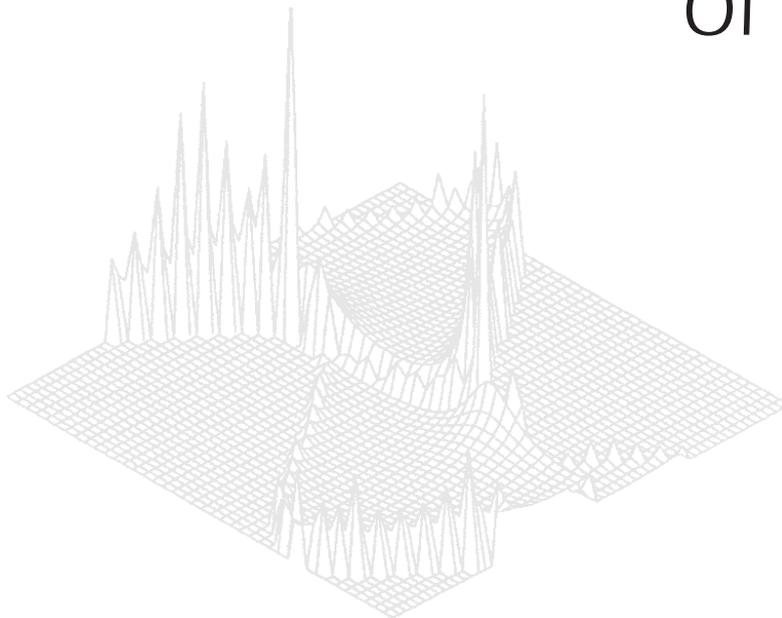

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Excitation Dynamics of an Atom due to Heavy Ion Impact in a Laser Field

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Abstract

Laser assisted inelastic scattering of heavy ions by alkali atoms is studied theoretically. The non-perturbative quasi-energy method, generalised for many states, is used to describe the laser–atom interaction, and the close coupling method using the impact parameter method is used for scattering calculations. We have calculated the transition probabilities and total cross section for the excitation of alkali atoms, due to simultaneous proton–photon collisions. We show the effect of laser and collision parameters, e.g. laser intensity, impact parameter, laser frequency, on the excitation process.

1. Introduction

Calculations of the collisionally aided radiative excitation of alkali atoms due to heavy ion impact in the presence of a laser field are presented here. Such a study is quite useful in the analysis of ion–atom collisions, for a quantitative modelling of low and high temperature plasmas (Mittleman 1982; Rahman and Guidotti 1992; Luans *et al.* 1991; Mohan and Prasad 1991; Massey *et al.* 1984). These processes are of interest because by using them we obtain information on the interaction between colliding particles, on the trend of gas phase chemical reactions in specific ways (Gudzenko *et al.* 1977) and on ways to develop new type of lasers (George 1986). The study of collision processes in the presence of a high intensity laser field has attracted considerable attention during the last few years. This is because of the availability of intense laser radiation sources and their application to populating resonantly excited states of atoms. Experimental evidence of simultaneous electron–photon excitation of atoms has been reported (Mason and Newell 1982).

The excitation of the alkali-metal atoms from their ns ground states has been a popular subject for experimentalists and theorists because these states are almost hydrogenic in nature. But the experimental study of alkali-metal atom valence electron excitation of heavier projectiles has been scarce. In the case of sodium, we find impacts limited to H^+ , H_2^+ , H_3^+ , H^- and H^0 and energies below 25 keV (Howard *et al.* 1982, 1983; Jitschin *et al.* 1986). Theoretical treatment of the $p+Na(3s)\rightarrow p+Na(3p)$ process above 25 keV has been limited to two Born approximation (Kubach and Sidis 1981) and one close coupling (Bell and Skinner 1962) calculations, and the Vainshtein–Presnyakov–Sobelman

approximation (Theodosiou 1987). In this paper, we present the effect of various collision and laser parameters on the interaction between the laser field and the system of ion and atom undergoing collisions.

We used the non-perturbative method developed by Agre and Rapport (1982) to study the laser–sodium atom interactions. We have already used this method quite successfully to study various collisional problems (Sharma and Mohan 1986, 1992; Sharma *et al.* 1993). The problem studied here is collision of an incoming proton with a sodium atom initially in the state i in the presence of a single mode laser beam moving to excited state j , with exchange of L photons between the atom and the laser field. We have used the impact parameter method to treat the proton–atom interaction. This method, which was developed originally by Seaton (1962) and by Stauffer and McDowell (1965, 1966) to describe electronic excitation in electron–atom collisions, is especially suited for spin-allowed electric dipole transitions. This feature makes this method complementary to the close coupling and L^2 distorted-wave methods which appear more suitable for spin forbidden transitions dominated by electron exchange effects.

2. Theory

We consider a collision between a proton and sodium atom in the ground state, in the presence of a laser field. The field is assumed to be purely monochromatic with angular frequency ω , linearly polarised with linear polarisation vector \hat{e} . We also assume that the dipole approximation is valid.

The time-dependent Schrödinger equation for the above proton–atom system in the presence of laser beam is

$$i \frac{\partial}{\partial t} \psi(r, t) = [H_0(r) + V_{\text{int}}^1(E, \omega, t) + V] \psi(r, t). \quad (1)$$

Here H_0 is the isolated Hamiltonian of the atom, $V_{\text{int}}^1(E, \omega, t)$ is the interaction of the laser field with the atom and V is the interaction of the incident proton with the atom. We can expand the total wave function of the system $\psi(r, t)$ in terms of the dressed states of the atom in the presence of a laser field, i.e.

$$\psi(r, t) = \sum_n C_n \phi_n(r, t), \quad (2)$$

where the subscript n runs over all states of the atom included. Substituting equation (2) in (1) and using the orthogonality conditions for the dressed states, we obtain the following set of coupled first order differential equations:

$$i \frac{\partial}{\partial t} C_k = \sum_n C_n(t) \langle \phi_k(r, t) | V | \phi_n(r, t) \rangle. \quad (3)$$

The interaction between the incident proton and atom is defined as

$$V = \frac{-Z}{R} + \sum_{j=1}^Z \frac{1}{r_{0j}}, \quad (4)$$

where R is the distance between the proton and atom, and r_j is the position vector of the j th electron with respect to the atomic nucleus and

$$\frac{1}{r_{0j}} = \frac{1}{|R - r_j|}. \quad (5)$$

The potential V becomes time-dependent due to the classical linear trajectory $R(t)$ defined by

$$|R(t)|^2 = (b^2 + v^2 t^2), \quad (6)$$

where b is the impact parameter, and v is the velocity of the proton. It is well known that the quantum levels of an atom get ‘dressed’ in the presence of a laser field and these dressed states are known as quasi-energy states (QES), which satisfy the equation

$$i \frac{\partial \phi}{\partial t} = H_a \phi, \quad (7)$$

where $H_a = H_0(r) + V_{\text{int}}^1(E, \omega, t)$. If we assume that the atom is interacting with radiation which is nearly resonant (i.e. $\epsilon_{n0} < \omega$, where ϵ_{n0} is the n photon detuning) and that the intensity of the radiation is not too high, then the solution of the Schrödinger equation (7) can be written as

$$\phi_n(r, t) = e^{-i(E_1 + \lambda_n t)} [a_1^n u_1(r) + a_2^n u_2(r) e^{-i\omega t} + a_3^n u_3(r) e^{-2i\omega t}], \quad (8)$$

where $u(r)$ are the unperturbed (or bare) atomic states, E_1 is the ground-state energy of the bare atom, the a_m^n are amplitudes corresponding to the bare atomic states, and λ_n are defined as quasi-energies. It is to be noted that we have written the solution (8) only for three states, but it can be generalised for more (multiplets) (Prasad *et al.* 1996a, 1996b).

We need to solve the set of coupled equations given by equation (3). On substituting equation (8) we get

$$\begin{aligned} i \frac{\partial}{\partial t} C_k &= \sum_n C_n e^{-i(E_1 + \lambda_n t)} (a_1^k u_1 + a_2^k u_2 e^{-i\omega t} + a_3^k u_3 e^{-2i\omega t}) \\ &\quad \times |V| e^{-i(E_1 + \lambda_n t)} (a_1^n u_1 + a_2^n u_2 e^{-i\omega t} + a_3^n u_3 e^{-2i\omega t}) \\ &= \sum_n C_n e^{-i(\lambda_n - \lambda_k)t} [a_1^n a_1^k u_{11} + a_1^n a_2^k u_{12} e^{-i\omega t} \\ &\quad + a_1^n a_3^k u_{13} e^{-2i\omega t} + a_2^n a_1^k u_{21} e^{i\omega t} + a_2^n a_2^k u_{22} \\ &\quad + a_2^n a_3^k u_{23} e^{-i\omega t} + a_3^n a_1^k u_{31} e^{2i\omega t} + a_3^n a_2^k u_{32} e^{-i\omega t} \\ &\quad + a_3^n a_3^k u_{33}]. \end{aligned}$$

Rearranging the terms we get

$$\begin{aligned}
i \frac{\partial}{\partial t} C_k = & \sum_n C_n e^{i(\lambda_n - \lambda_k)t} [a_1^n a_1^k u_{11} + a_2^n a_2^k u_{22} + a_3^n a_3^k u_{33}] \\
& + e^{i(\lambda_n - \lambda_k - \omega)t} [a_1^n a_2^k u_{12} + a_2^n a_3^k u_{23}] \\
& + e^{i(\lambda_n - \lambda_k + \omega)t} [a_2^n a_1^k u_{21} + a_3^n a_2^k u_{32}] \\
& + e^{i(\lambda_n - \lambda_k - 2\omega)t} [a_1^n a_3^k u_{13}] \\
& + e^{i(\lambda_n - \lambda_k + 2\omega)t} [a_3^n a_1^k u_{31}], \tag{9}
\end{aligned}$$

where the matrix elements are

$$u_{ij} = \langle \chi_i | V | \chi_j \rangle \tag{10}$$

and $\chi_i(r)$ represents the bare atomic states,

$$\chi_i(r) = R_{nl}(r) Y_{lm}(\theta, \phi), \tag{11}$$

and where $R_{nl}(r)$ describes the radial wave functions,

$$R_{nl}(r) = N_{nl} \exp(-r/\nu) (2r/\nu)^\nu \sum_{t=0}^{\infty} a_t r^{-t}, \tag{12}$$

and ν is the effective quantum number of the state n, l .

Equation (3) can be written in a matrix form as

$$i \dot{\mathbf{C}}(t) = \mathbf{Q}(t) \mathbf{C}(t), \tag{13}$$

where $\mathbf{C}(t)$ is a column matrix and $\mathbf{Q}(t)$ is a coupling matrix defined by

$$\mathbf{Q}(t) = \langle \phi_k(r, t) | V | \phi_n(r, t) \rangle. \tag{14}$$

Using the standard diagonalisation technique, the solution of equation (13) at $t = +\infty$ is given by (Sharma and Mohan 1986; Callaway and Baur 1965)

$$\mathbf{C}(+\infty) = \mathbf{U} \exp(-i\mathbf{M}_D) \mathbf{U}^+ \mathbf{C}(-\infty), \tag{15}$$

where \mathbf{U} is a unitary operator and \mathbf{M}_D is a diagonalised matrix obtained by the unitary transformation

$$\mathbf{M}_D = \mathbf{U}^+ \mathbf{M} \mathbf{U}, \tag{16}$$

where

$$\mathbf{M} = \int_{-\infty}^{+\infty} \mathbf{Q}(t') dt'. \quad (17)$$

Using equation (15), the transition probability for the transition from state i to f is given by

$$P_{i \rightarrow f} = |\mathbf{C}_{i \rightarrow f}(+\infty)|^2. \quad (18)$$

This probability can be integrated with respect to the impact parameter to find the total cross section for the transition from the initial state i to final state f , i.e.

$$\sigma_{i \rightarrow f} = 2 \int_0^{\infty} P_{i \rightarrow f}(b) b db. \quad (19)$$

The eigenvalues λ_n and the corresponding eigenvectors a_m^n were calculated by diagonalising the characteristic equation obtained by substituting equation (8) in (7) and by using the orthogonality conditions for the bare states. Substitution of these eigenvalues and eigenfunctions in equation (8) gives us the dressed states of the corresponding atoms. The dressed states are then substituted in equation (14) to give us the coupling matrix \mathbf{Q} . The elements of the coupling matrix \mathbf{Q} involve the dipole matrix and time-dependent terms which are integrated analytically to give us the matrix elements M_{ij} defined by equation (17). Next, we diagonalised \mathbf{M} to obtain \mathbf{M}_D and \mathbf{U} where \mathbf{M}_D is the diagonalised matrix and \mathbf{U} is the unitary matrix. Substitution of these in equations (16) and (18) gives us the required transition amplitudes and transition probability. Since the dressed states $\phi_n(r, t)$ are largely influenced by the radiation field, therefore, in addition to the collision parameters, the cross section for the above-mentioned process depends on various laser parameters, for example the frequency and intensity.

3. Results and Discussion

Here we have studied the laser assisted proton impact excitation of the sodium atom and the influence of laser and collision parameters on this process. The dressed states of the atom are calculated using the quasi-energy method described earlier. The quasi-energy matrix has been diagonalised using standard diagonalisation routines to yield the quasi-energies and corresponding eigenvectors. In constructing the quasi-energy matrix we need dipole matrix elements between adjacent atomic states. We have calculated these dipole matrix elements numerically. Though we show results for only three states, in our calculations we have included the lowest eight states of the sodium atom.

In Fig. 1 we show the variation of transition probabilities with laser frequency ω (in eV) in the presence of a laser field and without collisions. It can be seen that the transition probabilities vary with ω as expected. The transition probability for the $3s \rightarrow 3d$ transition rises sharply at $\omega = 1.808$ eV since the two photon resonance condition is satisfied here for the $3s \rightarrow 3d$ transition, as

shown in Fig. 1. Also, the transition probability for the $3s \rightarrow 3p$ transition rises sharply at $\omega = 2 \cdot 1018$ eV, which is again the one photon resonance frequency for the $3s \rightarrow 3p$ transition.

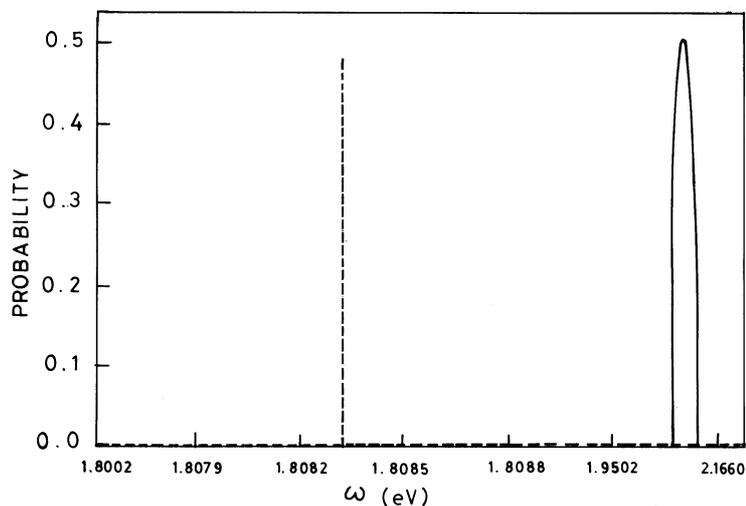


Fig. 1. Variation of the transition probability without collisions for the $3s-3p$ (solid line) and $3s-3d$ (dotted line) transitions with laser frequency (in eV) for intensity 10^6 W cm $^{-2}$.

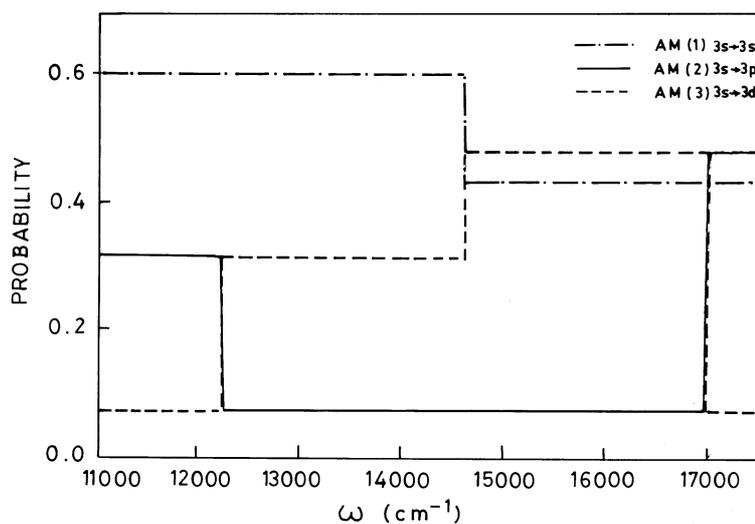


Fig. 2. Variation of the transition probability with collisions for $3s-3s$, $3s-3p$ and $3s-3d$ transitions with laser frequency (in eV) for intensity 10^6 W cm $^{-2}$ and collision velocity $v = 1$ a.u.

However, the behaviour of the transition probability is greatly influenced by the collision process, as shown in Fig. 2. We get a range of frequencies near resonances where the probability remains a maximum, unlike the process without

collisions where it is a maximum for one particular frequency. This type of process is only possible with collisions where all the levels participate in excitation depending upon the oscillator strengths between various levels. As we vary frequency ω from $10,000 \text{ cm}^{-1}$, we find there is a steady variation of $P^L(3s \rightarrow 3p)$ and $P^L(3s \rightarrow 3d)$ up to $\omega = 12218 \text{ cm}^{-1}$, and after this there is an abrupt fall of $P^L(3s \rightarrow 3p)$ and a rise of $P^L(3s \rightarrow 3d)$. This is because $\omega = 12218.3 \text{ cm}^{-1}$ (1.51483 eV) corresponds to the resonance frequency between the 3p and 3d states and these states are strongly coupled, since they have a large value of the oscillator strength ($f_{3p,3d} = 0.8433$) between them. With a further increase of frequency we again find a rise of $P^L(3s \rightarrow 3d)$ around $\omega = 14585 \text{ cm}^{-1}$, which is due to the two photon resonance condition being met at this particular frequency for the $3s \rightarrow 3d$ transition ($\omega_{\text{res}} = 29170.5 \text{ cm}^{-1}$ or 3.616 eV). Similarly, near

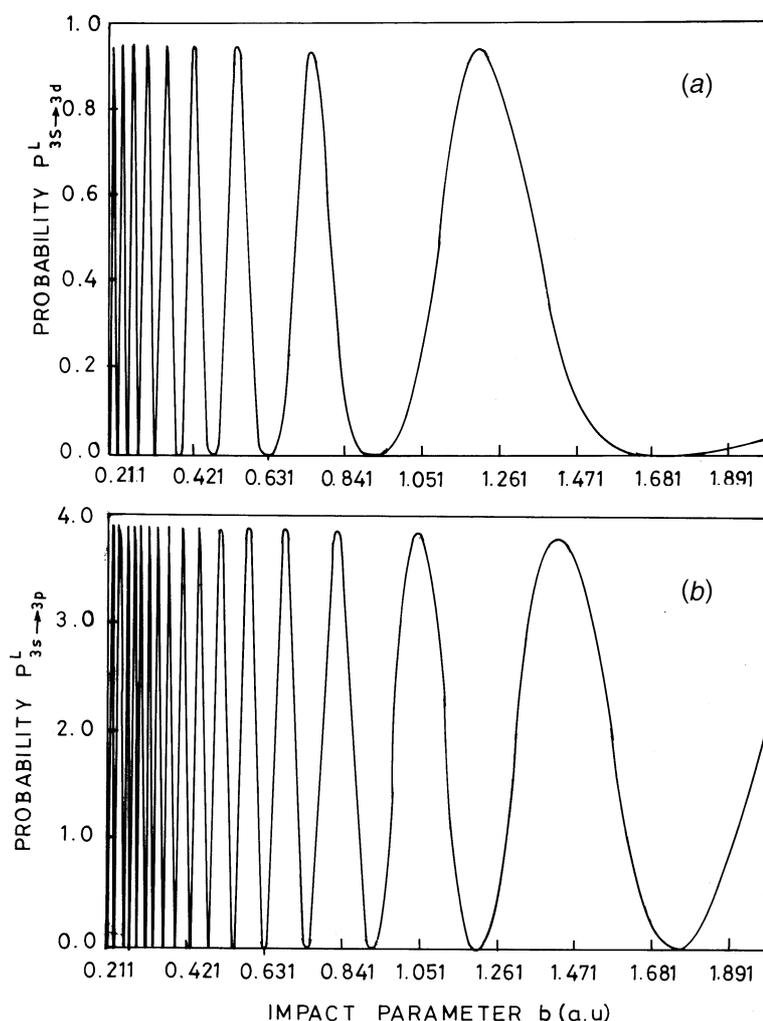


Fig. 3. Variation of collision probability with impact parameter b (in a.u.) for an intensity of 10^6 W cm^{-2} and $\omega = 2.1 \text{ eV}$ and $v = 1 \text{ a.u.}$ for (a) the $3s \rightarrow 3d$ transition and (b) the $3s \rightarrow 3p$ transition ($\times 10$).

$\omega = 16952 \text{ cm}^{-1}$ we find a rise of the $P^L(3s \rightarrow 3p)$ transition, which is due to the one photon resonance condition being met for the $3s \rightarrow 3p$ transition ($\omega_{\text{res}} = 16952 \cdot 2 \text{ cm}^{-1}$ or $2 \cdot 1017 \text{ eV}$). As the $3p \rightarrow 3d$ states are strongly coupled and are dipole allowed ($\Delta l = 1$) and the $3s$ and $3d$ states are a dipole forbidden transition ($\Delta l = 2$), there is a fall in the $P^L(3s \rightarrow 3d)$ probability. Thus the $P^L(3s \rightarrow 3d)$ probability remains quite steady between $\omega = 12220$ and 16952 cm^{-1} , instead of having a maximum at only $\omega = 14585 \text{ cm}^{-1}$ (Fig. 1). This remarkable behaviour is also reflected in the cross sections shown below in Fig. 4. Thus, it can be inferred that the collision process in a laser field can give a reaction which cannot be possible otherwise. The system gets enough energy to overcome some kind of (potential) barrier and chooses other channels, which are otherwise forbidden due to selection rules.

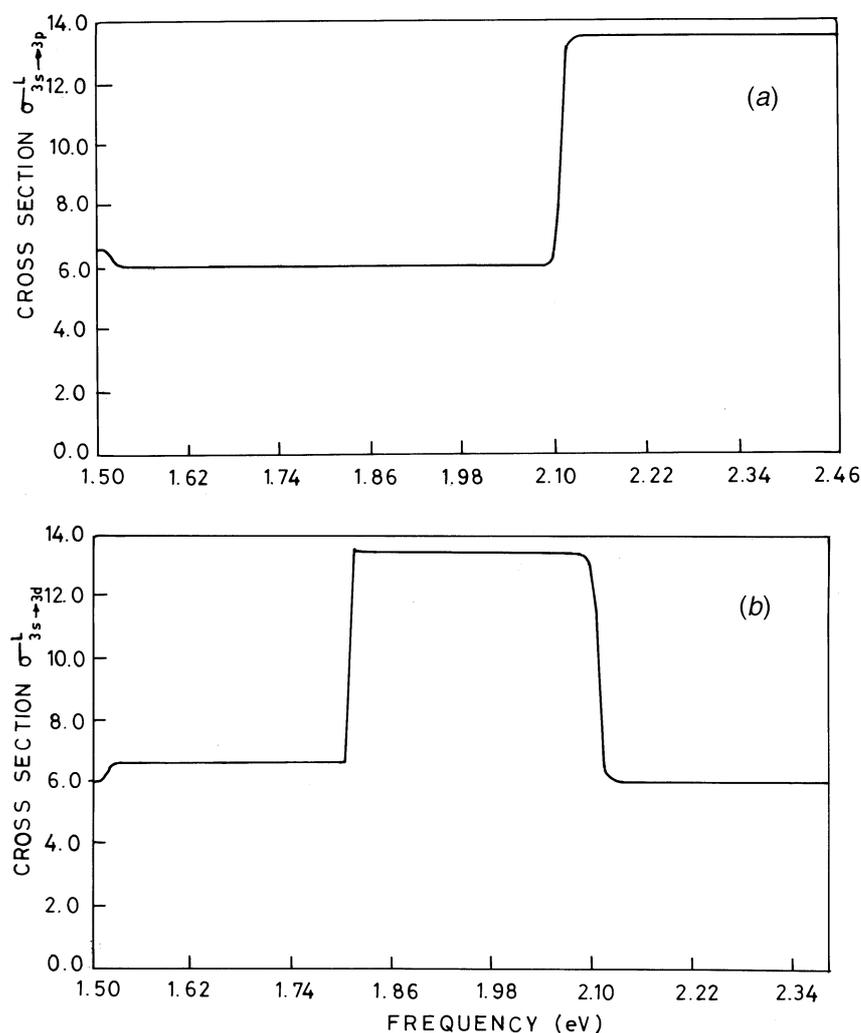


Fig. 4. Variation of the excitation cross section with laser frequency ω (in eV) for an intensity of 10^8 W cm^{-2} and $v = 1 \text{ a.u.}$ for (a) the $3s \rightarrow 3p$ transition and (b) the $3s \rightarrow 3d$ transition.

In Fig. 3 we show the variation of transition probability with impact parameter b (in a.u.) for (a) the $3s \rightarrow 3d$ and (b) the $3s \rightarrow 3p$ transition. Here we have taken the collision velocity to be $v = 1$ a.u. and the laser frequency $\omega = 2.1$ eV, while the laser intensity is $I = 10^6 \text{ W cm}^{-2}$. Clearly, it can be seen that the transition probability oscillates at small b , and decreases to zero at large b . This type of variation in transition probability with impact parameter is a general feature of the collision problem and can be explained on the basis of the multi-channel effect. At large impact parameter, the interaction is weak and the transition probability is small. Comparing the transition probabilities for two different transitions, we find that the transition probabilities $P^L(3s \rightarrow 3d)$ (Fig. 3a) has a large amplitude compared to $P^L(3s \rightarrow 3p)$ (Fig. 3b) since the two photon resonance condition is satisfied in the case of the $3s \rightarrow 3d$ transition.

Next we show the variation of the laser assisted excitation cross sections $\sigma^L(3s \rightarrow 3p)$ (Fig. 4a) and $\sigma^L(3s \rightarrow 3d)$ (Fig. 4b) with laser frequency ω (eV). For both transitions, we have taken the laser intensity as $I = 10^6 \text{ W cm}^{-2}$ and the collision velocity $v = 1$ a.u. We find that as the laser frequency increases from $\omega = 1.5$ eV, there is drop in $\sigma^L(3s \rightarrow 3p)$ (Fig. 4a) and rise in $\sigma^L(3s \rightarrow 3d)$ (Fig. 4b). This is because at $\omega = 1.5147$ eV the one photon resonance condition is met for the $3p$ and $3d$ transition, and these states are strongly coupled due to a large oscillator strength. With a further increase in laser frequency from $\omega = 1.54$ to 2.0 eV, we find both cross sections remain steady. However, around $\omega = 2.10$ eV, there is a jump in $\sigma^L(3s \rightarrow 3p)$ (Fig. 4a) due to the one photon resonance condition being met for the $3s \rightarrow 3p$ transition and $3s \rightarrow 3p$ is a dipole allowed transition. Also there is drop in the $\sigma^L(3s \rightarrow 3d)$ transition since it is a dipole forbidden transition with $(\Delta l = 2)$. Beyond this we find that $\sigma^L(3s \rightarrow 3p)$ remains greater than $\sigma^L(3s \rightarrow 3d)$ as expected.

In Fig. 5 we show the variation of the laser-assisted excitation cross section $\sigma^L(3s \rightarrow 3p)$ with collision velocity. We have taken the laser frequency to be

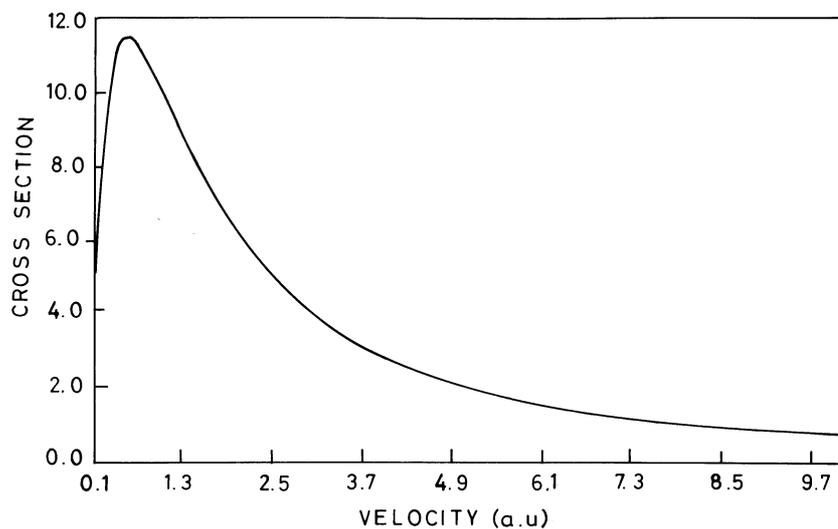


Fig. 5. Variation of the excitation cross section with collision velocity (in a.u.) for an intensity of $10^{10} \text{ W cm}^{-2}$ and $\omega = 2.10182$ eV.

$\omega = 2 \cdot 10182$ eV (equal to ω_{res}) and the laser intensity $I = 10^{10}$ W cm $^{-2}$. The variation of the cross section with velocity shows a rise at low velocities and then an exponential decrease of the cross section at higher velocities.

Table 1. Variation of transition probability with intensity for $v = 1$ a.u., $\omega = 2 \cdot 1$ eV (near the ω_{3s-3p} transition) and $b = 0 \cdot 01$ a.u.

Power (10^6 W cm $^{-2}$)	Collision probability	
	3s-3p transition	3s-3d transition
1	0.3186370	0.07485804
10	0.3186371	0.07485804
10^2	0.3186373	0.07485805
10^3	0.3186396	0.07485816
10^4	0.3186631	0.07485928
10^5	0.3188977	0.07487038
10^6	0.3211948	0.07497352

In Table 1 we show the effect of the laser intensity on the collision process, for a collision velocity $v = 1$ a.u., $b = 0 \cdot 01$ a.u. and $\omega = 2 \cdot 1$ eV (near the $3s \rightarrow 3p$ transition). It can be seen clearly that the collision probability increases with an increase in intensity.

4. Conclusion

We have described a combination of the non-perturbative quasi-energy approach for the radiation-atom interaction and the close coupling impact parameter method for the collision-aided radiative excitation of a dressed atom. The advantage of the non-perturbative treatment is that the interaction can be taken to all orders. Although we have presented our results only for the proton-atom excitation process, this method can be immediately generalised for any other ion-atom excitation.

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