Coherent interaction of light pulses and a three-level system

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We consider the properties of the coherent interaction between resonance radiation and a three-level medium. We find stationary solutions corresponding to the propagation of an undamped pulse, resonating with the transition between the lowest level and the upper or middle level in the presence of stationary radiation resonating with the transition between the upper two levels. It turns out that the irradiation of the upper transition by relatively weak light can lead to an appreciable delay of the pulse resonating with the lower forbidden transition at a pulse intensity well below the intensity of the usual 2π pulse. We suggest the use of this phenomenon to measure dipole moments and relaxation times of forbidden transitions. Moreover, it turns out to be possible to control short light pulses of one frequency by using continuous radiation or long light pulses of a different frequency.

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The coherent interaction of light pulses with a two-level system leads to a number of interesting effects such as, for instance, self-induced transparency (SIT), photon echo, and so on. Many theoretical and experimental studies (see, e.g.,[1]) have been devoted to this problem. In particular, a detailed study has been made of the properties of the propagation of undamped 2π pulses in media which absorb in resonance, which were first discovered by McCall and Hahn.[2] At the moment the most wide-spread application of coherent effects is the measurement of the dipole moment matrix elements of the corresponding transitions and also of the transverse relaxation times. Both in the measurements using SIT and in the photon echo set-up one needs fields $E \sim \hbar/\mu t_p > \hbar/\mu T_2$, i.e., pulse intensities $I > c(\hbar/\mu T_2)^2$, where μ is the dipole moment of the transition and T_2 the transverse relaxation time. As the threshold intensity $\propto \mu^{-2}$, one needs high intensity laser pulses to study forbidden transitions. When the light intensity is high other non-linear effects may become important which will complicate the measurement.

The estimate given above for the threshold intensity of coherent effects in two-level systems is connected with the fact that the amplitude of the lower level when it interacts, for instance, with a 2π pulse, decreases initially and only starts to increase after the transition speed vanishes. This is possible in a two-level system only when the lower level is completely emptied.

We show in the present paper that the presence of a third level which is coupled in resonance through stationary radiation with the two levels considered changes the situation considerably. The transition speed can vanish (after which the system can return to the initial state) if one takes into account the vanishing of the amplitude of the total level for both resonance fields. The amplitude of the given level increases if we take into account the transition involving the lower level under the influence of the resonance pulse and decreases if we take into account the transition to the third level under the action of the stationary radiation. If the dipole moments of the transitions are very different in magnitude, the return of the system to the initial state may start at a small level of emptying (small with respect to the parameter of the ratio of the dipole moments) of the lower level. This means that the intensity of the undamped pulse may be lowered as compared to the usual 2π pulse by many orders of magnitude, if the dipole moments are of very different orders of magnitude.

We consider the problem of the stationary propagation of light pulses which are in resonance with two cascade transitions in a three-level system. We assume that the inhomogeneous broadening of the two absorption lines is much smaller than the spectral width $1/t_p$ (t_p is the length of the pulse) while t_p, in turn, is much shorter than the transverse relaxation time which we put equal to infinity. Depending on the magnitude of $\Delta\omega = \omega_1 - \omega_2$ ($\omega_{1,2}$ are the frequencies of the cascade resonance transitions) as compared to the spectral width of the pulse two different situations are possible.

1. We consider first the case $t_p\Delta\omega \gg 1$. The equation of motion for the density matrix for the three-level system has a very complicated form, but as we neglect relaxation we can use directly the equations for the amplitudes of the occupation probabilities for the three levels which are in resonance. The contribution from the remaining levels is not in resonance and can be taken into account, using perturbation theory which leads to a dispersion of the phase velocity of the light. In the Maxwell equations and the equations for the level amplitudes we can split off the fast motion with frequencies ω_1 and ω_2 and their linear combinations, using the conditions $\omega_{1,2}t_p \gtrsim \Delta\omega t_p \gg 1$. For the slow motions we look for a stationary solution with a constant phase of the electromagnetic field which depends solely on the variable $\xi = t - x/v$, where v is the velocity of propagation of the envelope of the electromagnetic field.

Using the fact that the phase of the field is constant we can take the amplitudes of the three levels and also the envelopes of the two resonance fields to be real. We give the equations for these quantities at once in dimensionless form:

$$\dot{x}_1 = -y_1x_2, \quad \dot{x}_2 = y_1x_1 - y_2x_3,$$
$$\dot{x}_3 = y_2x_2, \quad \dot{y}_1 = x_1x_2, \quad \dot{y}_2 = \lambda^2x_2x_3. \tag{1}$$

Here $x_{1,2,3}$ are the amplitudes of the lower, middle, and upper level, respectively, $y_{1,2}$ the enveloping fields, in resonance, respectively, with the lower transition at frequency ω_1 and the upper one at frequency ω_2:

$$y_{1,2} = \frac{\mu_{1,2}}{2\hbar}E_{1,2}\Omega^{-1}, \quad \lambda = \frac{\mu_2}{\mu_1}\left[\frac{\omega_2(c/vn_1-1)}{\omega_1(c/vn_2-1)}\right]^{1/2},$$

the time is measured in units Ω^{-1}, where $\Omega^2 = 2\pi N\mu_1^2\{\hbar(c/vn_1 - 1)\}^{-1}$, $E_{1,2}$ are the field amplitudes, $\mu_{1,2}$ the dipole moments, and $n_{1,2}$ the non-resonance indexes of refraction for the lower and upper transition, respectively, while N is the particle density.

We can solve the set of Eqs. (1) with the initial conditions as $t \to -\infty$:

$$x_1=1, \quad x_2=x_3=y_1=0, \quad y_2=y_0,$$

by the substitution

$$\tau=\int_{-\infty}^{t} x_2\,dt \text{ when } x_2>0 \text{ and } \tau=\int_{\infty}^{t} x_2\,dt \text{ when } x_2<0.$$

The solutions of Eqs. (1) as functions of τ have the form

$$y_1=\sin\tau, \quad x_3=(y_0/\lambda)\,\mathrm{sh}\,\lambda\tau, \quad y_2=y_0\,\mathrm{ch}\,\lambda\tau$$
$$x_2=\pm[\sin^2\tau-(y_0/\lambda)^2\,\mathrm{sh}^2\,\lambda\tau]^{1/2}. \tag{2}$$

The function $\tau(t)$ can be found from the equation

$$d\tau/dt=[\sin^2\tau-(y_0/\lambda)^2\,\mathrm{sh}^2\,\lambda\tau]^{1/2}. \tag{3}$$

It follows from (2) and (3) that the solutions obtained are the same as the well known solution for 2π pulses[1,2] when $y_0 = 0$, where $x_3 = y_2 = 0$, i.e., where the upper level does not take part in the interaction between the light and the medium. When $y_0 \neq 0$ there is in the medium a stationary field, in resonance with the upper transition and not being absorbed because in the initial state the middle level is not populated.

It is clear from Eq. (2) for the field y_2 that as the pulse y_1 passes through an element of the medium the "guiding" field increases, and later decreases, although apparently the field y_2 should decrease when the middle level gets populated because of absorption. The fact is that the region of interaction of the pulse y_1 with the medium (which propagates with a velocity $v < c_{1,2}$) is a barrier for the photons of the field y_2 which propagate in the medium with a velocity c_2 outside the interaction region. In the stationary frame the density of the photons increases in the interaction region because of the conservation of their flux. If $y_0 \ll e^{-\lambda}$, the pulse $E_1(t)$ has two humps and consists of two approximate 2π pulses, which are farther apart the smaller y_0. If $e^{-\lambda} \ll y_0 \ll 1$, the pulse E_1 has a single hump with the same exponential tails, but its area is

$$\Theta=\frac{\mu_1}{\hbar}\int_{-\infty}^{\infty}E_1\,dt\approx\frac{4}{\lambda}\ln\frac{1}{y_0}\ll 2\pi.$$

One can qualitatively explain the diminishing of the area of the pulse as follows. When the usual 2π pulse propagates in a two-level medium each particle is lifted to the upper level and afterwards is dropped down, while the re-emission of the absorbed light only starts after the lower level is completely emptied. In the present case the speed of the transitions from the middle to the upper level is much higher than the speed at which the lower level is emptied (when $\mu_2 \gg \mu_1$). The re-emission starts therefore even when the lower level is as yet emptied weakly. Initially the re-emission (the center of the stationary pulse) corresponds to the vanishing of the amplitude of the middle level. It follows from (3) that the stationary propagation of the pulse E_1 is possible only when $y_0 < 1$. We note that $y_0 \sim 1$ corresponds to $E_2^2 \sim N\hbar\omega_2/\lambda^2$. When $\lambda \sim 1$ this means that on average there occurs one photon of the guiding field for each particle. It is clear that the stationary picture cannot be established when the radiation density is larger. When $\lambda \gg 1$ each photon of the guiding field is able to operate many times so that the maximum field y_2 decreases. As $y_0 \to 1$ the pulse has one hump and is stretched by a factor $(1 - y_0)^{-1/2}$, while its area $\Theta = 2\pi\sqrt{3/(1+\lambda^2)}$ can be both larger and smaller than 2π, depending on the magnitude of λ, while

$$E_1=(2\hbar/\mu)\,[6(1-y_0)/(1+\lambda^2)]^{1/2}\Omega/\mathrm{ch}[2^{1/2}(1-y_0)^{1/2}\Omega t].$$

One can easily obtain the analogous solution for the case where the upper, third level is common to the two resonance transitions. As before, a stationary guiding field y_0 for the upper $2 \to 3$ transition is necessary for the existence of a non-trivial response, different from a 2π pulse for the $1 \to 3$ transition. The responses are practically the same as the previous ones in the case $\lambda \ll 1$ (for any $y_0 < 1$). There is some difference in the case $\lambda \gg 1$, $y_0 \ll 1$, as in contrast to (2) now $x_3 = (y_0/\lambda)\sin\lambda\tau$, $y_2 = y_0\cos\lambda\tau$. This leads to a small correction to the enveloping pulse y_1, which oscillates with a frequency $\sim\lambda/t_p$. The spectrum of such a pulse has two scales: the main energy is concentrated in a frequency range $\sim t_p^{-1}$, while weak tails extend λ times further. In the particular case $\lambda = 1$ the pulse y_1 has the form of a normal 2π pulse, extended by a factor $(1 - y_0)^{-1/2}$ and with an area which is larger by the same factor.

2. We now consider the case $t_p\Delta\omega \ll 1$ when we can neglect the difference between the transition frequencies. The field y interacts in resonance simultaneously with both frequencies. The equations for the slow quantities have the form

$$\dot{x}_1=-yx_2, \quad \dot{x}_2=y(x_1+\lambda x_3), \quad \dot{x}_3=-\lambda yx_2, \quad \dot{y}=x_1x_2-\lambda x_2x_3, \tag{4}$$

and the initial conditions as $t \to -\infty$ are $x_1 = 1$, $x_2 = x_3 = y = 0$. Equations (4) have a solution which can be obtained through the change of variable

$$\tau=\int_{-\infty}^{t} y\,dt:$$
$$x_1=\frac{1}{p^2}(\cos p\tau+\lambda^2), \qquad x_2=\frac{1}{p}\sin p\tau,$$
$$x_3=\frac{\lambda}{p^2}(\cos p\tau-1), \tag{5}$$
$$y=\frac{2}{p^2}\sin\frac{p\tau}{2}\sqrt{2\lambda^2+(1-\lambda^2)\cos^2\left(\frac{p\tau}{2}\right)}, \quad p=\sqrt{1+\lambda^2},$$

while $\tau(t)$ is found from the equation $\dot{\tau} = y(\tau)$. Analysis of (5) shows that for any λ the pulse has a single hump and has the same form as the usual 2π pulse for $\lambda = 0$ and $\lambda = 1$, while for $\lambda \gg 1$

$$E=\frac{4\sqrt{2}\,\hbar\Omega}{\mu_1\lambda}\,\frac{\mathrm{ch}\,\Omega t}{\mathrm{ch}\,2\Omega t}. \tag{6}$$

The area of the pulse is equal to $\Theta = 4\pi\sqrt{(1+\lambda^2)}$.

The velocity of propagation of the pulse in the cases considered above is connected with the length of the pulse through the following relation: $t_p = \Omega^{-1}/\sqrt{(1-y_0^2)}$, i.e.,

$$\frac{c}{vn_1}-1=\frac{2\pi N\mu_1^2\omega_1 t_p^2}{\hbar}(1-y_0^2). \tag{7}$$

3. We turn to a discussion of the results. The most popular region of applications of coherent effects at the present moment is the measurement of the matrix elements of appropriate transitions and also of the relaxation times of levels. The problems considered enable us to suggest new variants of such measurements. In particular, if the lower transition is forbidden, we have $\mu_1 \ll \mu_2$ and $\lambda \gg 1$. Illuminating the upper transition with relatively weak light $E_2 \ll \hbar\Omega/\mu_2$ ($y_0 \ll 1$) we can create the conditions for the propagation of an undamped pulse with a field E_1 which is smaller by a factor λ than the field of the usual 2π pulse, i.e., the pulse energy can be smaller by a factor λ^2 for measurements by means of coherent effects.

Of the greatest interest is, in our opinion, the possibility to control short light pulses of one frequency by light of a different frequency. For instance, changing the intensity of the guiding field we can make the medium transparent or opaque for pulses of a well defined magnitude or choose in the same way from a train of ultrashort pulses of different magnitude the maximum one. A sufficiently long pulse can serve as the guiding field. If the front of the long pulse is sufficiently steep, we might by irradiating with it a layer of a resonance medium attempt to arrange a fast jamming for short pulses.

4. A list of possible suggestions for the use of coherent effects in three-level systems can easily be extended and depends, of course, on the experiment. We give some numerical estimates.

The guiding field affects the transmission of the pulse; even when its magnitude $y_0 \ll 1$, i.e., when the light flux $I_2 \ll I_0 = (c/\pi)(\hbar\Omega/\mu_2)^2$. A lower bound for I_2 follows from the requirement that the interaction be coherent $t_p \ll T_2$— the transverse relaxation time of the middle level, $I_2 \gg (c/\pi)(\hbar/\mu_2 T_2)^2$. This estimate is the same as to order of magnitude as the estimate for the SIT threshold[1] and lies within the wide band from 1 W/cm^2 (SF_6, alkali metal vapors) to 100 kW/cm^2 (ruby). When the intensity of the guiding field approaches the upper limit of the above-mentioned estimate, the conditions for the propagation of a short pulse at the lower transition change for any values of the ratios of the dipole moments and the frequencies. If, however, for instance, the lower transition is forbidden and $\mu_1 \sim 10^{-2}\,\mu_2$, the SIT threshold intensity for the lower transition when the guiding field is included is, roughly speaking, decreased by a factor $(\mu_2/\mu_1)^2 \sim 10^4$.

In conclusion we wish to turn our attention to an interesting detail: $\lambda^2 \propto (c/vn_1 - 1)/(c/vn_2 - 1)$. We remind ourselves that increasing λ corresponds to lowering the threshold energy for stationary pulses. If we perform experiments with a gas or a fluid, it could possibly have sense to use the dispersion of the medium when non-resonance transitions are taken into account. In the case $\Omega^2 t_p^2 \ll 1$ we have from (7) $(c/vn_1 - 1) \ll 1$ and it is determined by the time of the pulse, but, choosing a medium with a low dispersion we can make $(c/vn_2 - 1) \ll (c/vn_1 - 1)$ which leads to an increase in λ. This can be done by adding a buffer gas with the required dispersion and choosing its pressure. A similar effect can be achieved in a fluid or gas by changing their composition.

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