Aust. J. Phys., 1992, **45**, 47–53

Excitation of Atomic Hydrogen by Proton Impact in the Presence of a Resonant Laser Field Using the First Order Magnus Approximation

Bhupat Sharma and Man Mohan

Department of Physics, University of Delhi,
Delhi 110 007, India.

Abstract

We consider proton collisions from hydrogen atoms in the presence of a laser beam (taken in the electric dipole approximation) that resonantly (or nearly resonantly) excites the hydrogen atoms from the 1s to the 2p state. The laser beam is linearly polarised with polarisation either parallel (longitudinal) or perpendicular (transverse) to the direction of incidence of the proton. A non-perturbative quasi-energy approach is used to describe the laser–atom interaction, while the first-order Magnus approximation is used to describe the collision dynamics in the presence of the nearly resonant laser beam. We have calculated the integrated cross section σ(2s) for the excitation of the 2s state. It is found that σ(2s) is small for longitudinal polarisation, as compared with transverse polarisation. We have also compared our field-free results obtained by using the first-order Magnus approximation to that obtained by the close-coupling approximation. Although both methods give excellent results, the former method is quite demanding in terms of computer time.

1. Introduction

In this paper we study the proton–hydrogen atom collision in the presence of a resonant laser field using the first-order Magnus approximation, first introduced by Alder and Winther (1960) to describe the multiple Coulomb excitation of deformed nuclei by energetic heavier ions. In the past this method has been applied to electron–atom scattering (Takayanagi 1963; Callaway and Bauer 1965), to proton–hydrogen atom scattering (Callaway and Dugan 1966; Geltman 1969; Baye and Heenen 1973), to ionisation of atoms by heavy ion impact (Eichler 1977) and to atomic and molecular processes in the presence of a laser field (Leasure *et al.* 1981; Sharma and Mohan 1985, 1986*a*, 1986*b*; Mohan and Prasad 1990, 1991).

In the present paper we use this approximation to study proton impact excitation of the 2s state of the hydrogen atom in the presence of a resonant laser field. The validity of this approximation for the process is investigated by calculating the total cross section for the 2s state excitation of the hydrogen atom during collisions with heavier ions (protons) in the presence and absence of the laser field. The atom is resonantly excited to the 2p state by the high intensity laser pulse and the collision induces a transition to the 2s state. In the limit where the spontaneous emission and the splitting of the 2s–2p degeneracy are neglected, the integrated cross section σ(2s) for the atom to undergo a transition to the 2s state would be infinite. This divergence of the cross section

0004-9506/92/010047$05.00

is a consequence of the fact that the collision induced coupling between 2s–2p states is, at large distances, of the form of a non-oscillatory dipole.

2. Theory

We suppose the hydrogen atom is initially in the ground state and is resonantly excited to the 2p state by a picosecond laser pulse. The laser beam induces the hydrogen atom into a dressed state, a time-dependent linear superposition of 1s and 2p states. While the laser is switched on the atom is bombarded with protons and after the collisions are over the laser is turned off. We have calculated the cross section $\sigma(2s)$ for the atom to undergo a transition to the 2s state. We find that $\sigma(2s)$ is not infinite because the long range dipole coupling between the 2s and 2p states oscillates at a small frequency close to the Rabi frequency.

The electric field vector $\boldsymbol{E}(t)$ of the laser beam is defined by $\boldsymbol{E}(t) = \hat{\boldsymbol{\epsilon}} E_0 \cos(\omega t)$, where $\hat{\boldsymbol{\epsilon}}$ is a unit polarisation vector, E_0 is the amplitude of the laser beam and ω is its frequency. In the impact parameter method (Bates 1958) the incident proton is assumed to travel in a classical straight-line constant-velocity path with velocity $\boldsymbol{v}$ and impact parameter $\boldsymbol{b}$. The relative coordinate of the proton is described by $\boldsymbol{R}(t) = \boldsymbol{b} + \boldsymbol{v}t$. Since the incident proton is relatively massive, its trajectory will not be affected appreciably by the laser pulse and we can neglect the coupling between the laser and incident projectile. We take the laser beam to be perpendicular to $\boldsymbol{v}$ and the polarisation to be linear, and so $\hat{\boldsymbol{\epsilon}}$ is either parallel to $\boldsymbol{v}$ (longitudinal polarisation) or perpendicular to $\boldsymbol{v}$ (transverse polarisation). We define E_1^0 and E_2^0 as the energies of the ground and the first excited states of the hydrogen atom. The detuning of the laser from resonance is defined by $\epsilon_{21} = E_2^0 - E_1^0 - \omega$ (we use atomic units throughout this paper).

The collision duration τ_c is generally much shorter than the laser pulse duration (for a proton impact energy of 100 keV the collision duration is of the order of 10^{-15} s which is less than the laser pulse duration). Subject to this condition the particles situated in the radiation field collide. It is well established that an atom situated in a radiation field is characterised by a set of quasi-energy states (Shirley 1965; Zeldovich 1973; Chu 1985). We assume $\epsilon_{21} < \omega$, so that the atom is interacting with a nearly resonant field. Using the rotating wave approximation, the quasi-energy states (or the dressed states) of the hydrogen atom in the presence of a nearly resonant field with detuning ϵ_{21} are defined by (see e.g. Sharma and Mohan 1986*a*)

$$\Phi_n(\boldsymbol{r}, t) = \exp[-\mathrm{i}(E_1^0 + \lambda_n)t][a_1^n \,|\, 1\mathrm{s}\rangle + a_2^n \,|\, 2\mathrm{p}\rangle \exp(-\mathrm{i}\,\omega t)]\,, \tag{1}$$

where $\lambda_n = \frac{1}{2}\epsilon_{21} \pm \frac{1}{2}(|\,\epsilon_{21}\,|^2 + |\,V_{12}\,|^2)^{1/2}$ are the set of quasi-energies, and where $V_{12} = -0\cdot 744 E_0$ is the Rabi frequency and $\{a_i^n\}$ are the eigenvectors corresponding to the quasi-energies $\{\lambda_n\}$. The polarisation of the 2p state is the same as that of the laser. In equation (1), $|\,1\mathrm{s}\,\rangle$ is the normalised vector representing the bare atomic ground state. With $|\,2\mathrm{p}\,\rangle$ the normalised vector representing the bare $|\,2\mathrm{p}\,\rangle$ state with angular momentum projection quantum number m along $\boldsymbol{v}$, the vector is $|\,2\mathrm{p}_+\,\rangle$ for longitudinal polarisation and $|\,2\mathrm{p}_-\,\rangle$ for transverse polarisation, where

$$|\,2\mathrm{p}_\pm\rangle = \sqrt{\tfrac{1}{2}}(|\,2\mathrm{p}_{+1}\rangle \pm |\,2\mathrm{p}_{-1}\rangle)\,. \tag{2}$$

The total wavefunction Ψ of the system in terms of the quasi-energy states is written as (Gersten and Mittleman 1976)

$$\Psi = \sum_n C_n(t)\,\Phi_n(\boldsymbol{r},\,t) + \exp(-\mathrm{i}\,E_2^0 t)[C_{2\mathrm{s}}(t)\,|\,2\mathrm{s}\rangle + C_{2\mathrm{p}_-}\,|\,2\mathrm{p}_-\rangle] \quad \text{(longitudinal polarisation)} \tag{3a}$$

$$= \sum_n C_n(t)\,\Phi_n(\boldsymbol{r},\,t) + \exp(-\mathrm{i}\,E_2^0 t)[C_{2s}(t)\,|\,2\mathrm{s}\rangle + C_{2\mathrm{p}_0}^0\,|\,2\mathrm{p}_0\rangle + C_{2\mathrm{p}_+}\,|\,2\mathrm{p}_+\rangle] \quad \text{(transverse polarisation)}. \tag{3b}$$

Note that reflection symmetry in the $\boldsymbol{v}\,.\,\boldsymbol{b}$ plane implies $C_{2\mathrm{p}+}(t) = 0$ in the case of longitudinal polarisation.

Using the wavefunctions (3) and the total Hamiltonian of the system $H_0(\boldsymbol{r}) + V(\boldsymbol{r},t) + V_{\mathrm{L}}^{\mathrm{A}}(\boldsymbol{E},\omega)$, where $H_0(\boldsymbol{r})$ is the unperturbed Hamiltonian of the atom, $V(\boldsymbol{r},t)$ is the time-dependent interaction potential and $V_{\mathrm{L}}^{\mathrm{A}} = -\boldsymbol{r}\,.\,\hat{\epsilon}E_0\cos(\omega t)$ is the interaction of the atom with the laser field (in the dipole approximation), the time-dependent Schrödinger equation yields the set of coupled first-order linear differential equations

$$\mathrm{i}\,\frac{\partial \mathbf{C}}{\partial t} = \mathbf{Q}(t)\,\mathbf{C}(t)\,. \tag{4}$$

Here $\mathbf{C}(t)$ is the column matrix with elements $C_1(t)$, $C_2(t)$, etc. with the initial condition $\mathbf{C}_i(-\infty) = \delta_{oi}$ where i labels the initial state, and $\mathbf{Q}(t)$ is the coupling matrix composed of the matrix elements of the interaction potential

$$V(\boldsymbol{r},\,t) = \frac{1}{R(t)} - \frac{1}{|\,\boldsymbol{r} - \boldsymbol{R}(t)\,|} \tag{5}$$

between the atom and the projectile.

We now employ the dipole approximation used by Seaton (1962). With the expansion

$$\frac{1}{|\,\boldsymbol{r} - \boldsymbol{R}(t)\,|} = \sum_{\mu=0}^{\infty} P_\mu(\hat{\boldsymbol{r}}\,.\,\hat{\boldsymbol{R}})\,\frac{r_<^\mu}{r_>^{\mu+1}}; \qquad \hat{\boldsymbol{r}}\,.\,\hat{\boldsymbol{R}} = \cos\theta\,, \tag{6}$$

where θ is the angle between $\boldsymbol{r}$ and $\boldsymbol{R}(t)$ and $r_<$ is the smaller of r and R, and $r_>$ the greater, the only nonzero contribution to the optically allowed transitions comes from the odd values of μ satisfying $a_0 \le \mu \le (l_j + l_k)$, l_j and l_k being the atomic angular momentum quantum numbers. The dominant contribution comes from $\mu = 1$. Using only this term $|\,\boldsymbol{r} - \boldsymbol{R}(t)\,|^{-1}$ may be replaced by $\boldsymbol{r}\,.\,\boldsymbol{R}(t)/r_>^3$, and if we assume that $r_> = R(t)$ then we have

$$\langle j\,|\,1/[\boldsymbol{r} - \boldsymbol{R}(t)]\,|\,k\rangle = \langle j\,|\,\boldsymbol{r}\,.\,\boldsymbol{R}(t)/R^3\,|\,k\rangle\,. \tag{7}$$

So long as $R(t)$ is greater than r, the radius of the atomic electron, this approximation should be valid; it is therefore reasonable to use it for impact parameters greater than some mean atomic radius. When the full matrix $\mathbf{Q}$ was set up, several of the elements had factors $\exp(\pm\mathrm{i}\,\phi)$. A simple unitary transformation of $\mathbf{Q}$ removes these factors.

Using the first-order Magnus approximation, the solution of equation (4) at $t = +\infty$ is given by (Callaway and Bauer 1965; Pechukas and Light 1966; Jamieson *et al.* 1975; Sharma and Mohan 1986*a*; Mohan and Prasad 1990)

$$\mathbf{C}(+\infty) = \mathbf{U}\exp(-\mathrm{i}\,\mathbf{M}_\mathrm{D})\,\mathbf{U}^\dagger\mathbf{C}(-\infty)\,, \tag{8}$$

where $\mathbf{M}_\mathrm{D}$ is the diagonalised matrix whose diagonal elements are the eigenvalues of the matrix $\mathbf{M}$ defined by

$$\mathbf{M}(+\infty) = \int_{-\infty}^{+\infty} \mathbf{Q}(t')\,\mathrm{d}t' \tag{9}$$

and $\mathbf{U}$ is a unitary matrix whose columns are the eigenvectors of the matrix $\mathbf{M}$ satisfying the equation

$$\mathbf{M}_\mathrm{D} = \mathbf{U}^\dagger\mathbf{M}\mathbf{U}\,. \tag{10}$$

The exponential operator in (8) is unitary and therefore the normalisation of the vector $\mathbf{C}$ is preserved at all times. The probability for a transition to the 2s state at a given impact parameter is then $P(2\mathrm{s}) = |\,C_{2\mathrm{s}}(+\infty)\,|^2$. This probability can then be integrated with respect to the impact parameter to give the total cross section

$$\sigma(2\mathrm{s}) = 2\pi \int_0^\infty bP(2\mathrm{s})\,\mathrm{d}b\,. \tag{11}$$

The matrix $\mathbf{M}(+\infty)$ can easily be evaluated in terms of K_0 and K_1, which are modified Bessel functions. This yields a complex matrix because of the presence of i in $\mathbf{Q}$. The matrix may be made real to speed up the calculation by suitable changes in the basis set; in particular, multiplying the 2p wavefunctions by +i will yield a real $\mathbf{M}$ matrix.

3. Results and Discussion

Before mentioning the numerical results for $\sigma(2\mathrm{s})$, let us first describe in brief the general trend of the variation of the cross section with velocity for longitudinal and transverse laser beam. According to the first-order perturbation theory, the ratio of the probabilities for transverse ($\perp$) and longitudinal ($\|$) polarisation is given by

$$\frac{P_\perp(2\mathrm{s})}{P_\|(2\mathrm{s})} = \frac{(a_2^1)_\perp}{(a_2^1)_\|}\left|\frac{K_1(\beta)}{K_0(\beta)}\right|^2, \tag{12}$$

where $(a_2^1)_\perp$ and $(a_2^1)_\|$ are the components of the eigenvectors a_i^n belonging to quasi-energy $\lambda = \lambda_\perp$ for transverse and longitudinal polarisations respectively, and $\beta = (\epsilon_{21}-\lambda_1)b/V$. Note that for $\beta \ll 1$ we have $K_0(\beta) \approx -\ell\mathrm{n}\,\beta$ and $K_1(\beta) \approx 1/\beta$, so that over the range of impact parameters $a_0 \le b \le V/(\epsilon_{21}-\lambda_1)$ we have $P_\|(2\mathrm{s})$ smaller than $P_\perp(2\mathrm{s})$ by a factor of the order of $(\beta\,\ell\mathrm{n}\,\beta)^2$. We also note that the collision induced dipole coupling between the 2s and 2p states for a resonant laser field ($\epsilon_{21} = 0$) is given by

$$-\langle 2\mathrm{s}\,|\,\boldsymbol{r}\,.\,\boldsymbol{R}(t)/R^3\,|\,2\mathrm{p}\rangle\exp(-\mathrm{i}\,V_{12}\,t) = \pm\,3R^{-3}(\hat{\epsilon}\,.\,\boldsymbol{R})\exp(-\mathrm{i}\,V_{12}\,t)\,, \tag{13}$$

where for the resonant laser field we have $\epsilon_{21}-\lambda_1 \approx V_{12}$. For $\beta \ll 1$ the oscillatory term $\exp(-\mathrm{i}\, V_{12}\, t)$ can be set equal to unity during the collision time which is of the order of b/v. For transverse polarisation we find $\hat{\epsilon}\,.\,\boldsymbol{R} = \hat{\epsilon}\,.\,\boldsymbol{b}$ and the coupling is even in time t, but for longitudinal polarisation we find $\hat{\epsilon}\,.\,\boldsymbol{R} = Vt$ and the coupling is odd in t. Hence, the time average of the coupling over the collision duration vanishes for longitudinal polarisation (but not for transverse polarisation). It follows that the average coupling is stronger in the transverse case and hence $P_\perp(2\mathrm{s})$ is larger than $P_\parallel(2\mathrm{s})$.

Table 1. Total cross section $\sigma(2\mathrm{s})$ (in units of 10^{-17} cm^2) of atomic hydrogen against proton impact energy in the presence of a resonant laser field

The intensity of the laser beam is $I = 10^{10}$ W cm^{-2}

Energy (keV)	$\sigma_\parallel(2\mathrm{s})$	$\sigma_\perp(2\mathrm{s})$	$\sigma_{\mathrm{FF}}(2\mathrm{s})^{\mathrm{A}}$	$\sigma_{\mathrm{FF}}(2\mathrm{s})^{\mathrm{B}}$
15	4·35	174·09	3·19	3·41
25	6·86	58·19	3·39	3·98
40	4·32	29·56	2·99	2·33
50	2·11	18·09	1·13	1·39
60	1·11	12·30	0·97	0·82
75	1·00	8·9	0·45	0·42
145	0·133	2·61	0·051	0·040
200	0·112	1·10	0·0079	0·0087

[A] Our field-free cross section using the first-order Magnus method.
[B] Field-free results of Shakeshaft (1978) using the close-coupling method.

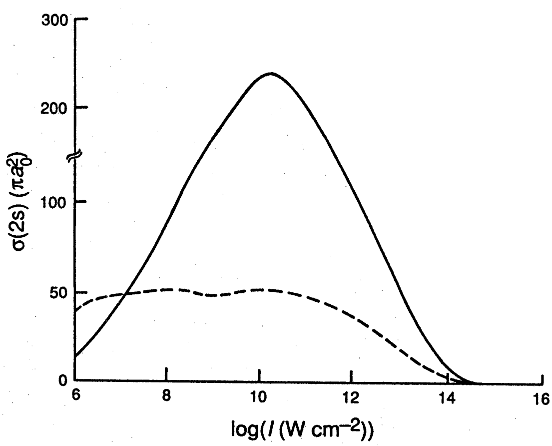

Fig. 1. Integrated cross section $\sigma(2\mathrm{s})$ against intensity of the field. The proton impact energy is 100 keV. The solid and dashed curves refer to transverse and longitudinal polarisation respectively.

In Table 1 we show the variation of $\sigma(2\mathrm{s})$ against proton impact energy for a transverse and longitudinal polarised laser beam. The intensity of the laser beam is taken to be $I = 10^{10}$ W cm^{-2}. The cross sections are obtained by using the solution (8). As expected, we find that $\sigma_\perp(2\mathrm{s})$ is greater than $\sigma_\parallel(2\mathrm{s})$ for all impact energies. We have also shown in Table 1 the field-free ($E_0 = 0$) cross section

$\sigma_{\rm FF}(2s)$ obtained using the Magnus approximation. Upon comparison of our field-free results with those obtained by Shakeshaft (1978) with the close-coupling method, we find that our results are in good agreement which shows that the Magnus approximation (or the diagonalisation method) is also valid for atomic scattering problems. We also notice that $\sigma_\perp(2s)$ is 10–100 times larger than its value in the absence of the laser field, which shows that the near resonant laser field greatly enhances the cross sections.

In Fig. 1 we show the effect of the laser intensity I on $\sigma(2s)$ for a fixed proton energy of 100 keV. We find that $\sigma_\perp(2s)$ increases linearly with I and has a maximum value at $I = 10^{10}\ \mathrm{W\,cm^{-2}}$. With a further increase in intensity $\sigma_\perp(2s)$ starts falling. However, $\sigma_\parallel(2s)$ varies quite slowly with an increase in intensity, and remains somewhat stationary near the region where $\sigma_\perp(2s)$ peaks. Such behaviour can be understood as follows. When the intensity of the laser beam is quite weak ($I \to 0$) the cross sections reduce to those obtained in the field-free case. However, with an increase in intensity the $|\,2p\,\rangle$ state population also increases and becomes roughly constant, equal to a half at and above the intensity $I_{\rm p}$ where $\sigma_\perp(2s)$ peaks. The main contribution to $\sigma_\perp(2s)$ comes from the range of impact parameters $a_0 \le b \le V/V_{12}$, while for greater impact parameters the 2s–2p coupling averages to zero over the collision time b/v. Since V_{12} increases with intensity (roughly as the square root of I), the significant range of impact parameter decreases. This leads to a decrease in $\sigma_\perp(2s)$. Consequently, $\sigma_\perp(2s)$ rises and then falls as I increases.

On the other hand, $\sigma_\parallel(2s)$ varies quite slowly compared with $\sigma_\perp(2s)$ and levels off at an intensity $I_{\rm p}$ where $\sigma_\perp(2s)$ peaks. We note from (13) that the 2s–2p coupling for longitudinal polarisation is odd in t and therefore averages to zero over the collision time if the factor $\exp(-{\rm i}\,V_{12}\,t)$ is set equal to unity. However, the $\sin(V_{12}\,t)$ component of $\exp(-{\rm i}\,V_{12}\,t)$ combines to make the longitudinal 2s–2p coupling even in t, so that it no longer averages to zero for $b \le V/V_{12}$. We also note that the magnitude of $\sin(V_{12}\,t)$ increases with I. Hence, while the range of b decreases with increasing I, the magnitude of the longitudinal 2s–2p coupling increases so that the two effects combine to make $\sigma_\parallel(2s)$ roughly constant for $I > I_{\rm p}$, as can be seen from Fig. 1. Finally Gersten and Mittleman (1976) also obtained analytic solutions for the cross sections of the H(2s) excitation by electron impact in a resonant laser field. However, they did not show the numerical results for the intensity dependence of $\sigma(2s)$.

4. Conclusions

The quasi-energy technique which is non-perturbative in nature provides a simple and elegant picture of atomic scattering problems in the high intensity limit (Chu 1985). We have used the quasi-energy method for the radiation part without allowance for the fine structure of the atom. We have also shown that the close-coupling diagonalisation method gives results close to other theoretical methods. Finally, our method is quite general and can be extended to other ion (atom)–atom collisions in the presence of a laser field.

Acknowledgments

The authors thank the CSIR, UGC and DST for financial support.

References

Alder, K., and Winther, A. (1960). *Dan. Vid. Selsk. Mat. Fys. Medd.* **32**, 1.
Bates, D. R. (1958). *Proc. R. Soc. London* A **245**, 299.
Baye, D., and Heenen, P. H. (1973). *J. Phys.* B **6**, 105.
Callaway, J., and Bauer, E. (1965). *Phys. Rev* A **140**, 1072.
Callaway, J., and Dugan, A. F. (1966). *Phys. Lett.* A **21**, 196.
Chu, S. I. (1985). *Adv. At. Mol. Phys.* **21**, 197.
Eichler, J. (1977). *Phys. Rev.* A **15**, 1856.
Geltman, S. (1969). 'Topics in Atomic Collision Theory', p. 199 (Academic: New York).
Gersten, J. I., and Mittleman, M. H. (1976). *Phys. Rev.* A **13**, 123.
Jamieson, M. J., Kalaghan, P. M., and Dalgarno, A. (1975). *J. Phys.* B **8**, 2140.
Leasure, S. C., Milfeld, K. F., and Wyatt, R. E. (1981). *J. Chem. Phys.* **74**, 6197.
Mohan, M., and Prasad, V. (1990). *Phys. Lett.* A **148**, 327.
Mohan, M., and Prasad, V. (1991). *J. Phys.* B. **24**, L81.
Pechukas, P., and Light, J. C. (1966). *J. Chem. Phys.* **44**, 3897.
Seaton, M. J. (1962). *Proc. Phys. Soc. London* **79**, 1103.
Shakeshaft, R. (1978). *Phys. Rev.* A **13**, 1930.
Sharma, B., and Mohan, M. (1985). *Chem. Phys. Lett.* **118**, 553.
Sharma, B., and Mohan, M. (1986*a*). *J. Phys.* B **19**, L433.
Sharma, B., and Mohan, M. (1986*b*). *Ind. J. Pure Appl. Phys.* **24**, 106.
Shirley, J. H. (1965). *Phys. Rev.* B **138**, 979.
Takayanagi, K. (1963). *Prog. Theor. Phys. Suppl.* **25**, 43.
Zeldovich, Ya. B. (1973). *Sov. Phys. Usp.* **16**, 427.

Manuscript received 20 February, accepted 29 August 1991