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Coherent propagation of laser pulse in a resonant medium and its application to isotope separation

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Abstract. The action of an intense laser pulse on resonant and slightly off-resonant atoms is considered. The induced oscillating average dipole moment in the two cases have different characteristics. This leads to a possible scheme of isotope separation by the action of a field gradient in the laser beam itself. Reaction of the medium back on the pulse is studied through the semi-classical Bloch-Maxwell equations to ascertain the effectiveness of the pulse over long propagation distances.

1. Introduction

Studies of the coherent propagation of short laser pulses in a resonant medium have brought to light a number of new phenomena like self-induced transparency (SIT) (McCall and Hahn 1967, 1969) and opened up new possibilities like selective excitation and separation of a species in an isotopic mixture. Those phenomena have a critical dependence on parameters like pulse-width and pulse-area. For example, in the case of SIT, the leading edge of the pulse inverts the atomic population and the trailing edge returns it to its initial value by stimulated emission. The energy lost by the leading edge to the atoms is fed back to the pulse at the trailing edge and the pulse thus propagates without net energy loss (transparency). For this to be realised, the pulse has to be much shorter than the relaxation times of the medium and the intensity should be sufficient to effect the population inversion. The parameter called the area under the pulse is a generalisation of the following idea. A two-level atom under resonance with the applied field oscillates between the two states with the Rabi frequency ω_R. If the width of the square pulse (envelope) is $n\pi/\omega_R$, the pulse is said to be an $n\pi$ pulse, with area $n\pi$. It is clear then that a π pulse leaves the atoms in the excited state while a 2π pulse takes them through a complete cycle and leaves them in the ground state. A $2n\pi$ pulse, as it propagates deep into the medium breaks up into n separate 2π pulses, because of the attenuation and amplification of parts of the pulse as they encounter atoms at the lower and upper levels respectively. These pulse-shaping properties of the medium (Lamb 1971) have to be taken into account when estimating the effectiveness of a pulse in producing a specific change in the atomic properties of the medium because of the dependence of the shape on depth.

In the context of isotope separation using lasers, Diels (1976) has proposed a scheme for the selective excitation of one species in a mixture based on the following idea. If the parameters of the pulse are chosen in such a way that it appears as a π pulse to the atoms to be selected and a 2π pulse to the other (majority) species, the atoms of the former will be left in the excited state and the latter in the ground state at the end of the pulse. Since the majority of the atoms sees a 2π pulse, the area will remain practically constant by virtue of SIT. Another scheme (Friedmann and Wilson 1976) uses the above π–2π concept to achieve not simply selective excitation but separation as well. A pulse on first traversing the atomic beam leaves only the desired species population-inverted. On being reflected by a mirror it re-traverses the atomic beam and returns the atoms to their ground state. The absorption in the forward flight of the photon beam and the stimulated emission in the return flight impart a total momentum of $2\hbar\omega/c$ to the atoms to be selected.

In this paper, we propose a selective photo-deflection scheme based on the following idea. When a two-level atom is in resonance with the laser field, the oscillating dipole moment which results from the coherent superposition of the two states is $\pi/2$ out of phase with the applied field. For a slight mismatch in the frequency, the average dipole moment acquires an in-phase component which will experience a deflecting force from the radial gradient of the laser field. Tuning the field frequency to the line-centre of the majority species and choosing the height and width of the pulse such that it is a 2π pulse for the majority atoms and a 4π one for the minority atoms we can achieve the following. The majority species suffer on an average neither change of state nor deflection as a result of a passage of the pulse. The atoms of the minority species, on the other hand, are also left in their ground state but suffer a deflection along the gradient of the laser field. Frequency pulling effects would be small in view of the tuning to the majority species. Pulse break-up effects would also be negligible because it is a 2π pulse to the majority of atoms. There would, however, be a small red-shift in the carrier frequency as a result of the momentum imparted to the minority atoms. The overall performance of this scheme would depend very much on the availability of laser pulses with an optimum intensity profile leading to a maximum value for the momentum imparted. We shall return to a discussion of these problems after a brief outline of the theory of two-level atoms and pulse propagation in a medium composed of two species of two-evel atoms.

2. Two-level atom theory

In order to clarify the principle of the proposed method, we briefly outline the theory of a single atom in resonance with the field $E = E_0 \cos \omega_0 t$ of a light wave.

The Schrödinger equation for the system in the semi-classical approximation is given by

$$i\hbar\frac{\partial\phi}{\partial t} = (H_0-\boldsymbol{\mu}.\boldsymbol{E}_0 \cos \omega_0 t)\psi \tag{1}$$

where H_0 is the Hamiltonian of the atom with eigenstates ϕ_1 and ϕ_2 corresponding to the energies E_1 and E_2 respectively. Writing

$$\psi = a_1(t)e^{-iE_1t/\hbar}\phi_1(x)+a_2(t)e^{-iE_2t/\hbar}\phi_2(x) \tag{2}$$

we obtain from equation (1)

$$\left.\begin{aligned}\mathring{a}_1 &= i\frac{G}{2}a_2e^{i(\omega_0-\omega)t}\\ \mathring{a}_2 &= i\frac{G}{2}a_1e^{-i(\omega_0-\omega)t}\end{aligned}\right\} \tag{3}$$

where

$$\omega = (E_2-E_1)/\hbar,\ \ G = (\phi_1|\boldsymbol{\mu}.\boldsymbol{E}_0|\phi_2)/\hbar. \tag{4}$$

In deriving equation (3) we have assumed that $\omega_0 \simeq \omega$ and hence used the rotating wave approximation according to which the off-resonant terms involving $\exp(\pm i(\omega+\omega_0)t)$ are neglected. Equation (3) may now be recast in the fimiliar Bloch form

$$\left.\begin{aligned}\mathring{R}_1 &= \delta R_2\\ \mathring{R}_2 &= -\delta R_1+GR_3\\ \mathring{R}_3 &= -GR_2\end{aligned}\right\} \tag{5}$$

where we have defined

$$\left.\begin{aligned}R_1 &= a_1a_2^*e^{-i\delta t}+c.c.\\ R_2 &= -ia_1a_2^*e^{-i\delta t}+c.c.\\ R_3 &= |a_2|^2-|a_1|^2\end{aligned}\right\} \tag{6}$$

and $\delta = \omega_0-\omega$ as the frequency mismatch. It is clear that R_3 represents physically the population difference between the two levels. The meaning of the components R_1 and R_2 may be seen by considering the expression for the average dipole moment along the direction of E_0 given by

$$\begin{aligned}\mu(t) &\equiv \langle\psi|\hat{\mu}|\psi\rangle = \mu_{12}a_1a_2^*e^{-i\omega t}+c.c.\\ &= \mu(R_1 \cos \omega_0 t-R_2 \sin \omega_0 t)\end{aligned} \tag{7}$$

where $\mu = \mu_{12} = (\phi_1 | \ddot{\mu} | \phi_2)$. It is now seen from equation (7) that R_1 is the component of the average dipole moment oscillating in phase with the field and R_2, the component $\pi/2$ out of phase. Hence, R_1 and R_2 characterize the dispersion and absorption respectively.

It is easy to write the solutions of equation (5) corresponding the initial condition $\mathbf{R}(0) = (0, 0, -1)$ (which implies that the atom is in the state ϕ_1 at $t = 0$). These are

$$\left.\begin{aligned} R_1 &= -\frac{G\delta}{\Omega^2}\,(1-\cos\Omega\,t) \\ R_2 &= -\frac{G}{\Omega}\sin\Omega t \\ R_3 &= -1+\frac{G^2}{\Omega^2}\,(1-\cos\Omega t) \end{aligned}\right\} \tag{8}$$

where $\Omega = \sqrt{\delta^2+G^2}$. Note that at exact resonance ($\delta = 0$) R_1 is zero and the atom will not experience any force due to a gradient in the laser field. On the other hand for an atom with a frequency $\omega_0+\Delta(\delta = \Delta)$, R_1 is non-zero and there is a possibility of the atom experiencing a gradient force. Thus the single-atom two-level theory brings out the fact that an atom with a frequency mismatch will be deflected by the gradient in the incident field. However, when we have a collection of atoms of both types as in an atomic beam we need to consider the details of propagation of a laser pulse in such a medium. We discuss this in the next section.

3. Pulse Propagation

The laser field induces oscillating dipole moments in that atoms (in the sense of expection values) and when we have a large number of atoms these constitute a macroscopic dipole moment P per unit volume which can exchange energy with the radiation field. The field experienced by the atom is then not the applied field but the one obtained by solving the semiclassical Bloch-Maxwell equations self-consistently. In addition to the system of equations (5) we have also the wave equation

$$\nabla^2 E - \frac{n^2}{c^2}\frac{\partial^2 E}{\partial t^2} = \mu_0 \frac{\partial^2 P}{\partial t^2} \tag{9}$$

where n is the refractive index and μ_0 is the free space permeability. We now take the applied field in the form of a plane wave so that inside the medium we have a field of the form

$$E(Z, t) = \epsilon(Z, t)\cos(k_0 Z - w_0 t + \phi) \tag{10}$$

where $\phi(Z, t)$ is a phase developed during the propagation and $\epsilon(Z, t)$ is the amplitude. The medium polarization is similarly described as

$$P = N\mu[R_1(Z, t)\cos(k_0 Z - \omega_0 t + \phi) + R_2(Z, t)\sin(k_0 Z - \omega_0 t + \phi)] \tag{11}$$

where N is the number of atoms per unit volume. Substituting equations (10) and (11) in equation (9) and making use of the usual slowly varying envelope approximation, we obtain

$$\frac{\partial \epsilon}{\partial Z} + \frac{n}{c}\frac{\partial \epsilon}{\partial t} = \frac{\omega_0 c \mu_0 \mu N}{2n} R_2$$

$$\epsilon\left(\frac{\partial \phi}{\partial Z} + \frac{n}{c}\frac{\partial \phi}{\partial t}\right) = \frac{\omega_0 c \mu_0 \mu N}{2n} R_1. \tag{12}$$

We now consider the medium to consist of two species of atoms with the transition frequencies centred around ω_0 and $\omega_0 + \Delta$ with the lineshape function $g(\delta w)$. We shall characterize their Bloch vectors by $\boldsymbol{R}$ and $\boldsymbol{S}$. In equation (12) we will have now to use in place of R_j the average

$$C_R < R_j > + C_S < S_j >$$

where C_R and C_S are the fractions of the two species of atoms with

$$< R > = \int_{-\infty}^{\infty} d(\delta\omega) g(\delta\omega) R(\delta\omega, Z, t). \tag{13}$$

Two other modifications must be introduced in equation (5) for $\boldsymbol{R}$ and $\boldsymbol{S}$. First δ must be replaced by $\delta - \phi$. Second, for long pulses we would have to add the terms involving the phase relaxation time T_2 and the energy relaxation time T_1. We may also note that $\boldsymbol{R}$, $\boldsymbol{S}$ and G are functions of Z, t with $G = \mu\epsilon/\hbar$. The complete set of equations which are relevant to our discussion may then be rewritten as

$$\begin{aligned} \mathring{R}_1 &= (\delta - \mathring{\phi})R_2 - R_1/T_2 \\ \mathring{R}_2 &= -(\delta - \mathring{\phi})R_1 - R_2/T_2 + GR_3 \\ \mathring{R}_3 &= -GR_2 - (R_3 - R_{30})/T_1. \end{aligned} \tag{14}$$

$$\frac{\partial G}{\partial Z} + \frac{n}{c}\frac{\partial G}{\partial t} = \alpha[C_R < R_2 > + C_S < S_2 >]$$

$$G\left(\frac{\partial \phi}{\partial Z} + \frac{n}{c}\frac{\partial \phi}{\partial t}\right) = \alpha[C_R < R_1 > + C_S < S_1 >]. \tag{15}$$

Here R_{30} is the initial population difference and $\alpha = \frac{\mu_0 \omega_0 C \mu^2 N}{2n\hbar}$. The equations for S are obtained from equation (14) by replacing $\boldsymbol{R}$ by $\boldsymbol{S}$ and δ by $\delta + \Delta$.

4. Isotope separation

In the context of isotope separation the two species of atoms correspond to two distinct isotopes with an isotopic shift Δ. The concentration of the unwanted isotope $C_R \simeq 1$ while that of the isotope to be selected $C_s \simeq 0$. The laser pulse is tuned to the centre frequency of the unwanted isotope. This has three important consequences. First, the in-phase component of the polarization $< R_1 >$ is close to zero and it follows from equation (15) that the phase modulation ϕ of the pulse during the passage through the isotopic mixture would be negligible. Thus the "frequency pulling" effect would be small and ϕ may be omitted in equation (14). Second, the pulse shape during its propagation will not be affected significantly by the presence of the other isotope. For example, one may choose the pulse amplitude G and its duration T ($T \ll T_1, T_2$) so that it acts as a 2π pulse for the majority isotope and as a 4π pulse for the other. Such a pulse remains practically a 2π pulse over its entire period of propagation. A third consequence is that since $< R_1 >$ is close to zero the majority isotope is unaffected by the gradient in the laser field. The component $< S_1 >$ is, however, non-zero and is effective is selectively deflecting the second isotope by virtue of a gradient in the laser field. Such a gradient may be introduced in practice by suitably tailoring the incident laser pulse. For instance, one can have the laser field polarized along X axis with a gaussian behaviour along X and nearly flat along Y, propagating in the Z direction. The geometrical arrangement required would be that the atomic beam in the form of a ribbon streaming in the Y direction with its thickness measured along X axis while the laser beam is incident in the Z direction as shown in Figure 1. The width of the gaussian profile may ideally be chosen to correspond to the thickness of the atomic beam. The net impulse experienced by an average atom of the isotope to be selected is then given by

$$p = \frac{\hbar}{2} \int_0^T < \quad \frac{\partial G}{\partial x} \, dt. \tag{16}$$

One has then to choose the pulse parameters so as to maximize the impulse p.

We now consider the requirements on the parameters of the pulse. A simple gaussian field profile of the laser pulse of the form $G = G_0 e^{-x^2/\sigma^2}$ would produce a maximum force on the atom encountered around $x = \sigma/\sqrt{2}$. The net impulse imparted per pulse would then be proportional to the duration T of the pulse on the basis of a single-atom picture. It is indeed possible to increase T without violating the short pulse requirement. The latter requirement only specifies that the Rabi-frequency for both the species of atoms must be much larger than the increase relaxation times T_2^{-1} and T_1^{-1}. As long as G satisfies this condition, a large T would only take the atoms through a large number of cycles. For a 2π pulse (i.e., $GT = 2\pi$), we find that for imparting a momentum

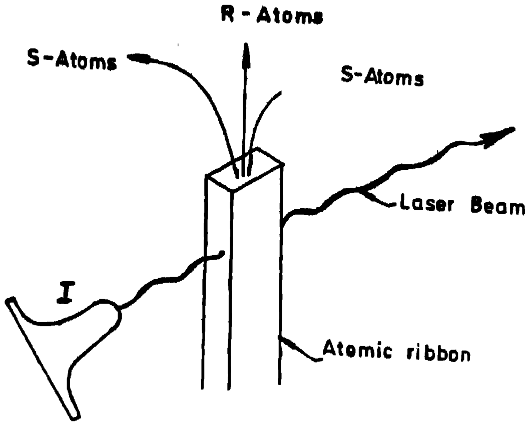

Figure 1. A schematic of the photo-deflection method; the R-atoms are of the majority species to be rejected and S-atoms are of the minority species to be selected. I is the intensity profile along the width of the laser beam.

of $\hbar\omega/c$ per pulse, the width of the gaussian $\sigma \sim n\lambda$. In such a case, if at all it is feasible, the effectiveness of the pulse is confined to a rather small section of the atomic beam; moreover, the carrier frequency of the light wave would acquire a large spread. It may be more practicable to have a $2n\pi$ pulse with G close to the isotopic shift Δ so that the induced dipole moment stays close to its maximum value while $\sigma \sim n\lambda$. For $n \sim 1000$, this implies a pulse duration of the order of 1μ sec. Finally, to achieve a sizable deflection it would be necessary to apply a large number of these pulses before the atoms leave the interaction region. Economy would require re-using the same pulse a number of times by reflecting it by means of a mirror-cavity arrangement.

5. Results and Discussion

We have solved the Bloch-Maxwell equations (14) and (15) for pulse propagation numerically for some typical values of the parameters. Preliminary results indicate that the effects introduced by the medium-pulse interaction do not adversely affect the qualitative conclusions drawn from the single-atom picture discussion in section 2.

For obtaining the solutions of equations (14) and (15) we have chosen the following initial and boundary conditions on $G(Z,t)$, $R(Z,t)$ and $S(Z,t)$:

$$G(0,t) = \begin{cases} G_0 & 0 < t < T \\ 0 & t \geqslant T \end{cases}$$

$$R_1(Z_1, 0) = R_2(Z, 0) = S_1(Z, 0) = S_2(Z, 0) = 0$$

$$R_3(Z, 0) = S_3(Z, 0) = -1.$$

We next introduce the new variables, $\tau = t - Z/c$ and $Z' = Z$ so that equation (15) takes the form

$$\frac{dG}{dZ'} = \alpha[C_R \langle R_2 \rangle + C_s \langle S_2 \rangle]. \tag{15A}$$

The solution is propagated in the mesh (Z_n', τ_m) by letting G evolve along the Z axis according to equation (15A) and $R(S)$ along the τ axis from one mesh point to its neighbour (Icsevgi and Lamb 1969). The modification in the pulse shape as it propagates into the medium is illustrated in figure 2. Figure 2 depicts

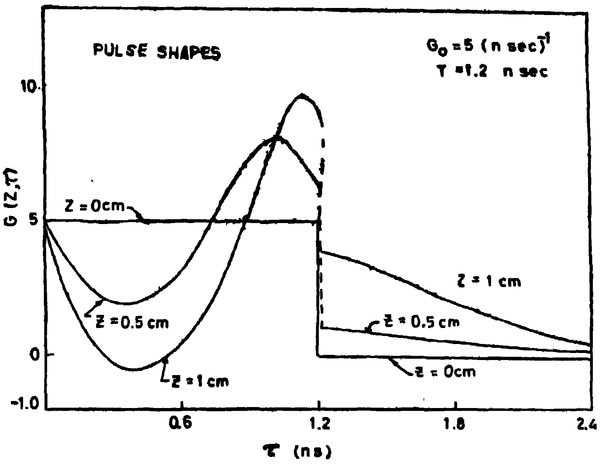

Figure 2. Distortion of a pulse during its propagation in the medium.

the behaviour of $G(Z,t)$ in time t for two different values of the depth Z inside the medium and for a typical set of parameters, viz. $G_0 = 5$ $(n \text{ sec})^{-1}$, $T = 1{\cdot}2$ $(n \text{ sec})^{-1}$, $\alpha = 6{\cdot}46$ $(n \text{ sec})^{-1}$, width of the gaussian atomic line profile $\delta = 0{\cdot}1$

$(n\,\text{sec})^{-1}$, isotopic shift $\Delta = 8{\cdot}4\ (n\,\text{sec})^{-1}$ and the peak wavelength $\lambda = 5915$ Å. It is seen from figure 2 that the area of the pulse is preserved as the pulse propagates in the medium. This is to be expected on the basis of the area-theorem because the applied pulse is a 2π pulse.

We need a gradient in the intensity of the laser beam to realize the deflection of the desired species. If we choose the field strength at some point in the cross-section of the laser beam to give us a 2π pulse, the pulse would have different areas at other points and would accordingly suffer a different type of distortion as it propagates inside the medium. The average dipole moments generated and the gradient force acting on them will vary not only with Z but also with x. We have therefore to obtain G, $\boldsymbol{R}$, $\boldsymbol{S}$ and $\partial G/\partial x$ as functions of x, Z and τ. The effectiveness of the deflection scheme depends on the location (x, Z) of the atom in the ribbon. Some sample results on the impulse p imparted to an average atom per pulse are displayed in figure 3. These results are of a preliminary nature because the computations were carried out only over the duration of the

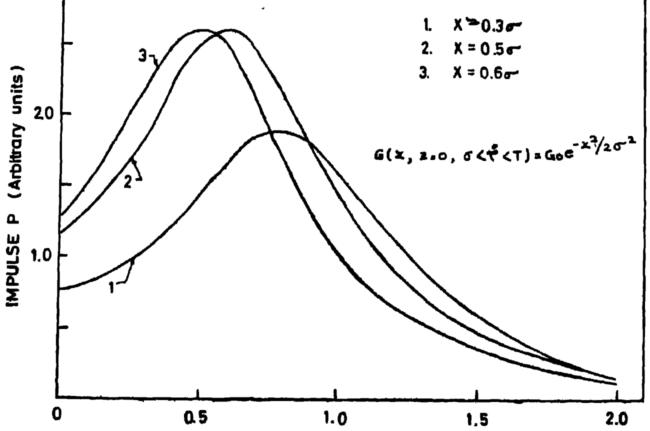

Figure 3. Impulse p imparted to an average atom in the beam is shown as a function of the location (x, Z) of the atom in the interaction region.

applied pulse. Clearly, as shown in Figure 2, as the pulse propagates deep inside the medium, a considerable area of the pulse remains over a much longer duration of time. If the contribution from this area were taken into account, the rate of fall of the impulse p with Z shown in figure 3 would be much less. This trend would be desirable from the point of view of isotope separation.

It may be mentioned finally, that there are, in fact, a large number of parameters in the problem to be varied for arriving at the optimum conditions for isotope separation. Such studies are in progress and the results will be reported later.

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