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Interaction of strong laser beam with He-atom using non-perturbative Floquet method

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Abstract. Non-perturbative Floquet method is used to investigate the interaction of strong linearly polarized light with He-atom. In the calculations, six lowest singlet target states and fifteen photon states are taken for absolute convergence. For allowed transitions, accurate oscillator strengths using configuration interaction wave-functions are used. A number of interesting features such as nonlinear effects at high intensities, line broadening, a dynamic Stark effect are obtained and explained.

Keywords. Photon states; oscillator strengths; target states.

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1. Introduction

Recently there has been considerable attention in the study of interaction of intense laser beam with multielectron atoms (Lambropoulos and Tang 1987; Perry and Szoke 1988) due to its importance in the production of powerful lasers and laser fusion reactions. Here in this paper we have used the Floquet theory (non-perturbative) for the calculation of multi-photon excitation of the He-atom. The Floquet theory (Shirley 1965; Moloney and Faisal 1979; Leasure and Wyatt 1979; Chu 1986) relates the solution of Schrödinger equation with a periodic Hamiltonian (laser + atom system) to the solution of another Schrödinger equation with a time-independent Hamiltonian represented by an infinite matrix (called a Floquet matrix). In the present calculations we have included lowest six singlet states of target atom and fifteen photon states (i.e. $N = 0, \pm 1, \pm 2, \pm 3, \pm 4, \pm 5, \pm 6, \pm 7$) for absolute convergence.

2. Floquet theory formalism

The Schrödinger equation for a quantum mechanical system in an external field, which is periodic function of time with period T, can be written (in a.u.) as

$$i(\partial\psi_p/\partial t) = H\Psi_p(\mathbf{r}, t) \tag{1}$$

where $H = H_0 - \mu(\mathbf{r}).\ \mathbf{E}_0 \cos\omega t$; H_0 is the Hamiltonian for the isolated atom, $\mu(r)$ is the atomic dipole moment function, $|\mathbf{E}_0|$ is the amplitude and ω is the frequency of the field.

We can expand the total wavefunction $\Psi_p(\mathbf{r}, t)$ of the system in terms of field free atomic eigenfunctions

$$\Psi_p(\mathbf{r}, t) = \sum_{q=1}^{N} a_{qp}(t)\chi_q(\mathbf{r}) \tag{2}$$

when the e.m. field is turned off adiabatically, the wavefunction of the system becomes the usual stationary states, hence,

$$\Psi_p(\mathbf{r}, t) = \chi_q(\mathbf{r}). \tag{3}$$

Substituting (2) into (1) and using the orthogonality conditions for the atomic states, we obtain

$$i\frac{\partial a_{qp}}{\partial t} = E_q^0 a_{qp}(t) - |\mathbf{E}_0|\cos\omega t \sum_{s=1}^{N} V_{qs}a_{sp}(t) \tag{4}$$

where

$$V_{qs} = \langle \chi_q(\mathbf{r})|\mu(\mathbf{r})\cdot\hat{E}_0|\chi_s(\mathbf{r})\rangle \tag{4a}$$

is the dipole matrix element and $\hat{E}_0$ is a unit vector in the direction of $\mathbf{E}_0$. In the matrix notation the above equation can be written as

$$i(\partial A/\partial t) = H_c(t)A(t), \tag{5}$$

where $H_c(t)$ is a periodic function of time with period $T = 2\pi/w$ i.e. $H_c(t) = H_c(t+T)$. According to Floquet theory (5) admits the following solution

$$A^F(t) = \Phi(t)\exp(-i\mu t), \tag{6}$$

where $\Phi(t)$ is another periodic function of time, and μ is the diagonal matrix called the characteristic exponent.

According to Shirley (1965), the Fourier series expansions of $A^F(t)$ and $H_c(t)$ are given by

$$A^F(t) = \sum_{m=-\infty}^{+\infty} f_{r,l}^m \,\mathrm{emp}(im\omega t)\exp(-i\mu_l t) \tag{7}$$

and

$$H_c(t) = (H_c)_{r,l} = \sum_{m=-\infty}^{+\infty} \mathscr{H}_{r,l}^m \exp(im\omega t) \tag{8}$$

where $f_{r,l}^m$ and $\mathscr{H}_{r,l}^m$ are the Fourier amplitudes corresponding to a particular value of m. Here the indices r and l range over the atomic states and the Fourier index m indicates the photon number.

Substituting (7) and (8) in (5) yield the following eigenvalue equation for the Fourier components $f_{r,l}^m$ and the characteristic value μ_l i.e.

$$\sum_j \sum_n (\mathscr{H}_{r,j}^{m-n} + n\omega\delta_{r,j}\delta_{nm})f_{rl}^n = \mu f_{r,l}^m. \tag{9}$$

Once the eigenvalue $f_{r,l}^m$ corresponding to the eigenvalue μ_l are found by diagonalizing the Floquet matrix H_F (the terms within the bracket of (9)), the transition amplitude can be determined from (7). More explicitly the transition probability from the initial atomic state $|i\rangle$ to final atomic state $|j\rangle$ is given by (Shirley 1965; Leasure and Wyatt 1979; Moloney and Faisal 1979)

$$P_{i\to j}(t,t_0)=|\langle j|U(t,t_0)|i\rangle|^2 \tag{10}$$

where $U(t,t_0)$ is the evolution operator.

For continuous coherent operation of the laser, we average (10) over $t-t_0=T$, and obtain the long time average transition probability as

$$\bar{P}_{i\to j}=\lim_{T\to\infty}\frac{1}{T}\int_0^T P_{ij}(\tau)\,d\tau$$
$$=\sum_n\sum_{\mu_l}|f^n_{i,\mu_l}f^0_{j,l}|^2. \tag{11}$$

The determination of $\bar{P}_{i\to j}$ requires the construction of the Floquet Hamiltonian H_F which involves the dipole-matrix element between the various target levels. We have used the recent calculations (Berrington *et al* 1985; Berrington and Kingston 1987) for the evaluation of accurate dipole matrix elements from oscillator strengths for the He-atom. The sophisticated configuration interaction wavefunctions, needed to evaluate oscillator strengths have been obtained by using CI V3 code (Hibbert 1975). The theoretical energies obtained for six lowest singlet levels and the oscillator strengths for different allowed transitions between them are shown in tables 1 and 2 respectively. In our calculations we have truncated the Floquet Hamiltonian H_F to

Table 1. Excitation energies (in eV) for $n=1$, $n=2$ and $n=3$ states in He atom relative to groundstate 1^1S.

State	Thresholds (eV)
1^1S	0·0
2^1S	20·617
2^1P^0	21·219
3^1S	22·921
3^1D	23·075
3^1P^0	23·088

Table 2. Oscillator strength for allowed transitions among the lowest six states in the He atom.

Transitions	f(length)
$1^1S\to 2^1P^0$	0·2145
$1^1S\to 3^1P^0$	0·0006
$2^1S\to 2^1P^0$	0·3662
$2^1S\to 3^1P^0$	0·1755
$2^1P^0\to 3^1S$	0·0514
$2^1P^0\to 3^1D$	0·7254
$3^1S\to 3^1P^0$	0·5663
$3^1P^0\to 3^1D$	0·0221

contain fifteen Floquet photon blocks ($N=0$, ± 1, ± 2, ± 3, ± 4, ± 5, ± 6, ± 7) as described by Mohan and Sharma (1986) although the results were found to be converged after truncating H_F by including only seven photon blocks.

Using the standard diagonalization routines from SSP Library, the Floquet Hamiltonian H_F of dimension (90×90) is diagonalized to yield the characteristic Floquet exponents and the corresponding Fourier components defined by (9). These are then substituted into (11) to yield infinite-time average transition probability.

3. Results and discussion

The results for the $(s^2)1^1S \rightarrow (1s2p)2^1P^\circ$ transition are shown in figure 1 with the variation of laser frequency (in cm^{-1}) for several laser intensities. Clearly the probability $\bar{P}_{1\rightarrow 3}$ at $I = 10^6$ cm^{-2}, increases quite steadily from $\omega = 161470{\cdot}0\,\mathrm{cm}^{-1}$ to, $\omega = 161491{\cdot}8\,\mathrm{cm}^{-2}$ where it has an extremum value. However, with further increase in the frequency, the probability $\bar{P}_{1\rightarrow 3}$ decreases. At a higher intensity $I = 10^7$ Wcm^{-2}, the probability $\bar{P}_{1\rightarrow 3}$ has the similar variation except the extremum occurs at $\omega = 161491{\cdot}6\,\mathrm{cm}^{-1}$. Thus, the shifting of resonance by $\Delta\omega = 0{\cdot}2\,\mathrm{cm}^{-1}$ with increase of laser power 10^1 Wcm^{-2}, is a clear indication of the dynamic Stark shift which increases with increasing intensity of the laser beam. Further broadening of overall probability curve at $I = 10^8$ Wcm^{-2} as compared to that at $I = 10^6\,\mathrm{Wcm}^{-2}$ shows that the line broadening effect is much more pronounced in He atom at high intensities as also shown by others (Geltman 1980; Mohan and Sharma 1986).

In figure 2 we have shown the variation of $(1s2p)2^1p^0 \rightarrow (1s3d)$ 3^1D transition probability with frequency of the laser beam. Comparison of the two figures shows

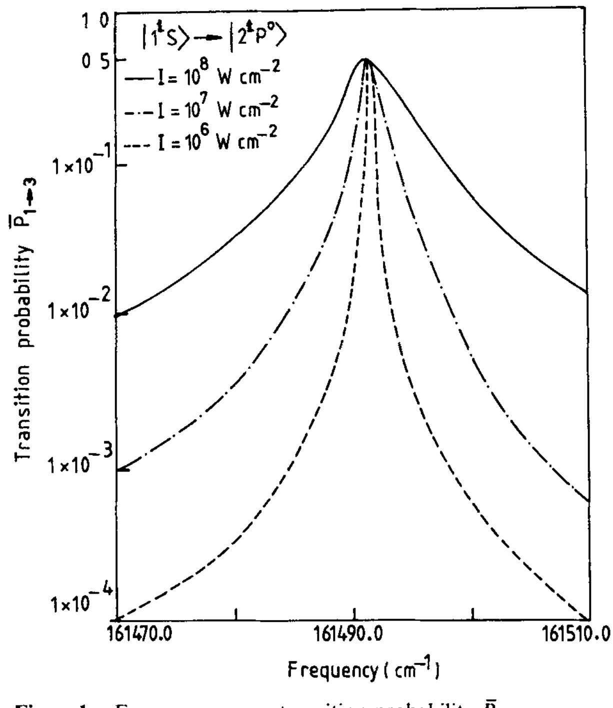

Figure 1. Frequency versus transition probability $\bar{P}_{1-3}$.

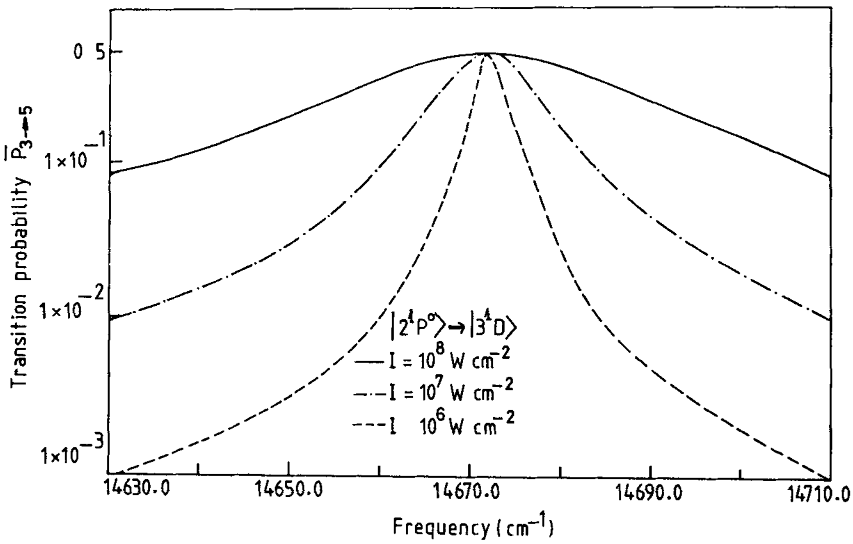

Figure 2. Frequency versus transition probability $\bar{P}_{3-5}$.

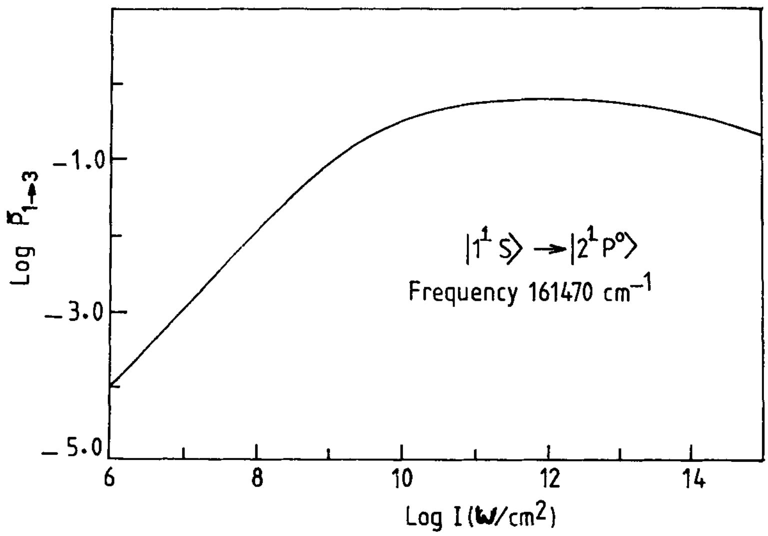

Figure 3. Log intensity versus log transition probability $\bar{P}_{1-3}$.

the asymmetrical behaviour for transition $1^1S \rightarrow 2^1P^0$ at laser intensities 10^7 W/cm^2 and 10^8 W/cm^2. This is due to low coupling between 1^1S and 2^1P^0 with respect to the coupling between 2^1P^0 and 3^1D. This asymmetrical behaviour becomes more pronounced with increase of laser intensity.

Figure 3 shows the variation of transition probability $\mathbf{P}_{1-3}$ for the $1^1S \rightarrow 2^1P^0$ transition with intensity I (W/cm^2) at $\omega = 161470\,\text{cm}^{-1}$. Here the probability varies linearly with intensity up to $10^{10}\,\text{Wcm}^{-2}$ as expected from perturbation theory. However, as the intensity of the field is increased further, the probability reaches to the saturation value up to $10^{13}\,\text{Wcm}^{-2}$. Beyond $I \geqslant 10^{14}\,\text{Wcm}^{-2}$ there is a decline in probability showing the nonlinear effect at high intensity.

Table 3. Variation of transition probability $\bar{P}_{i\to j}$ with intensity I, for frequency $\omega = 161470\,\mathrm{cm}^{-1}$.†

Intensity I(W/cm^2)	$\bar{P}_{i\to 1}$	$\bar{P}_{i\to 2}$	$\bar{P}_{i\to 3}$	$\bar{P}_{i\to 4}$	$\bar{P}_{i\to 5}$	$\bar{P}_{i\to 6}$
10^6	0·9999023	0·5501(−14)	0·976055(−4)	0·210658(−13)	0·2721(−14)	0·53851997(−12)
10^{10}	0·667904	0·18716854(−6)	0·330954(0)	0·716908(−8)	0·92546(−7)	0·47642(−8)
10^{14}	0·6069	0·235957(−2)	0·379416(0)	0·515699(−3)	0·1633(−2)	0·9054(−2)

†The number in parentheses indicates the power of 10 by which the entry should be multiplied.

In our calculations t low intensities ($10^6\,\mathrm{Wcm}^{-2} \leqslant I \leqslant 10^{13}\,\mathrm{Wcm}^{-2}$) for frequency near the resonance of $1^1S \to 2P^0$ transition, we find that 1^1S and 2^1P^0 levels are strongly coupled as compared to coupling between 1^1S and other higher levels e.g. 2^1S, 3^1S, 3^1P and 3^1D, which results a negligible probability flow to these channels while for high intensities i.e. for $I \geqslant 10^{14}\,\mathrm{Wcm}^{-2}$, there is a substantial probability flow to these channels (also shown in table 3) which results the mixing of all the levels. This mixing leads finally to drop in the probability for $1^1S \to 2^1P^0$ transition.

4. Conclusions

We have used the Floquet theory which is non-perturbative method and takes into account of all multiphoton processes and therefore our results are valid for high intensities. Further work for understanding the behaviour of complex-multi-electron atoms and ions using the present method is in progress.

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