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DOUBLE PHOTON ELECTRIC DIPOLE AND QUADRUPOLE ABSORPTION OF ATOMIC HYDROGEN *

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ABSTRACT

The two photon processes with one electric dipole photon and another electric quadrupole photon are investigated. These processes with selection rules different from two-electric dipole photon process may be tested by experiments with high power lasers.

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1. Introduction

With the high power laser techniques, it is possible to study the excited states of atoms and molecules by two photon spectroscopy. The two photon processes of electric dipole photon were calculated by many authors [1]. In this paper we restricted our investigation to one dipole and one quadrupole photon absorption, which is slightly smaller than the two electric dipole photon process.

2. Theory of one electric dipole and one electric quadrupole photon absorption

The incoming beams are two plane polarized laser beams with the vector potential

$$\vec{A} = \frac{1}{\sqrt{2\omega\Omega}}\,\vec{e}\,e^{i\vec{k}\cdot\vec{r}} \tag{1}$$

$$\vec{A}' = \frac{1}{\sqrt{2\omega'\Omega}}\,\vec{e}'\,e^{i\vec{k}'\cdot\vec{r}}$$

We now study the two photon absorption of electric dipole and quadrupole, these two photons are from two beams respectively.

The incoming plane polarized laser wave may be expressed as some spherical waves with different parities.

$$\vec{e}\,e^{i\vec{k}\vec{r}} = \sum_{L,M;\lambda=0,1} \vec{e}\cdot\vec{Y}^{(\lambda)*}_{LM}\left(\frac{\vec{k}}{\omega}\right)\vec{a}^{(\lambda)}_{LM} \tag{2}$$

$$\vec{a}^{(1)}_{LM} = \sqrt{\frac{L}{2L+1}}\,g_{L+1}(\omega r)\,\vec{Y}_{L,L+1,M}\left(\frac{\vec{r}}{r}\right) + \sqrt{\frac{L+1}{2L+1}}\,g_{L-1}(\omega r)\,\vec{Y}_{L,L-1,M}\left(\frac{\vec{r}}{r}\right)$$

$$\vec{a}^{(0)}_{LM} = g_L(\omega r)\,\vec{Y}^{(0)}_{LM}\left(\frac{\vec{r}}{r}\right) \tag{3}$$

$$g_L(\omega r) = (2\pi)^{3/2}\, i^L\, \frac{j_{L+\frac{1}{2}}(\omega r)}{\sqrt{\omega r}}$$

$j_{L+\frac{1}{2}}(\omega r)$ is the Bessel function, $\vec{Y}^{(\lambda)}_{L,M}$, $\vec{Y}_{J,L,M}$ are the vector spherical harmonic functions, as shown in reference (2).

With the expression (2), the transition rate of the hydrogen atom from the initial state $| n_1 l_1 >$ to the final state $< n_2 l_2 |$ through the absorption of one electric dipole photon and one quadrupole photon may be derived by a straightforward application of Feynman diagram for perturbation theory of QED [2]. The result is

$$W^{n_2\ell_2}_{n_1\ell_1} = 2\pi\delta(E_2 - E_1 - \omega - \omega')\left(\vec{e}\cdot\vec{Y}^{(1)*}_{10}\left(\frac{\vec{k}}{\omega}\right)\right)^2\left(\vec{e}'\cdot\vec{Y}^{(1)*}_{20}\left(\frac{\vec{k}'}{\omega'}\right)\right)^2 N\cdot N'\, M^{n_2\ell_2}_{n_1\ell_1}(\omega) \qquad (4)$$

$$M^{n_2\ell_2}_{n_1\ell_1}(\omega) = \frac{e^4}{4(2\ell_1+1)\omega\omega'}\sum_{m_1 m_2}\left|\sum_{n\ell m}\frac{\langle n_2\ell_2 m_2|\vec{\alpha}\cdot\vec{a}^{(1)}_{20}(\frac{\vec{r}}{r})|n\ell m\rangle\langle n\ell m|\vec{\alpha}\cdot\vec{a}^{(1)}_{10}(\frac{\vec{r}}{r})|n_1\ell_1 m_1\rangle}{E_n - E_1 - \omega} + \frac{\langle n_2\ell_2 m_2|\vec{\alpha}\cdot\vec{a}^{(1)}_{10}(-\frac{\vec{r}}{r})|n\ell m\rangle\langle n\ell m|\vec{\alpha}\cdot\vec{a}^{(1)}_{20}(-\frac{\vec{r}}{r})|n_1\ell_1 m_1\rangle}{E_n - E_1 - \omega'}\right|^2 \qquad (5)$$

where N, N' are the number of photons cross unit area per unit time in both laser beams which may be in connection to the Poynting vector of laser wave in classical approximation.

With the approximation $\omega r \ll 1$, one obtains

$$\langle n_2\ell_2 m_2|\vec{\alpha}\cdot\vec{a}^{(1)}_{LM}\left(\frac{\vec{r}}{r}\right)|n\ell m\rangle = \sqrt{\frac{L+1}{L}}\,\frac{4\pi(i\omega)^L}{(2L+1)!!}\langle n_2\ell_2|r^L|n\ell\rangle\langle\ell_2 m_2|Y_{LM}\left(\frac{\vec{r}}{r}\right)|\ell m\rangle \qquad (6)$$

$M^{n_2\ell_2}_{n_1\ell_1}(\omega)$, as shown in (5), may be reexpressed as

$$M^{n_2\ell_2}_{n_1\ell_1}(\omega) = \frac{(4\pi e)^4\omega\omega'^3}{2700(2\ell_1+1)}\sum_{m_1 m_2}\left|\sum_{\ell m}P^{n_2\ell_2}_{n_1\ell_1\ell}(\omega,1,2)\,C^{\ell m}_{\ell_1 m_1,10}\,C^{\ell_2 m_2}_{\ell m 20} + P^{n_2\ell_2}_{n_1\ell_1\ell}(\omega,2,1)\,C^{\ell m}_{\ell_1 m_1,20}\,C^{\ell_2 m_2}_{\ell m 10}\right|^2 \qquad (7)$$

where $P^{n_2\ell_2}_{n_1\ell_1\ell}(\omega, L_1, L_2)$ is the sum of the radial part of integral for the intermediate states.

$$P^{n_2\ell_2}_{n_1\ell_1\ell}(\omega, L_1, L_2) = \sum_n\frac{\langle n_2\ell_2|r^{L_2}|n\ell\rangle\langle n\ell|r^{L_1}|n_1\ell_1\rangle}{E_n - E_1 - \omega}$$
$$C^{\ell m}_{\ell_1 m_1, LM} = \langle \ell m|Y_{LM}\left(\frac{\vec{r}}{r}\right)|\ell_1 m_1\rangle \qquad (8)$$

3. <u>Selection rules</u>

The electric dipole and quadrupole selection rules limit that

$$\ell_2 = \ell_1 + 1;\quad \ell_2 = \ell_1 + 3;$$
$$\ell_2 = \ell_1 - 1, \text{ for } \ell_1 > 0; \qquad (9)$$
$$\ell_2 = \ell_1 - 3, \text{ for } \ell_1 > 2.$$

$M^{n_2\ell_2}_{n_1\ell_1}(\omega)$ in Eq.(5) for different cases are given as follows

$$M^{n_2\ell_1+1}_{n_1\ell_1}(\omega) = \frac{(4\pi e)^4\omega\omega'^3}{2700(2\ell_1+1)}\sum_{m_1 m_2}\left|\sum_m P^{n_2\ell_1+1}_{n_1\ell_1\ell_1+1}(\omega,1,2)\,C^{\ell_1+1\, m_2}_{\ell_1+1\, m\, 20}\,C^{\ell_1+1\, m}_{\ell_1 m_1,10} + P^{n_2\ell_1+1}_{n_1\ell_1\ell_1}(\omega',2,1)\,C^{\ell_1 m}_{\ell_1 m_1,20}\,C^{\ell_1+1\, m_2}_{\ell_1 m\, 10} + P^{n_2\ell_1+1}_{n_1\ell_1\ell_1-1}(\omega,1,2)\,C^{\ell_1-1\, m}_{\ell_1 m_1,10}\,C^{\ell_1+1\, m_2}_{\ell_1-1\, m\, 20} + P^{n_2\ell_1+1}_{n_1\ell_1\ell_1+2}(\omega',2,1)\,C^{\ell_1+2\, m}_{\ell_1 m_1,20}\,C^{\ell_1+1\, m_2}_{\ell_1+2\, m\, 10}\right|^2 \qquad (10)$$

here $\ell_2 = \ell_1+1$. For $\ell_1 > 0$, we have $\ell_2 = \ell_1 - 1$

$$M^{n_2\ell_1-1}_{n_1\ell_1}(\omega) = \frac{(4\pi e)^4\omega\omega'^3}{2700(2\ell_1+1)}\sum_{m_1 m_2}\left|\sum_m P^{n_2\ell_1-1}_{n_1\ell_1\ell_1+1}(\omega,1,2)\,C^{\ell_1+1\, m}_{\ell_1 m_1,10}\,C^{\ell_1-1\, m_2}_{\ell_1+1\, m\, 20} + P^{n_2\ell_1-1}_{n_1\ell_1\ell_1}(\omega',2,1)\,C^{\ell_1 m}_{\ell_1 m_1,20}\,C^{\ell_1-1\, m_2}_{\ell_1 m\, 10} + P^{n_2\ell_1-1}_{n_1\ell_1\ell_1-1}(\omega,1,2)\,C^{\ell_1-1\, m}_{\ell_1 m_1,10}\,C^{\ell_1-1\, m_2}_{\ell_1-1\, m\, 20} + P^{n_2\ell_1-1}_{n_1\ell_1\ell_1-2}(\omega',2,1)\,C^{\ell_1-2\, m}_{\ell_1 m_1,20}\,C^{\ell_1-1\, m_2}_{\ell_1-2\, m\, 10}\right|^2 \qquad (11)$$

For $\ell_2 = \ell_1+3$, Eq.(5) becomes

$$M^{n_2\ell_1+3}_{n_1\ell_1}(\omega) = \frac{(4\pi e)^4\omega\omega'^3}{2700(2\ell_1+1)}\sum_{m_1 m_2}\left|\sum_m P^{n_2\ell_1+3}_{n_1\ell_1\ell_1+1}(\omega,1,2)\,C^{\ell_1+1\, m}_{\ell_1 m_1,10}\,C^{\ell_1+3\, m_2}_{\ell_1+1\, m\, 20} + P^{n_2\ell_1+3}_{n_1\ell_1\ell_1+2}(\omega',2,1)\,C^{\ell_1+2\, m}_{\ell_1 m_1,20}\,C^{\ell_1+3\, m_2}_{\ell_1+2\, m\, 10}\right|^2 \qquad (12)$$

For $\ell_1 \geq 3$, $\ell_2 = \ell_1 - 3$,

$$M^{n_2\ell_1-3}_{n_1\ell_1}(\omega) = \frac{(4\pi e)^4\omega\omega'^3}{2700(2\ell_1+1)}\sum_{m_1 m_2}\left|\sum_m P^{n_2\ell_1-3}_{n_1\ell_1\ell_1-1}(\omega,1,2)\,C^{\ell_1-1\, m}_{\ell_1 m_1,10}\,C^{\ell_1-3\, m_2}_{\ell_1-1\, m\, 20} + P^{n_2\ell_1-3}_{n_1\ell_1\ell_1-2}(\omega',2,1)\,C^{\ell_1-2\, m}_{\ell_1 m_1,20}\,C^{\ell_1-3\, m_2}_{\ell_1-2\, m\, 10}\right|^2 \qquad (13)$$

4. Results for ground state of hydrogen atoms

Now we give the $M^{n_2 l_2}_{n_1 l_1}(\omega)$ of the ground state for atomic hydrogen, i.e. $n_1 = 1$, $l_1 = 0$.

$$M^{n_2 1}_{1\,0}(\omega) = \frac{(4\pi e)^4 \omega\omega'^3}{2700} \sum_{m,m_2} \left| \sum_m P^{n_2\,1}_{1\,0\,1}(\omega,1,2)\, C^{1\,m}_{0\,0\,1\,0}\, C^{1\,m_2}_{1\,m\,2\,0} \right|^2 \tag{14}$$

For $n_2 > 1$, $C^{1\,m}_{0\,0\,1\,0}\, C^{1\,m}_{1\,m\,20} = -\frac{\sqrt{5}}{10\pi}\,\delta_{m0}\cdot\delta_{m_2 0}$.

$$M^{n_2 3}_{1\,0}(\omega) = \frac{(4\pi e)^4 \omega\omega'^3}{2700} \sum_{m,m_2} \left| \sum_m P^{n_2 3}_{1\,0\,1}(\omega,1,2)\, C^{1\,m}_{0\,0\,10}\, C^{3\,m_2}_{1\,m\,20} + P^{n_2 3}_{1\,0\,2}(\omega',2,1)\, C^{3\,m_2}_{2\,m\,10}\, C^{2\,m}_{0\,0\,20} \right|^2 \tag{15}$$

where $C^{2\,m}_{0\,0\,2\,0}\, C^{3\,m_2}_{2\,m\,1\,0} = C^{1\,m}_{0\,0\,1\,0}\, C^{3\,m_2}_{1\,m\,2\,0} = \frac{3}{4\pi}\sqrt{\frac{3}{35}}\,\delta_{m0}\,\delta_{m_2 0}$.

With the nonrelativistic approximation, $P^{n_2 l_2}_{1\,0\,l}(\omega, L_1, L_2)$ can be accurately evaluated by using the Coulomb Green's function method.

$$g_l(\nu, r, r') = \sum_n \frac{R_{nl}(r)\, R^*_{nl}(r')}{E_n + \frac{e^2}{2\nu^2 a}}$$

$$= -\frac{2m}{\sqrt{rr'}} \int_0^\infty dx \quad \exp\left(-\frac{r+r'}{a\nu}\cosh x\right)\left(\coth\frac{x}{2}\right)^{2\nu} I_{2l+1}\left(\frac{2\sqrt{rr'}}{a\nu}\sinh x\right) \tag{16}$$

where $\nu = \frac{1}{(1 - 2a\omega/e^2)^{1/2}}$, a is the Bohr radius.

A useful integral representation involving $g_l(\nu, r, r')$ has been introduced and derived by Rapoport and Zon (3)

$$J_l(\beta,\beta') = -\frac{1}{2m_0}\int_0^\infty dr\,dr'\,(rr')^{l+1} \exp\left(-\frac{\beta r+\beta' r'}{a\nu}\right) g_l(\nu, r, r')$$

$$= \frac{2^{l+1}(2l+1)!\,(a\nu)^{2l+3}}{(l+1-\nu)\,[(1+\beta)(1+\beta')]^{2l+1}}\; {}_2F_1\left(2l+2,\, l+1-\nu,\, l+2-\nu,\, \frac{(1-\beta)(1-\beta')}{(1+\beta)(1+\beta')}\right) \tag{17}$$

where ${}_2F_1$ is the usual hypergeometric function. Substituting wave functions $R_{n_1 l_1}(r)$ and $R^*_{n_2 l_2}(r)$ we obtain the radial second order matrix element in terms of $J_l(\beta,\beta')$

$$P^{n_2 l_2}_{n_1 l_1 l}(\omega, L_1, L_2) = \left[D^{n_2 l_2}_{n_1 l_1 l L_1 L_2}(\beta,\beta')\, J_l(\beta,\beta')\right]_{\beta=\frac{\nu}{n_2},\ \beta'=\frac{\nu}{n_1}} \tag{18}$$

A few explicit expressions of $D^{n_2 l_2}_{1\,0\,l\,L_1 L_2}(\beta,\beta')$ are given as examples

$$D^{2\,1}_{1\,0\,1\,1\,2}(\beta,\beta') = \frac{2m}{\sqrt{6}}\,\nu^4 \frac{\partial^4}{\partial\beta^3\partial\beta'}$$

$$D^{4\,3}_{1\,0\,1\,1\,2}(\beta,\beta') = \frac{2m\nu^6}{384\sqrt{35}}\,\frac{\partial^6}{\partial\beta^5\partial\beta'} \tag{19}$$

$$D^{4\,3}_{1\,0\,2\,2\,1}(\beta,\beta') = \frac{2m\nu^4}{384\sqrt{35}\,a^2}\,\frac{\partial^4}{\partial\beta^3\partial\beta'}$$

where the units, in which $\hbar = c = 1$, are used.

In order to simplify the calculation we put

$$M^{n_2 l_2}_{n_1 l_1}(\omega) = \frac{(4\pi e)^4 a^{10} m^2 \omega\omega'^3}{2700(2l_1+1)}\,\mathcal{M}^{n_2 l_2}_{n_1 l_1}(\omega) \tag{20}$$

The quantity $\mathcal{M}^{n_2 l_2}_{n_1 l_1}(\omega)$ has zero dimension. Using the expressions (16)---(20), the numerical calculation of $\mathcal{M}^{21}_{10}(\omega)$, $\mathcal{M}^{43}_{10}(\omega)$ has been done. The results are shown in Fig.1, Fig.2. It may be compared to the future experiments with high laser power.

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FIGURE CAPTIONS

Fig.1 1s → 2p Two photon relative absorption probability and dipole photon frequency.

Fig.2 1s → 4d Two photon relative absorption probability and dipole photon frequency.

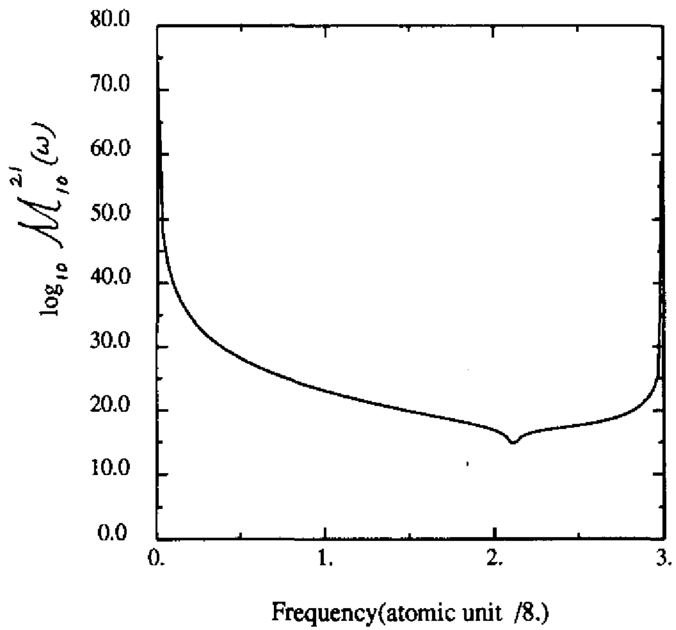
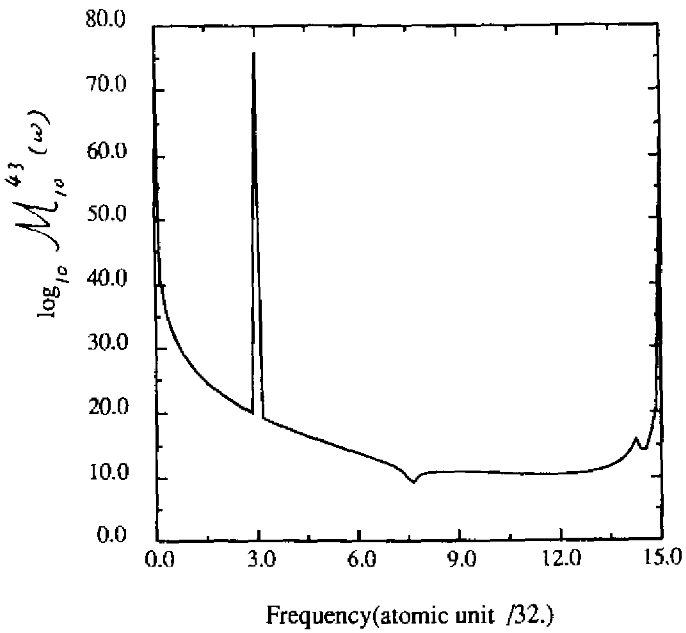


Fig.1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70.0
60.0
50.0
40.0
30.0
20.0
10.0
0.0
0.0
3.0
6.0
9.0
12.0
15.0
Frequency(atomic unit /32.)

Fig.2