^2, respectively. In this situation we have also calculated the population in states $|g\rangle$ and $|a\rangle$ as a function of time to show that the population difference reaches a limiting value in the long-time limit and no population inversion occurs with the evolution of the system (Fig. 3). To obtain gain under this condition, probe fields need not be short-pulsed. At the lower intensity of the coherent field, 6.4×10^7 W/cm^2, the gain can be obtained at a higher value of intensity 9.6×10^6 W/cm^2 for the lower probe field and both probe fields have to be pulsed at a duration of 3.45 ps. The results of nonperturba-

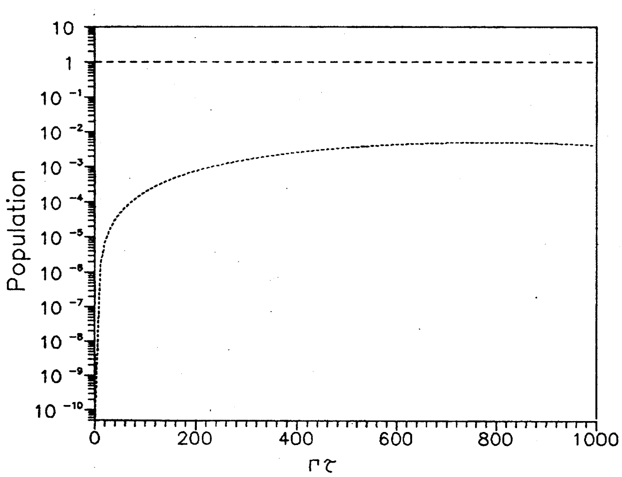

FIG. 3. Population of the lower and the upper lasing level as a function of time. ––––, population of the ground $X\,^1\Sigma_g$ $(v=0, j=1)$ level; —, population of the lowest AI level of $^1\Sigma_g$ symmetry.

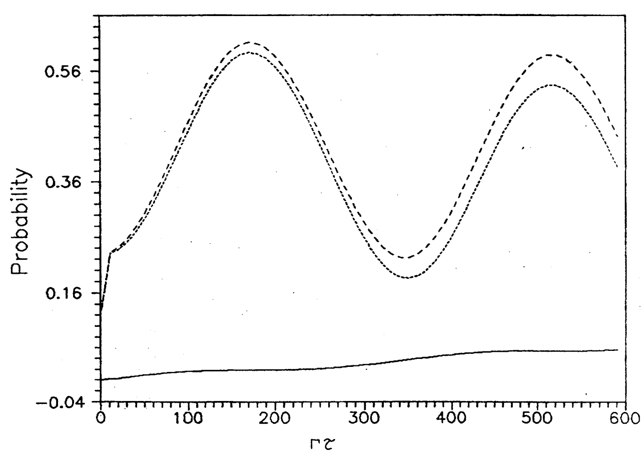

FIG. 4. Absorption, emission, and gain probabilities as a function of time; $\Gamma\tau = 100$ corresponds to $\tau = 3.45$ ps. —, absorption probability; –––, emission probability; ···, gain. Both incoherent and coherent pumping are present. The coherent field intensity is 6.4×10^7 W/cm^2, the upper probe intensity of wavelength 315 nm is 9.6×10^7 W/cm^2, and the lower probe field strength is 9.6×10^6 W/cm^2.

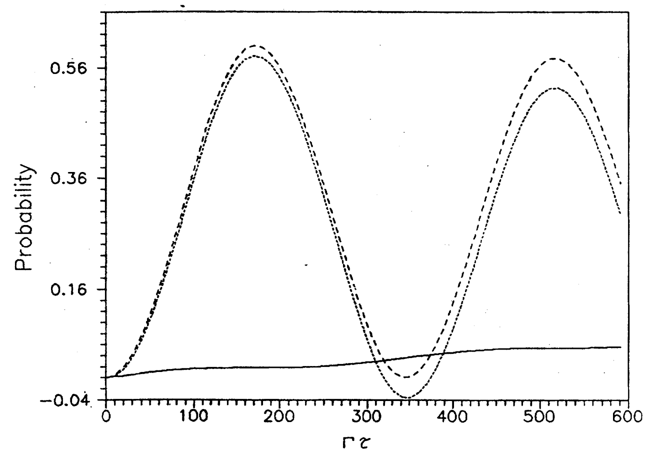

FIG. 5. Same as in Fig. 4, but in the absence of incoherent pumping.

tive calculations have been shown in Figs. 4–8. In the presence of incoherent pumping in the short-time region $\Gamma\tau<10$, gain arises mainly due to the single-photon emission to the $B\ ^1\Sigma_u$ state and the gain on both probe fields evolves with an increase in time (Fig. 4). This feature has already been shown in a parametric calculation [11]. It may be noticed that the sharp kink occurring for smaller values of $\Gamma\tau<10$ in Fig. 4 does not appear for coherent pumping (Fig. 5). In Fig. 6 we have shown the evolution of populations in the ground and in the AI state, respectively. The populations are found to oscillate completely out of phase with time. Hence the system oscillates between noninversion and inversion alternately. The gain curves in Figs. 4 and 5 show this oscillatory structure as a function of time without and with inversion alternately. It is found that within the first quarter period of noninversion (Fig. 6) gain persists up to a time given by $\tau=3.45$ ps, after which gain switches over to lasing with population inversion. We have shown here the gain profile as a function of detuning from the AI state at a value of

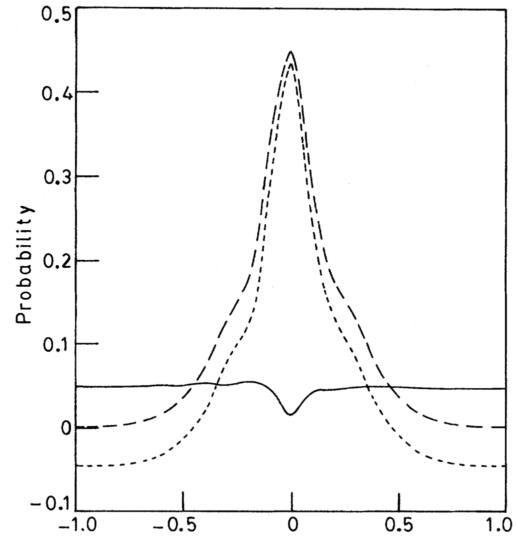

FIG. 7. Absorption, emission, and gain profile as a function of detuning from the AI state $[\epsilon=\delta/(\Gamma/2)]$ $\Gamma\tau=95$. The probe and coherent intensities are the same as in Fig. 4.

time given by $\Gamma\tau=95$, i.e., for a pulse duration of 3.2 ps (Fig. 7), where both pumping mechanisms are present. A comparison of this profile with the profile at a smaller intensity of the lower probe field (Fig. 2) shows that an increase in the intensity of the lower probe field leads to a broadening of the gain profile. Throughout this calculation we have considered the square pulse, i.e., during the evolution of the system the probe field intensities do not change. In addition, we have repeated the calculation for a coherent field intensity 6.4×10^9 W/cm^2; probe field intensities have been chosen to be the same as in Figs. 4 and 5, using the probe pulse shape as $I(t)=I_0\ \mathrm{sech}^2(t/T)$, where T is the pulse duration. We have shown the evolution of the gain for a square pulse and a $\mathrm{sech}^2(t/T)$ pulse, with $T=100/\Gamma$ a.u.$=3.45$ ps (Fig. 8). It

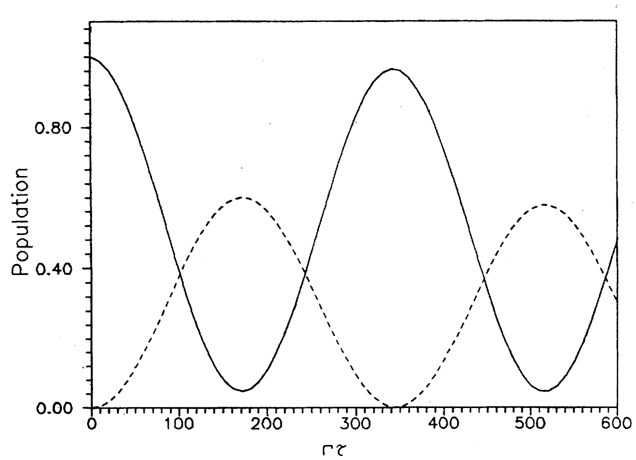

FIG. 6. Population of the AI state as a function of time. —, population of the $X\ ^1\Sigma_g$ ($v=0,j=1$) level; ----, population of the lower AI state of $^1\Sigma_g$ symmetry. The field intensities of coherent and probe fields are the same as in Fig. 4.

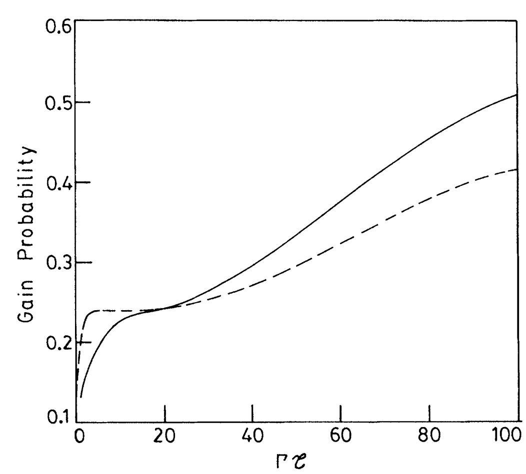

FIG. 8. Time evolution of gain $G(t)$: —, for a square pulse; ----, for a $\mathrm{sech}^2(t/T)$ pulse. The field intensities are the same as in Fig. 4. Both coherent and incoherent pumping are present.

is found that the evolution of the gain for a square pulse is much faster than in the case of a $\text{sech}^2(t/T)$ pulse shape.

In conclusion, we have shown that in the presence of a coherent pumping field of intensity in the range 6.4×10^7–6.4×10^9 W/cm^2, $(1+1)$-photon lasing with or without population inversion can be obtained in the H_2 molecule when the probe fields are of wavelengths around 105 nm [connecting $X\ ^1\Sigma_g\ (v=0,j=1)\rightleftharpoons B\ ^1\Sigma_u\ (v=4,j=2)$ levels] and 315 nm [connecting $B\ ^1\Sigma_u\ (v=4,j=2)\rightleftharpoons{}^1\Sigma_g$ states]. The optimum intensity of the upper probe field is 9.6×10^7 W/cm^2 and that of the lower probe field can be varied from 9.6×10^4 to 9.6×10^6 W/cm^2, depending upon the strength of the coherent pumping. Results of the square-shaped pulse and the $\text{sech}^2(t/T)$-shaped pulse have been compared.

Note added. After submission of this paper, we have come to learn that a recent paper by Walmsley *et al.* [18] has demonstrated gain without inversion in a molecular system by using femtosecond pump and probe pulses.

ACKNOWLEDGMENTS

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