Resonance scattering of a short laser pulse on a two-level system: Time-dependent approach

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We discuss the time-dependent power spectrum of fluorescence light produced by a two-level system driven by a smooth, short, resonant laser pulse. We show how the multipeak structure of the spectrum develops in time. The possibility of a smooth transition to the conventional Mollow spectrum is discussed.

In our recent Letter,[1] we have reported a new phenomenon in the resonant interaction of a strong, coherent laser light with the atom. Novel spectral properties were found in the fluorescence induced by a short pulse driving a two-level system.

In the well-studied case of cw excitation,[2] the power spectrum of the scattered light consists of three peaks: the central one at the frequency of the incoming laser light and the two satellites displaced by an amount proportional to the atomic transition dipole moment and to the amplitude of the laser light, known as the Rabi frequency.[3]

In the Letter,[1] using the analytically soluble model of the hyperbolic secant pulse, we showed that the energy spectrum of the scattered light is multipeaked and the number of peaks increases linearly with the area of the pulse. The analytic results of Ref. 1 were obtained under the assumption that the Fourier spectral width of the pulse Γ_p is much larger than the spontaneous width of the transition Γ_s. In this case, the impact of radiative losses on the motion of the atom may be neglected.

The distortion of this perfect situation caused by the finite value of Γ_s has been studied by Lewenstein *et al.*[4] They report that the multipeak structure of the spectrum gets smeared with increasing Γ_s but remains very visible for $\Gamma_s \sim 0.1\Gamma_p$.

The phenomenon of multipeak structure in the spectrum is still lacking a simple, heuristic explanation. In Ref. 4 it is argued that the structure is due to the interference of contributions emitted by the atom at different times during the interaction. Obviously, the energy spectrum, which measures the total number of photons of given frequency radiated regardless of the emission time, allows for maximum interference between the contributions produced at different moments.

To shed some light on this problem, it is instructive to study a more conventional notion of the power spectrum, i.e., the rate at which the photons of given frequency are emitted. In the case of pulse excitation, it is a time-dependent quantity. In fact, the whole process is transient in nature.

When discussing a time-dependent power spectrum, one runs into difficulty with the uncertainty relation between time and frequency. This difficulty can be satisfactorily overcome by taking into account a filter necessary to resolve the spectrum.[5] Such a filter is characterized by its width Γ_F, which determines the resolution of the spectrometer and at the same time its reciprocal $1/\Gamma_F$ is the accumulation time of light in the spectrometer. When we have the resolution Γ_F, we cannot then know the emission time with an accuracy better than $1/\Gamma_F$.

In this paper, as in Ref. 1, for simplicity we will neglect the back action of the emitted radiation on the motion of the atomic dipole. Therefore, if a carrier frequency of the laser $-\omega_L$ is equal to the separation of the atomic-energy levels, within the rotating-wave approximation[4] we can write explicit formulas for the time dependence of probability amplitudes $\alpha(t)$ and $\beta(t)$ of finding the atom in its lower and upper states, respectively, for an arbitrarily shaped pulse.

Splitting the time-dependent, slowly-varying envelope of the field $\epsilon(t)$ into its typical field strength ϵ_0 and the dimensionless shape of the pulse $f(t)$

$$\epsilon(t)=\epsilon_0 f(t) , \tag{1}$$

we can then write the Schrödinger amplitudes $\alpha(t)$ and $\beta(t)$

$$\alpha(t)=\cos[z(t)/2] , \tag{2}$$

$$\beta(t)=\sin[z(t)/2] , \tag{3}$$

where the time-dependent area of the pulse is defined as

$$z(t)=\Omega_0 \int_{-\infty}^{t} f(\tau)d\tau , \tag{4}$$

and the Rabi frequency Ω_0 is proportional to the transition's dipole moment and the typical (usually maximal) field amplitude ϵ_0.

Solutions (2) and (3) describe a two-level atom which was in its lower state before the pulse arrived $[\alpha(-\infty)=1,\beta(-\infty)=0]$. It evolves to its lower state in the remote future if the total area of the pulse is $2\pi n$. The time-dependent spectrum of a light signal described by the electric field operator $\hat{E}(t)$, which is a sum of its positive $\hat{E}^{(+)}(t)$ and negative $\hat{E}^{(-)}(t)$ frequency parts, according to Ref. 5, reads

$$S(t,\omega,\Gamma_F)=2\Gamma_F\int_{-\infty}^{t}dt_1\int_{-\infty}^{t}dt_2\,e^{-(\Gamma_F-i\omega)(t-t_1)}e^{-(\Gamma_F+i\omega)(t-t_2)}\langle\hat{E}^{(-)}(t_1)\hat{E}^{(+)}(t_2)\rangle\ . \tag{5}$$

The exponential memory function describing the ideal Fabry-Perot interferometer was used in (5). The simplified relation, neglecting retardation and three-dimensional propagation properties of the vector field is usually used to connect the emitted field directly with the dipole moment operator[6]

$$\hat{E}^{(-)}(t)=Ce^{i\omega_L t}\hat{\sigma}^{+}(t)\ . \tag{6}$$

Omitting the irrelevant for our purpose proportionality factor C, we get

$$S(t,\omega,\Gamma_F)=2\Gamma_F\int_{-\infty}^{t}dt_1\int_{-\infty}^{t}dt_2\,e^{-[\Gamma_F-i(\omega-\omega_L)](t-t_1)}e^{-[\Gamma_F+i(\omega-\omega_L)](t-t_2)}\langle\hat{\sigma}^{+}(t_1)\hat{\sigma}^{-}(t_2)\rangle\ . \tag{7}$$

The second-order correlation function of dipole-moment operators can be expressed (compare Ref. 1) by the Schrödinger amplitudes $\alpha(t)$ and $\beta(t)$, yielding the spectrum in the form of a sum of two terms

$$S(t,\omega,\Gamma_F)=S_s+S_q\ , \tag{8}$$

where

$$S_s(t,\omega,\Gamma_F)=2\Gamma_F\left|\int_{-\infty}^{t}dt_1\,e^{-[\Gamma_F-i(\omega-\omega_L)](t-t_1)}\alpha(t_1)\beta^{*}(t_1)\right|^2 \tag{9}$$

and

$$S_q(t,\omega,\Gamma_F)=2\Gamma_F\left|\int_{-\infty}^{t}dt_1\,e^{-[\Gamma_F-i(\omega-\omega_L)](t-t_1)}|\beta(t_1)|^2\right|^2\ . \tag{10}$$

We argued in Ref. 1 that the two terms should be designated the "semiclassical part" and "quantum part," respectively. In the context of cw excitations they are usually called coherent and incoherent parts. While the semiclassical part is driven by the motion of the atomic dipole moment, the quantum part is proportional to the square of the population of the upper level.

With the explicit solutions (2) and (3), all one has to do to obtain a time-dependent spectrum of the resonance fluorescence induced by the pulse of the envelope $f(t)$ is to compute the area (4) and the Fourier transforms (9) and (10). This can be done for almost arbitrary pulse shapes on a microcomputer (all our figures were obtained this way) or for some specific pulse shapes analytically.

We quote here the celebrated hyperbolic secant pulse case:

$$f(t)=\operatorname{sech}(\Gamma_p t)\ . \tag{11}$$

Here

$$\alpha(t)={}_2F_1(-n,n,\tfrac{1}{2};x) \tag{12}$$

and

$$\beta(t)=2n\sqrt{x(1-x)}\,{}_2F_1(-n+1,n+1,\tfrac{3}{2};x)\ , \tag{13}$$

where

$$x=\frac{\tanh(\Gamma_p t)+1}{2},\quad n=\Omega_0/2\Gamma_p$$

and ${}_2F_1(\ldots,\ldots,\ldots;x)$ is a hypergeometric function.

For n integer (the pulse has an area equal to $2\pi n$) one can directly multiply the finite sums (12) and (13) and then perform the Fourier integrals term by term to obtain

$$S_s=\frac{2n^2\Gamma_F}{\Gamma_p^2}x(1-x)\left|\sum_{k=0}^{2n-1}C_k^n\frac{1}{\xi+k+\frac{1}{2}}\,{}_2F_1(k+1,1,\xi+k+\tfrac{3}{2};x)\right|^2 \tag{14}$$

and

$$S_q=\frac{8n^4\Gamma_F}{\Gamma_p^2}x^2(1-x)^2\left|\sum_{k=0}^{2(n-1)}D_k^n\frac{1}{\xi+k+1}\,{}_2F_1(k+2,1,\xi+k+2;x)\right|^2\ , \tag{15}$$

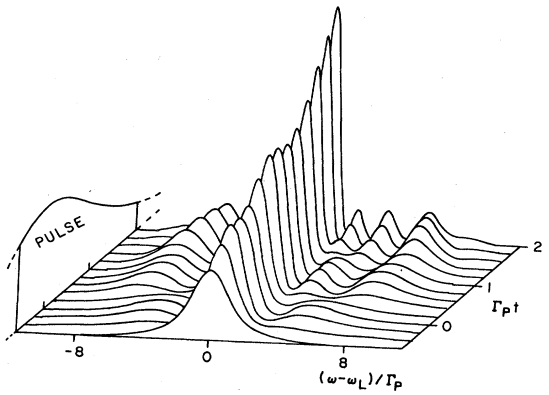

FIG. 1. Physical spectrum of light scattered from a two-level system driven by 8π hyperbolic secant pulse. The seven-peak structure develops in stages as the area of the pulse grows. The width of the filter Γ_F is only $0.1\Gamma_p$.

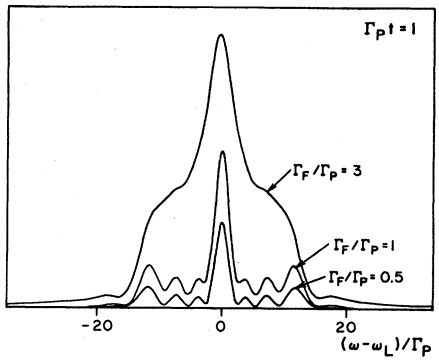

FIG. 3. Physical spectrum of light scattered from a two-level system driven by 8π Gaussian pulse. Curves correspond to filter widths Γ_F of 0.5,1,3. Complete smearing is obtained for large Γ_F instead of the three-peak spectrum.

where

$$\xi=\Gamma_F-i(\omega-\omega_L)$$

and the coefficients C_k^n and D_k^n are given by

$$C_k^n=\sum_{l=0}^{k}\frac{(-n)_l(n)_l(-n+1)_{k-l}(n+1)_{k-l}}{l!(\frac{3}{2})_l(k-l)!(\frac{1}{2})_{k-l}}, \tag{16}$$

$$D_k^n=\sum_{l=0}^{k}\frac{(-n)_l(n)_l(-n)_{k-l}(n)_{k-l}}{l!(\frac{3}{2})_l(k-l)!(\frac{3}{2})_{k-l}}, \tag{17}$$

with the usual Pochhammer notation

$$(\alpha)_n=\alpha(\alpha+1)\cdots(\alpha+n-1).$$

In Fig. 1, the time-dependent spectrum of fluorescence induced by the hyperbolic secant pulse of the area equal to 8π is plotted for a very narrow filter ($\Gamma_F=0.1\Gamma_p$). The spectrum evolves from its initial single peak form developing more and more peaks as the area of the pulse accumulates. For long times, we get a seven-peak spectrum which is nearly identical with the energy spectrum $W(\omega)$ discussed in Ref. 1. In fact, the energy spectrum $W(\omega)$ is related to the long-time limit of the power spectrum with vanishingly narrow filter:

$$W(\omega)=\lim_{t\to\infty}\lim_{\Gamma_F\to 0}\frac{S(t,\omega,\Gamma_F)}{2\Gamma_F}. \tag{18}$$

Obviously, the accumulation time of the field in the interferometer is ten times longer than the duration of the pulse and the interference responsible for the multipeak structure is nearly maximal in this case.

The effect of decreasing the accumulation time is illustrated in Fig. 2. With all the other parameters as before,

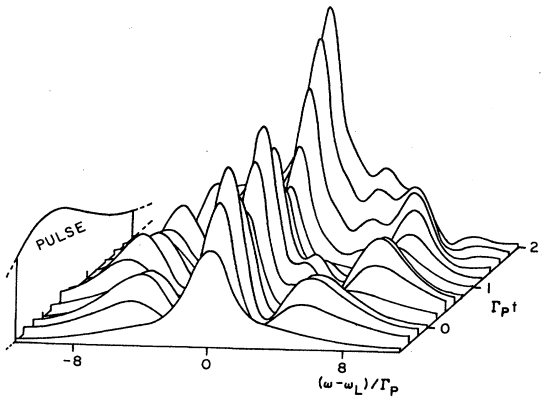

FIG. 2. Same as Fig. 1, but for $\Gamma_F=\Gamma_p$. Shorter accumulation time enhances satellites corresponding to the instantaneous Rabi frequency.

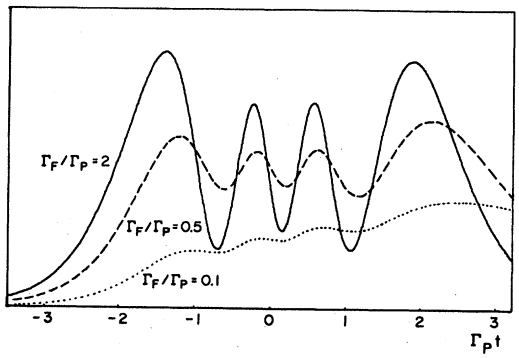

FIG. 4. Total intensity of light leaving the filter is plotted versus time (8π hyperbolic secant pulse) for three ratios of Γ_F/Γ_p. Rabi oscillations are very strong for $\Gamma_F/\Gamma_p=2$ and accumulation of light in the filter leads to the monotonically rising curve for $\Gamma_F/\Gamma_p=0.1$.

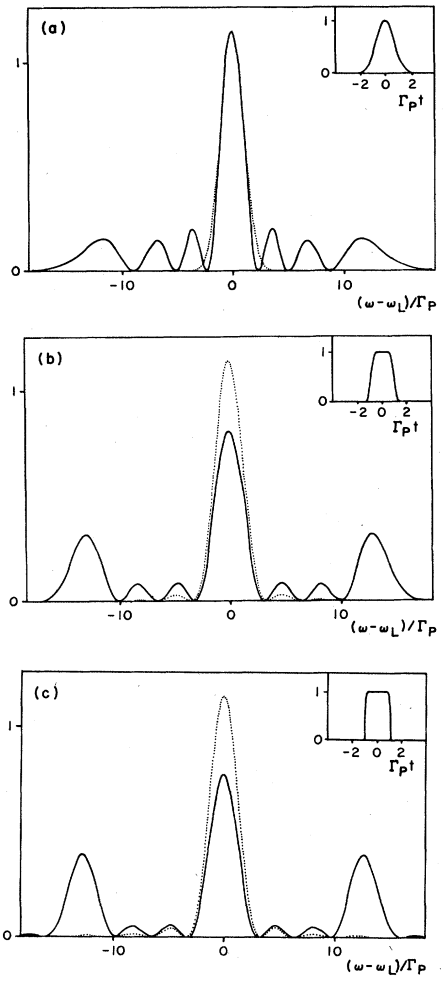

FIG. 5. Energy spectra (or long time, very small filter width power spectra) are plotted for the 8π exponential pulses described by formula (20) for $\alpha=2$, 6, and 26 in (a), (b), and (c), respectively. The time shape of each pulse is shown in the inset. Their Fourier spectra are shown as dotted lines. Note the appearance of the strong Mollow satellites. Note also the superposition of the structure due to the elastic scattering and the saturation phenomena.

now the width of the filter $\Gamma_F=\Gamma_p$. We see the enhancement of the satellite which corresponds to the local, instantaneous Rabi frequency. We get closer to a Mollow-like spectrum. The obvious prescription: take Γ_F much bigger than Γ_p to reproduce the Mollow spectrum; it does not work, however. It is true that the accumulation time is very short in this case, but we lose the spectral resolution. The result is a complete smearing of the structure.

This last point is illustrated in Fig. 3. Here we have the spectra for the 8π Gaussian pulse and increasing width of the filter. As a kind of crosscheck, Fig. 4 shows the total intensity of the scattered light as it leaves the interferometer. We have plotted here the integral

$$I(t,\Gamma_F)=\int S(t,\omega,\Gamma_F)d\omega \tag{19}$$

as a function of time for several values of Γ_F/Γ_p. This intensity oscillates strongly, reflecting the Rabi flopping of the atom if Γ_F/Γ_p is large; it becomes monotonic for $\Gamma_F/\Gamma_p \ll 1$ because of the effective accumulation of the field in the interferometer in this case.

Figures 5(a)—5(c) show in yet another way the smooth transition from our multipeak spectrum to the conventional Mollow-like spectrum[7]. We took the 8π pulses with the envelope

$$f_\alpha(\tau)=e^{-(\Gamma_p t)^\alpha} \tag{20}$$

for α equal to 2, 6, and 26. The pulse in the t domain (see the insets) changes shape from a Gaussian, through one with a well-defined plateau, to a rectangular one.

The energy spectra of the resonance fluorescence change from the pure multipeak to one with well-defined Mollow-type satellites. Their displacement corresponds, of course, to the Rabi frequency at the maximum of the pulse, while their widths are still determined by the width of the pulse. The dotted line represents the Fourier spectrum of the pulse itself. Of course, the Fourier spectrum is single peaked only for the Gaussian pulse. Only in this case do we have pure saturation-type and/or coherence-type splitting. For $\alpha>2$, the Fourier spectrum of the pulse itself is multipeaked and the additional structure in the fluorescence spectrum is partially due to the elastic scattering of the photons from those peaks. In other words, for $\alpha>2$ (or for the rectangular pulse), even the resonance fluorescence spectrum of the harmonic oscillator would be multipeaked.

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