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EFFECT OF THE LASER TRANSVERSE SPATIAL DISTRIBUTION ON MULTIPHOTON IONIZATION PROCESSES*

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In this paper, the dependence of the photoionization cross section σ_2, total ion number N_i and the FWHM of photoionization spectral line in resonant multiphoton ionization processes (RMPI) upon the parameter C of the laser transverse spatial distribution has been studied. Considering the effects of spatial coordinate on the resonant multiphoton absorption cross section σ_1, the limited dimension of the laser-molecular interaction volume and threshold of laser intensity for RMPI, we deduce the expression of σ_2 *vs* the parameter C. We calculated the value of σ_2 for (2+2)PI of benzene by numerical method, and compared the result with that obtained by Bruzzeste *et al.* In additon, we also present the total ion number of photoionization and the FWHM of photoionization spectral line as a function of the parameter C.

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I. INTRODUCTION

The resonant multiphoton ionization (RMPI) technique has been widely used in studying the higher excited states of atoms and molecules and has obtained numerous results. When a laser beam is used as the excitation source of multiphoton ionization (MPI), its transverse spatial distribution will affect the MPI process, but little attention has been given in this field. For the first time Aymer *et al.*[1] and Armenante *et al.*[2] have studied the effect of gaussain field on the MPI cross section, and then Bruzzese *et al.*[3] has further studied the effect of spatial distribution of two transverse modes on inoization cross section for resonant 2-photon ionization [(2+2)PI] of benzene. However, they all ignored the relation between the resonant absorption cross section and the spatial coordinates, i. e., they considered the resonant absorption cross section as a constant, which was independent of spatial coordinates. Meanwhile, they did not consider the spatial restriction of laser-molecule interaction and the intensity threshold of RMPI. In fact, RMPI cross section greatly depends on these factors. To describe precisely the effects of laser transverse spatial distribution on MPI cross section, by considering these factors and ac-stark frequency shift, we derived the expressions of the 2-photon ionization cross section σ_2 and the ion number

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N_i versus the laser transverse distribution parameter C from the rate equation of (2+2)PI, and calculated the dependence of σ_2 upon C for benzene by numerical method. Then we compared this result with that of Bruzzese *et al.* In addition, we present the dependence of the total ion number N_i and the width of MPI spectral line upon the parameter C.

II. THE IONIZATION RATE OF (2+2)PI

We consider a molecule which is ionized by two-photon-resonant four-photon absorption: the ground state $|0>$ is coupled to the resonant excited state $|1>$ by a 2-photon transition, while $|1>$ is ionized by the absorption of two additional photons. When the excitation source is a laser beam with a finite-bandwidth, the expression of ionization rate can be written as[4]

$$P_i = 1 - \frac{1}{2}(1 + \frac{A}{B})\exp[(A-B)t] - \frac{1}{2}(1 - \frac{1}{2})\exp[(A+B)t], \tag{1}$$

where

$$A = W^{(a)} + \gamma_1 + \kappa, \tag{2}$$

$$B = [W^{(a)^2} + (\gamma_1 + \kappa)^2]^{1/2}. \tag{3}$$

In the two formulas above, γ_1 is the ionization rate of state $|1>$, κ is the total other relaxation rate from the intermediate state $|1>$, $W^{(a)}$ is the two-photon resonant absorption rate. Let Γ_L represent the laser line width and Ω represent the Rabi fequency of the 2-photon resonant absorption, which is proportional to exciting light intensity[5]. When $\Gamma_L \gg \Omega$, γ_1, $W^{(a)}$ can be expressed as[4]

$$W^{(a)} = \Omega^2 \frac{2\Gamma_L + \gamma_1 + \kappa}{(\Delta_0 - \delta\omega_{10})^2 + (2\Gamma_L + \gamma_1 + \kappa)^2}, \tag{4}$$

where $\Delta_0 = 2\omega - \omega_{10}$ is the detuning of 2-photon resonance, ω is the exciting frequency, ω_{10} is the resonant frequency between states $|0>$ and $|1>$, $\delta\omega_{10}$ is the difference of ac-stark frequency between state $|0>$ and state $|1>$, which is proportional to exciting light intensity[5, 6].

From Eq.(1) the (2+2)PI rate is obtained as

$$W^{(i)} = \frac{d}{dt} P_i = \frac{A^2 + B^2}{2B} \{\exp[-(A-B)t] - \exp[-(A+B)t]\}. \tag{5}$$

Let σ_1 denote the 2-photon resonant absorption cross section, σ_2 denote the 2-photon ionzation cross section,

$$\sigma_1 I^2 = W^{(a)}, \tag{6}$$

$$\sigma_2 I^2 = \gamma_1. \tag{7}$$

We have

$$\sigma_1 = (\Omega/I)^2 \frac{2\Gamma_L + \gamma_1 + \kappa}{(\Delta_0 - \delta\omega_{10})^2 + (2\Gamma_L + \gamma_1 + \kappa)^2}. \tag{8}$$

Because $\Omega \propto I$, Ω/I is a constant and let $K=\Omega/I$, so Eq.(8) can be written as

$$\sigma_1 = K^2 \frac{2\Gamma_L + \gamma_1 + \kappa}{(\Delta_0 - \delta\omega_{10})^2 + (2\Gamma_L + \gamma_1 + \kappa)^2} \tag{9}$$

and

$$\sigma_2 = \gamma_1 / I^2.$$

When the laser intensity is high enough, typical value $I \simeq 10^{29}$ photon $\cdot$ cm$^{-2} \cdot$ s^{-1}, RMPI is saturated, the population of state $|1>$ is constant, we have

$$\sigma_1 I^2 n_0 \simeq (\sigma_2 I^2 + \kappa) n_1, \tag{10}$$

where n_0 and n_1 represent the populations of states $|0>$ and $|1>$, respectively. Under this condition, $\sigma_2 I^2 \simeq \kappa$[2], and in the case of stationary, $n_0 \gg n_1$, so that

$$\sigma_1 I^2 \ll \sigma_2 I^2 \text{ and } \sigma_1 I^2 \ll \kappa. \tag{11}$$

From Eqs.(2) and (3), we obtain $A \simeq B = (\sigma_2 I^2 + \kappa)$, Eq.(5) can be approximately written as

$$W^{(i)} = \frac{A^2 - B^2}{2B} \{1 - \exp[-(A+B)t]\} = \sigma_1 I^2 [1 - \exp[-(A+B)t]]. \tag{12}$$

Generally, the magnitude of κ is 10^{11} s^{-1}[2, 3], when the duration of laser pulse is nanosecond, $(A+B)t \gg 1$, so Eq.(12) reduces to

$$W^{(i)} = \frac{2\sigma_1 \sigma_2 I^4}{\sigma_2 I^2 + \kappa}. \tag{13}$$

In the case of weak intensity, or the lifetime of state $|1>$ is long, the RMPI is generally unsaturatied. When the time of laser-molecule interaction is less than the lifetime of state $|1>$, $(A+B)t < 1$[7]. We develop the exponent in Eq.(5) as a series and omit the second and higher-order terms, then we have

$$\begin{aligned} W^{(i)} &= \frac{A^2 - B^2}{2B} [1 - (A-B)t + 1 + (A+B)t] \\ &= (A^2 - B^2)t = 2\sigma_1 I^2 (\sigma_2 I^2 + \kappa) t \\ &= 4\sigma_1 \sigma_2 I^4 t. \end{aligned} \tag{14}$$

III. THE DEPENDENCE OF RMPI ON LASER TRANSVERSE SPATIAL DISTRIBUTION

The most common laser transverse spatial distribution is the combination of a pure gaussian mode (0, 0) and the first order transverse mode (0, 1), the intensity distribution is expressed in cylindrical coordinate (z, ρ) as

$$I(z, \rho) = \frac{2W_0}{DW^2} I_0 e^{-x} [1 + C(1-x)]^2, \tag{15}$$

where D is the normalizing constant, I_0 is the peak of the pulse laser, C is the coupling coefficient of laser transverse mode. $C=0$ implies gaussian field. In Eq.(15) $x=2\rho^2/W^2$, $W^2=W_0^2(1+z^2/z_0^2)$, $z_0=\dfrac{\pi W_0^2}{\lambda}$; where W_0 is the beam waist, z_0 is the intrinsic coherent length of laser. For simplifying, in Eq.(15) we assume the phases of two modes to be the same. The effects of fluctuation of phases on the line width of the laser beam has been included in Γ_L of Eq.(4).

A. The expression of resonant absorption cross section

In the study of the effects of laser transverse distribution on RMPI, Bruzzese *et al.*[2,3] take the resonant absorption cross section σ_1 as a constant, which is independent of spatial coordinates. But according to the results of Eqs.(8) and (15), σ_1 should be the function of spatial coordinates. Substituting Eq.(15) into Eq.(9), we obtain the expression of σ_1,

$$\sigma_1(z,\rho)=\frac{K^2\{2\Gamma_L+k+\dfrac{4\sigma_2}{D^2}I_0^2e^{-2x}[1+C(1-x)]^4/(1+\dfrac{z^2}{z_0^2})^2\}}{\{\Delta_0-\dfrac{2}{D}(\delta\omega_{10})e^{-x}[1+C(1-x)]^2/(1+\dfrac{z^2}{z_0^2})\}^2+\{4\Gamma_L+\kappa+\dfrac{4\sigma_2}{D^2}I_0^2e^{-2x}[1+C(1-x)]^4/(1+\dfrac{z^2}{z_0^2})^2\}^2}, \tag{16}$$

where $(\delta\omega_{10})_0$ is the ac-stark frequency shift corresponding to intensity I_0. From Eq.(16), we can see that σ_1 is not only the function of spatial coordinate, but also the function of laser line width Γ_L, the detuning quantity of resonant frequency Δ_0, ac-stark frequency shift $\delta\omega_{10}$, and laser transverse spatial distribution parameter C.

B. The expression of photoionization rate

Substituting Eqs. (15) and (16) into Eq. (13), we obtain the expression of photoionization rate under saturation case

$$W^{(i)}(z,\rho)=\frac{2\sigma_2K^2\{2\Gamma_L+\kappa+\dfrac{4\sigma_2}{D^2}I_0^2e^{-2x}[1+C(1-x)]^4/(1+\dfrac{z^2}{z_0^2})^2\}}{\{\Delta_0-\dfrac{2}{D}(\delta\omega_{10})_0e^{-x}[1-C(1-x)]^2/(1+\dfrac{z^2}{z_0^2})\}^2+\{2\Gamma_L+\kappa+\dfrac{4\sigma_2}{D^2}I_0^2e^{-2x}[1+C(1-x)]^4/(1+\dfrac{z^2}{z_0^2})^2\}}$$
$$\times\frac{\dfrac{16}{D^4}I_0^4e^{-4x}[1+C(1-x)]^8/(1+\dfrac{z^2}{z_0^2})^4}{\dfrac{4\sigma_2}{D^2}I_0^2e^{-2x}[1+C(1-x)]^4/(1+\dfrac{z^2}{z_0^2})^2+\kappa}. \tag{17}$$

Substituting Eqs. (15) and (16) into Eq. (14), we obtain the expression of photonionization rate under unsaturation case

$$W^{(i)}(z,\rho)$$
$$=\frac{2\sigma_2K^2\{2\Gamma_L+\kappa+\frac{4\sigma_2}{D^2}I_0^2e^{-2x}[1+C(1-x)]^4/(1+\frac{z^2}{z_0^2})^2\}\{\frac{16}{D^4}I_0^4e^{-4x}[1+C(1-x)]^8/(1+\frac{z^2}{z_0^2})^4\}}{\{\Delta_0-\frac{2}{D}(\delta\omega_{10})_0e^{-x}[1-C(1-x)]^2/(1+\frac{z^2}{z_0^2})\}^2+\{2\Gamma_L+\kappa+\frac{4\sigma_2}{D^2}I_0^2e^{-2x}[1+C(1-x)]^4/(1+\frac{z^2}{z_0^2})^2\}^2} \tag{18}$$

Because $W^{(i)}$ is the function of σ_1, whether it is in the case of saturation or unsaturation, $W^{(i)}$ is not only the function of spatial coordinate, but also the function of laser line width, detuning of resonant frequency, ac-stark frequency shift and the transverse distribution.

C. The dependence of PI ion number on C

When the excitation laser is a rectangular pulse with width T, the total ion number in the RMPI process is

$$N_i=n\int_0^T dt\int_v W^{(i)}(\boldsymbol{r})d^3\boldsymbol{r}, \tag{19}$$

where n is the particle number density, v is the laser-molecule interaction volume. Let l and l' represent the longitudinal and the radial limits of integration respectively. So, the laser-molecule interaction volume is confined in longitudinal interval $[-\frac{l}{2},\ \frac{l}{2}]$, and in radial interval $[0,\ l']$. While the laser intensity decreases approximately in accordance with $e^{-2\rho^2}/W_0^2$. An intensity threshold is required for realizing RMPI, we let I_{th} represent this value. As the threshold condition is satisfied, the corresponding radial coordinate $\rho_0\simeq W_0\cdot[\ln(I/I_{th})]^2$. Generally, the beam waist W_0 is small, I_0 is only slightly larger than I_{th}, so ρ_0 is smaller than l'. Equation.(19) can be written as

$$N_i=2\pi n\int_0^T dt\int_{-\frac{l}{2}}^{\frac{l}{2}}dz\int_0^{\rho_0}W^{(i)}(\boldsymbol{r})\rho d\rho\ . \tag{20}$$

1. *The saturation case.* In the case of saturation, $W^{(i)}$ is independent of time t, Eq.(20) can be written as

$$N_i=2\pi nT\int_{\frac{-l}{2}}^{\frac{l}{2}}dz\int_0^{\rho_0}W^{(i)}(z,\rho)\rho d\rho. \tag{21}$$

Let τ_e be the characteristic time of laser phase fluctuation and τ_i be the characteristic time of photoionization of molecules. We will discuss the effects of laser transverse distribution on photoionization for two limit conditions—— $\tau_i\gg\tau_e$ and $\tau_i\ll\tau_e$:

(a) When $\tau_i\gg\tau_e$, because $\Gamma_L\gg\sigma_2I^2$, κ, so Eq.(17) reduces to

$$W^{(i)}(z,\rho)=\frac{4\sigma_2K^2\Gamma_L}{\{\Delta_0-\frac{2}{D}(\delta\omega_{10})_0e^{-x}[1+C(1-x)]^2/(1+\frac{z^2}{z_0^2})\}^2+4\Gamma_L^2}\times\frac{\frac{16}{D^4}I_0^4e^{-4x}[1+C(1-x)]^8/(1+\frac{z^2}{z_0^2})^4}{\frac{4\sigma_2}{D^2}I_0^2e^{-2x}[1+C(1-x)]^4/(1+\frac{z^2}{z_0^2})^2+\kappa}. \tag{22}$$

Substituting Eq.(22) into Eq.(21), and let $\zeta=\frac{z}{z_0}$, $\xi=e^{-2x}$. After a series of calculations, we have

$$N_i=\frac{1}{4}n\lambda z_0^2T\int_{-\zeta_0}^{+\zeta_0}d\zeta\int_{\xi_0}^{1}\frac{1+\zeta^2}{\xi}W^i(\xi,\zeta)d\xi$$

$$=4n\lambda z_0^2\sigma_2TK^2\Gamma_L(\frac{I_0}{D^2})\int_{-\zeta_0}^{+\zeta_0}d\zeta\int_{\xi_0}^{1}\frac{1}{\{\Delta_0-\frac{2}{D}(\delta\omega_{10})_0\frac{\sqrt{\xi}}{1+\zeta^2}[1+C(1+\frac{1}{2}\ln\xi)]^2\}^2+4\Gamma_L^2}$$

$$\times\frac{\xi[1+C(1+\frac{1}{2}\ln\xi)]^8/(1+\zeta^2)^3}{\frac{\xi}{(1+\zeta^2)^2}[1+C(1+\frac{1}{2}\ln\xi)]^4+\frac{\kappa D^2}{4\sigma_2I_0^2}}d\xi, \tag{23}$$

where $\zeta_0=\frac{1}{2z_0}$, $\xi_0=e^{-2x_0}=e^{-4\rho_0^2/W_0^2}=(\ln\frac{I_{th}}{I_0})^2$.

(b) When $\tau_i\ll\ _e$, because $\Gamma_L\ll\sigma_2I^2$, κ, Γ_L can be omitted in the Eq.(17), thus Eq.(17) can be written as

$$W^{(i)}(z,\rho)$$

$$=2\sigma_2K^2\frac{\frac{16}{D^4}I_0^4e^{-4x}[1+C(1-x)]^8/(1+\frac{z^2}{z_0^2})^4}{\{\Delta_0-\frac{2}{D}(\delta\omega_{10})_0e^{-x}[1+C(1-x)]^2/(1+\frac{z^2}{z_0^2})\}^2+\{\kappa+\frac{4\sigma_2}{D^2}I_0^2e^{-2x}[1+C(1-x)]^4/(1+\frac{z^2}{z_0^2})^2\}^2}. \tag{24}$$

Substituting Eq.(24) into Eq.(21), after the same calculations as in case (a), we obtain

$$N_i=8n\lambda z_0^2\sigma_2TK^2(\frac{I_0}{D})^4\int_{-\zeta_0}^{+\zeta_0}d\zeta\int_{\xi_0}^{1}d\xi$$

$$\times\frac{\xi^2[1+C(1+\frac{1}{2}\ln\xi)]^8/(1+\zeta^2)^3}{\{\Delta_0-\frac{2}{D}(\delta\omega_{10})_0\sqrt{\xi}[1+C(1+\frac{1}{2}\ln\xi)]^2/(1+\zeta^2)\}^2+\{\kappa+\frac{4\sigma_2}{D^2}I_0^2\xi\frac{[1+C(1+\frac{1}{2}\ln\xi)]^4}{(1+\zeta^2)^2}\}^2}. \tag{25}$$

2. *The unsaturation case.*

(a) When $\tau_i\gg\tau_e$, and also under $\Gamma_L\gg\sigma_2I^2$, κ, Eq.(18) reduces to

$$W^{(i)}(z,\rho)=8\sigma_2K^2\Gamma_L\frac{\frac{16}{D^4}I_0^4e^{-4x}[1+C(1-x)]^8/(1+\frac{z^2}{z_0^2})^4}{\{\Delta_0-\frac{2}{D}(\delta\omega_{10})_0e^{-x}[1+C(1-x)]^2/(1+\frac{z^2}{z_0^2})\}^2+4\Gamma_L^2}\times t. \tag{26}$$

Take the same variable substitution as in case 1, then

$$N_i=16n\lambda z_0^2\sigma_2K^2T^2\Gamma_L\int_{-\zeta_0}^{\zeta_0}d\zeta\int_{\xi_0}^{1}d\xi\times\frac{\xi[1+C(1+\frac{1}{2}\ln\xi)]^8(1+\zeta^2)^3}{\{\Delta_0-\frac{2}{D}(\delta\omega_{10})_0\sqrt{\xi}[1+C(1+\frac{1}{2}\ln\xi)]^2/(1+\zeta^2)\}^2+4\Gamma_L^2}, \tag{27}$$

where $\zeta_0=\frac{1}{2z_0}$, $\xi_0=\exp(-\rho_0^2/W_0^2)\simeq I_{th}/I_0$.

(b) As $\tau_i\ll\tau_e$, Γ_L can be omitted, then Eq.(18) reduces to

$$W^{(i)}(z,\rho)$$

$$=4\sigma_2K^2\frac{\{\kappa+\frac{4\sigma_2}{D^2}I_0^2e^{-2x}[1+C(1-x)]^4/(1+\frac{z^2}{z_0^2})^2\}\{\frac{16}{D^4}I^4e^{-4x}[1+C(1-x)]^8/(1+\frac{z^2}{z_0^2})^4\}t}{\{\Delta_0-\frac{2}{D}(\delta\omega_{10})e^{-x}[1+C(1-x)]^2/(1+\frac{z^2}{z_0^2})\}^2+\{\kappa+\frac{4\sigma_2}{D^2}I_0^2e^{-2x}[1+C(1-x)]^4/(1+\frac{z^2}{z_0^2})^2\}^2} \tag{28}$$

Substituting Eq. (28) into Eq. (20), weobtained finally the expression of the total ion number

$$N_i=8n\lambda z_0^2\sigma_2K^2T^2(\frac{I_0}{D})^4\int_{-\zeta_0}^{+\zeta_0}d\zeta\int_{\xi_0}^{1}d\xi$$

$$\times\frac{\xi[1+C(1+\frac{1}{2}\ln\xi)]^8/(1+\zeta^2)^3\times\{\kappa+\frac{4\sigma_2}{D^2}I_0^2\xi[1+C(1+\frac{1}{2}\ln\xi)]^4(1+\zeta^2)^2\}}{\{\Delta_0-\frac{2}{D}(\delta\omega_{10})_0\frac{\sqrt{\xi}}{1+\zeta^2}[1+C(1+\frac{1}{2}\ln\xi)]^2\}^2+\{\kappa+\frac{4\sigma_2}{D^2}I_0^2\frac{\xi}{(1+\zeta^2)^2}[1+C(1+\frac{1}{2}\ln\xi]^4\}^2} \tag{29}$$

IV. NUMERICAL CALCULATION

In order to analyze and compare with the previous works, we conduct numerical computation for the dependence of MPI cross section σ_2, the total ion number N_i, and the width (FWHM) of ionization spectral line upon the laser transverse spatial distribution with computer.

A. Calculation method and the choice of parameters

We take the experimental data from Ref. [2] for (2+2) PIof benzene as follows: excitation wavelength $\lambda=4716$Å, laser line width $\Gamma_L=1\,cm^{-1}(3\times10^{10}\,s^{-1})$, pulse duration $T=10^{-8}$ s, $I_0=1.5\times10^{29}$ photon · cm^{-2} · s^{-1}, $z_0=0.01$ cm, molecular density $n=3.2\times10^6$

cm^{-3}, $K^2 \simeq 10^{-40}\ cm^4$, $\kappa \simeq 1 \times 10^{11}\ s^{-1}$ for $^1B_{2u}$ state of benzene. We take the integral limit $l = 0.2$ cm, $\rho_0 \simeq 10^{-3}$ cm. Because the ac-stark frequency shift $(\delta\omega_{10})_0$ of $^1B_{2u}$ state of benzene has not been reported, so we take the parameter of NO instead of that of benzene, i.e., $(\delta\omega_{10})_0 = 1.75\ cm^{-1}$[8].

B. The dependence of two photon ionization cross section on parameter C for (2+2)PI

A least square fit of Eqs.(23) and (25) to the N_i-I_0 experimental curve yields 2-photon ionization cross section σ_2 for a given C. Hence, taking different C values, we obtain the curve of σ_2 versus C.

In the case of saturation, as $\tau_i \gg \tau_e$, for the strictly resonant and two detuning cases, the calculated results of σ_2 vs C are shown in Fig.1. When σ_1 is constant (take $\sigma_1 = 10^{-51}$ $cm^4 \cdot s$[2]), the calculated results of σ_2 vs C are shown in Fig.2. When $\tau_i \ll \tau_e$, the calculated

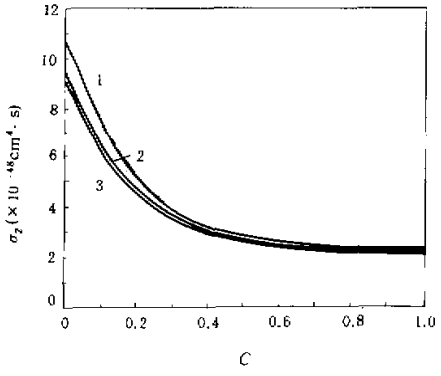

Fig. 1. The 2-photon ionization cross section σ_2 as a function of the parameter C, in the saturtion case and $\tau_i \gg \tau_e$, 1, $\Delta_0 = 1.75\ cm^{-1}$; 2, $\Delta_0 = 0$; 3, $\Delta_0 = -1.75\ cm^{-1}$.

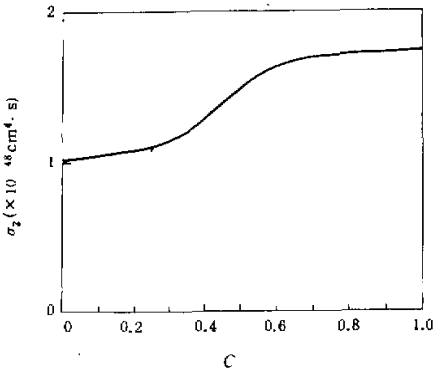

Fig. 2. The 2-photon ionization cross section σ_2 as a function of C when σ_1 is constant and $\Delta_0 = 0$, in the saturation case and $\tau_i \gg \tau_e$.

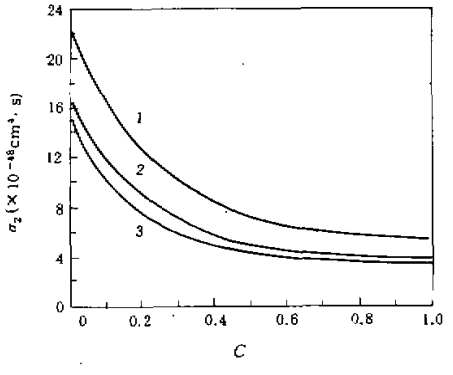

Fig. 3. The 2-photon ionization cross section σ_2 as a function of C, in the saturation case and $\tau_i \ll \tau_e$. 1, $\Delta_0 = 1.75\ cm^{-1}$; 2, $\Delta_0 = 0$; 3, $\Delta_0 = -1.75\ cm^{-1}$.

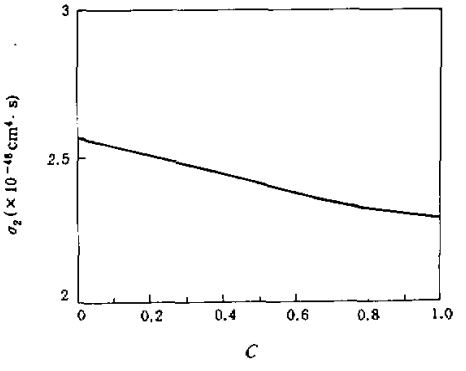

Fig.4. The 2-photon ionization cross section σ_2 as a function of C when when σ_1 is constant and $\Delta_0=0$ in the saturation case and $\tau_i \ll \tau_e$.

results of σ_2 vs C are shown in Fig.3. When σ_1 is a constant, the results are shown in Fig.4.

C. The dependence of the total ion number N_i on C for (2+2)PI

In the case of saturation, for $\tau_i \gg \tau_e$ the curve of N_i versus C corresponding to different Δ_0 are shown in Fig.5 and that for $\tau \ll \tau$ are shown in Fig.6.

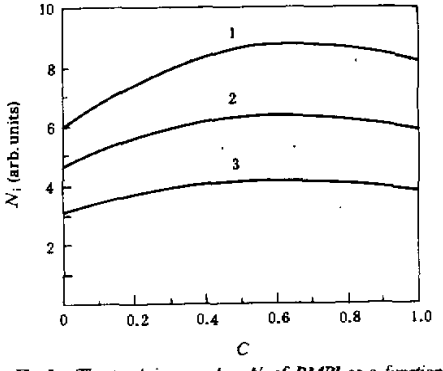

Fig. 5. The total ion number N_i of RMPI as a function of C, in the saturation case and $\tau_i \gg \tau_e$. 1, $\Delta_0=1.75\,cm^{-1}$; 2, $\Delta_0=0$; 3, $\Delta_0=-1.75\,cm^{-1}$.

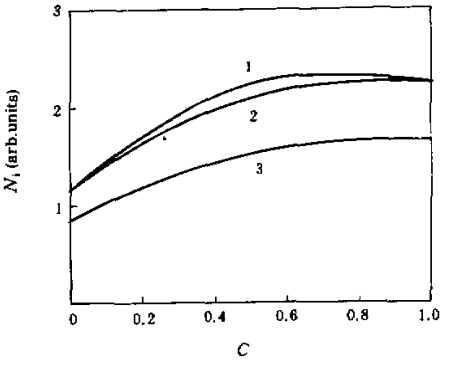

Fig. 6. The total ion number N_i of RMPI as a function of C, in the saturation case and $\tau_i \ll \tau_e$. 1, $\Delta_0=1.75\,cm^{-1}$. 2, $\Delta_0=3$; 3, $\Delta_0=-1.75\,cm^{-1}$.

From Fig.5 and Fig.6, it can be found that whether $\tau_i \gg \tau_e$ or $\tau_i \ll \tau_e$, N_i exhibits extreme on each curve.

In the case of unsaturation, the forms of the dependence of σ_2 and N_i on C are similar to those in the saturation case, the results are not given here.

D. The dependence of the width (FWHM) of ionization spectral line upon C for (2+2)PI

The line width in RMPI can be defined as

$$N_i(\omega_0 \pm \frac{1}{2}\mathrm{FWHM}) = \frac{1}{2}N_i(\omega_0), \tag{30}$$

where ω_0 is the frequency corresponding to the peak of ionization spectral line.

Because N_i is a function of the detuning Δ_0 and the laser transverse distribution parameter C, when C is given, we can get the curve of N_i versus Δ_0. Let the maximum value of N_i is denoted by $(N_i)_{max}$ and that of the detuing Δ_0 corresponding to $(N_i)_{max}$ denoted by $(\Delta_0)_{max}$. Taking two values of Δ_{01} and Δ_{02} on both sides of $(\Delta_0)_{max}$ respectively, which satisfy the condition, $N_{i1} = N_{i2} = \frac{1}{2}(N_i)_{max}$, we obtain

$$\mathrm{FWHM} = |\Delta_{01} - \Delta_{02}|. \tag{31}$$

Thus taking C as a variable, we get the relation between FWHM and C. In the case of saturation, as $\tau_i \gg \tau_e$ and $\tau_i \ll \tau_e$, the results are shown in Fig.7. In the case of unsaturation, the results are shown in Fig.8. From Fig.8, it can be found that under the condition of unsaturation FWHM unvaries with C.

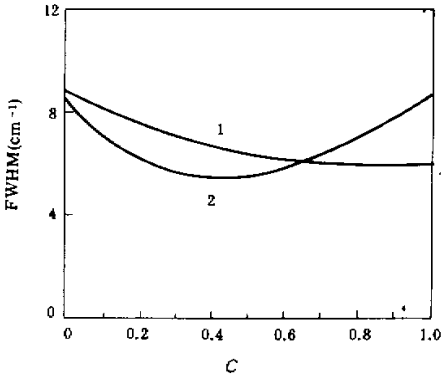

Fig.7. FWHM of photoionization spectral line as a function of C in the saturation case. 1, $\tau_i \gg \tau_e$; 2, $\tau_i \ll \tau_e$.

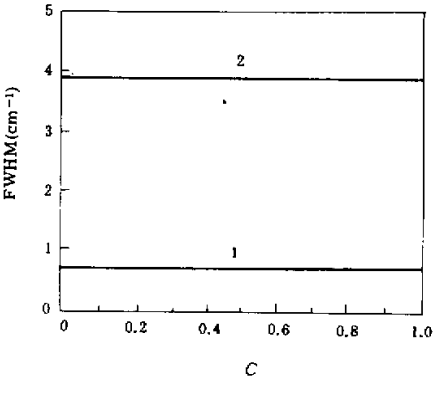

Fig.8. FWHM of photoionization spectral line as a function of C in the unsaturation case. 1, $\tau_i \gg \tau_e$, 2, $\tau_i \ll \tau_e$.

V. CONCLUSIONS

(1) From Figs.1 and 3, it can be found that, in the case of saturation, whether $\tau_i \gg \tau_e$ or $\tau_i \ll \tau_e$ the ionization cross section σ_2 for (2+2)PI decreases with the increase of the parameter C, and the detuning Δ_0 only affects the absolute value of σ_2, but does not change the tendency of σ_2 dependence of C. For a given value of C, when Δ_0 is positive, σ_2 is slightly greater than that at exact resonance. Assuming 2-photon absorption cross section σ_1 is a constant, i.e. σ_1 is independent of the spatial coordinates, we obtain Figs.2 and 4, which show that σ_2 increases with the increase of C for $\tau_i \gg \tau_e$ and that σ_2 slightly decreases with the increase of C for $\tau_i \ll \tau_e$. These results agree with that of Bruzzese[3]. When σ_1 is a function of spatial coordinates, the tendency of σ_2 versus C is completely different.

Thus, in the process of RMPI the dependence of σ_1 on spatial coordinates cannot be ignored.

(2) In the process of (2+2)PI, total ionization ion number N_i depends on the laser transverse spatial distribution parameter C, and it exhibits a maximum at any detuning Δ_0, as shown in Figs.5 and 6. These results have never been reported before.

(3) From the computation, the dependence of the FWHM of RMPI on C is found to be quite different between the saturation case and the unsaturation case. In the saturation case, when $\tau_i \gg \tau_e$, the FWHM shows a decreasing tendency for $C<0.4$ and it is invariable for $C>0.4$. When $\tau_i \ll \tau_e$, the FWHM exhibits a minimum at $C=0.4$, while the width of which is nearly half as wide as that of the widest one. In the unsaturation case, the FWHM is independent of the laser transverse spatial distribution.

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