Non-Markovian Strong-Field Excitation of Optical Coherent Transients

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A transient, time-delayed, four-wave mixing experiment is performed in strontium vapor under stochastic excitation conditions. The first two broad-bandwidth laser pulses are correlated and their delay is adjustable. Thus they combine into a non-Markovian excitation process with adjustable memory time. One of the correlated pulses is allowed to be strong. In contrast with previous investigations, it is shown both theoretically and experimentally that under strong-field conditions the atoms are not able to keep the excitation memory over time intervals longer than the inverse laser bandwidth.

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For years, a large amount of work has been devoted to fluorescence induced by resonant interaction with a broad-bandwidth, intense electromagnetic field.[1] The analytical description of this interaction illustrates the linear stochastic differential equation problem. Two important parameters in this problem are the correlation time of the stochastic field τ_m, and the characteristic time of evolution of the driven system T. As long as $\tau_m < T$, averaged solutions are obtained without further specification of the stochastic process. Since T is a decreasing function of the driving field intensity, one ultimately enters the long correlation time domain $\tau_m > T$ on increasing the field strength. Then the problem is solved only for some specific driving processes. Much attention has been paid for years to the special case of Markov processes, which enables us to substitute a master equation for the initial stochastic differential equation.[2] Many efforts have been devoted to the more specific case where excitation is achieved by a randomly time-varying jumplike Markov process.[1] In contrast, we consider a non-Markov excitation process which is provided by a sequence of time-delayed, correlated, broadband, light pulses. This excitation scheme is used in a transient time-delayed four-wave mixing (TDFWM) configuration which is of practical interest to investigate ultrafast phenomena with a temporal resolution as short as the coherence time of the broad-band pulses.[3-7] The strong-field situation in similar configurations was considered in recent experiments[8,9] but the observed features were not conclusively interpreted. Those experiments raise the following question: How long is an atom which interacts with a given field at a given time able to keep memory of this interaction and to recognize the same instantaneous field at a later time? We answer this question both experimentally and theoretically in a specific situation.

In this experiment, strontium vapor is resonantly excited on the transition $5s^2\,{}^1S_0$-$5s5p\,{}^3P_1$ at $\lambda = 689$ nm by a two-pulse sequence. The upper-level decay rate to the ground level is $4.8\times10^4\ s^{-1}$. The cell is heated at 420 °C, which corresponds to a vapor pressure of 2.7×10^{-4} Torr. A small angle is made between the beam wave vectors $\mathbf{k}_1$ and $\mathbf{k}_2$. The two pulses are beam split from a single pulse of duration $\tau_L \sim 10$ ns which issues from a neodymium-doped yttrium-aluminum-garnet laser-pumped broad-band dye laser. This way, two correlated pulses are prepared. The angled beams build a Bragg grating inside the atomic level population. The Bragg vector is $\mathbf{K} = \mathbf{k}_2 = \mathbf{k}_1$. The two pulses time overlap inside the interaction volume. They synchronously excite the atomic system, save for a small adjustable shift $t_{12} \ll \tau_L$. The sign of t_{12} is defined in such a way that t_{12} is positive when the pulse along $\mathbf{k}_1$ reaches the sample before the pulse along $\mathbf{k}_2$, 60 ns after the first two pulses, a non-time-overlapping weak probe pulse, which propagates along $\mathbf{k}_2$, is scattered in direction $\mathbf{k}_1$ by the grating. The scattered intensity is measured as a function of t_{12}.

The coupling of a stochastic field $\mathbf{E}(t)\cos[\omega t - \mathbf{k}\cdot\mathbf{r} + \varphi(t)]$ with the two-level atoms is characterized by the Rabi frequency $\Omega(t) = \mathbf{E}(t)e^{i\varphi(t)}\mu h^{-1}$, where μ is the dipole moment of the transition. The autocorrelation function of the field,

$$g(\tau) = \langle \Omega(t)\Omega^*(t+\tau)\rangle / \langle |\Omega(t)|^2 \rangle , \qquad (1)$$

is directly determined in a side experiment.[10] For that purpose the laser beam is split into two time-separated components with an adjustable delay τ. Both components overlap on a photodiode array detector where they interfere. The Fourier component of the intensity spatial distribution, at the frequency of the interference pattern, corresponds to the fringe contrast. Its variation as a function of τ draws $g(\tau)$ (Fig. 1). The integral $\int_0^\infty g(\tau)\,d\tau$ is the coherence time of the light, which identifies with its inverse spectral width. In Fig 1, $\tau_c = 15$ ps. Parameters of importance are expressed in terms of the effective interaction rate $T^{-1} = \langle |\Omega(t)|^2 \rangle \tau_c$. The ratios τ_L/T and τ_c/T are the effective numbers of interactions, respectively, during the pulse duration and during the light coherence time. In the present TDFWM situation the excitation is achieved by a composite field with long correlation time. This field reads $\chi(\mathbf{r},t) = \Omega_2(t) + \Omega_1(t)e^{i\mathbf{K}\cdot\mathbf{r}}$. It combines

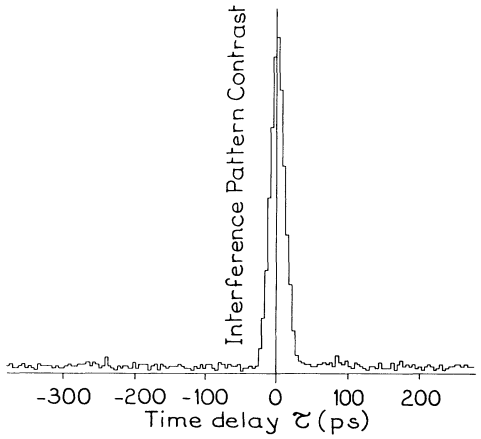

FIG. 1. Autocorrelation function of the light source, recorded as the time-delay variation of the fringe contrast on the interference pattern.

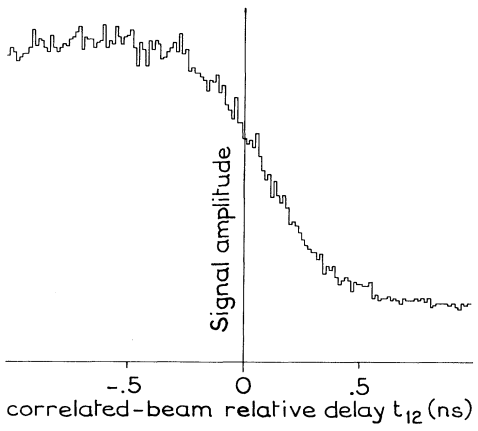

FIG. 2. Experimental TDFWM signal in the weak-field limit. Each point of the curve is averaged over about 200 laser shots.

$\Omega_1(t)$ and $\Omega_2(t)$ which propagate along $\mathbf{k}_1$ and $\mathbf{k}_2$, respectively. The correlation of the two components is expressed by $\Omega_1(t) \propto \Omega_2(t+t_{12})$. The composite field value at time t is thus correlated to its value at time $t \pm t_{12}$, where the geometrical delay t_{12} can be made much larger than the coherence time of the light source. The relevant correlation time τ_m for this composite field is whichever one of τ_c and τ_{12} is larger. The weak-field condition $\tau_L/T \ll 1$ is fulfilled by $\Omega_1(t)$.

The signal intensity as a function of t_{12} has been recorded for different values of $\Omega_2(t)$ which range from the weak-field regime (Fig. 2) to the strong-field situation (Fig. 3, where $T=0.1$ ns) within the limits of the requirement $\tau_c/T \ll 1$. The weak-field profile has been discussed previously.[7] It reflects the atomic dipole capability for long-time storage of excitation memory. Interacting with the same instantaneous field at two different times, atoms are able to recognize the correlation between those two interactions even if their time separation exceeds τ_c. Thus atoms detect long-time correlation (i.e., over intervals larger than τ_c) inside the composite field. The weak-field profile corresponds to the switch over of the signal emission from direction $\mathbf{k}_1$ to direction $2\mathbf{k}_2 - \mathbf{k}_1$ when t_{12} is varied across zero. This switchover occurs on the time scale of the inverse Doppler width Δ_D^{-1}. A drastic change occurs in the profile when $\Omega_2(t)$ is increased. The strong-field profile exhibits a sharp dip at $t_{12}=0$. Its width is close to that of $g(\tau)$ in Fig. 1. By varying the spectral width of the source we have verified that the width of the dip is proportional to τ_c. Out of the dip region, the profile is as flat as it would be if the excitation pulses were not correlated. Indeed, with uncorrelated pulses, the time scale of variation of the signal intensity as a function of t_{12} is $\tau_L \sim 10$ ns. This suggests that the memory time is reduced to τ_c in the strong-field regime. Since $\Delta_D \tau_c \ll 1$, the Doppler phase has not had enough time to build up and the atoms behave as motionless radiators. The following calculation confirms this picture.

The sample is regarded as an ensemble of motionless atoms. The atomic levels are labeled a and b. Atoms are described by $\sigma_0(\mathbf{r},t)$, which denotes the level-population difference, and by $\sigma_{\pm 1}(\mathbf{r},t)$ which is connected to the nondiagonal density-matrix element $\rho_{ab}(\mathbf{r},t)$ according to

$$\sigma_1(\mathbf{r},t) = \sqrt{2}\rho_{ab}(\mathbf{r},t)e^{i(\omega t - \mathbf{k}_2 \cdot \mathbf{r})}, \qquad \sigma_{-1}(\mathbf{r},t) = \sigma_1^*(\mathbf{r},t). \tag{2}$$

The equation of motion reads

$$\dot{\sigma}(\mathbf{r},t) = \mathbf{m}(\mathbf{r},t)\sigma(\mathbf{r},t), \tag{3}$$

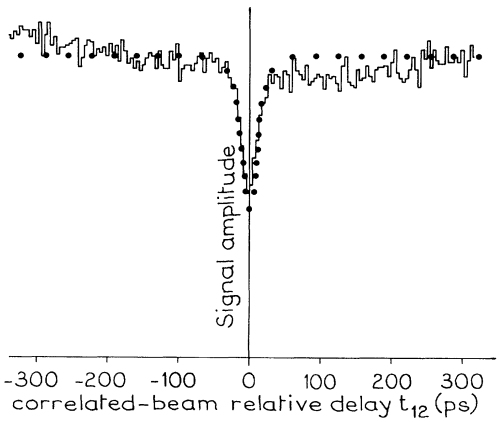

FIG. 3. Full line, experimental TDFWM signal when $[\langle |\Omega_2(t)|^2 \rangle \tau_c]^{-1} \cong 100$ ps. The corresponding autocorrelation function is that of Fig. 1. Dotted line, theoretical profile according to Eq. (12). The recorded autocorrelation function of Fig. 1 has been substituted for $g(\tau)$ in Eq. (12).

where

$$\mathbf{m}(\mathbf{r},t)=\frac{i}{\sqrt{2}}\begin{bmatrix}0 & \chi(\mathbf{r},t) & 0\\ \chi^*(\mathbf{r},t) & 0 & -\chi(\mathbf{r},t)\\ 0 & -\chi^*(\mathbf{r},t) & 0\end{bmatrix} \quad (4)$$

and

$$\boldsymbol{\sigma}(\mathbf{r},t)=\begin{bmatrix}\sigma_1(\mathbf{r},t)\\ \sigma_0(\mathbf{r},t)\\ \sigma_{-1}(\mathbf{r},t)\end{bmatrix}. \quad (5)$$

The signal field results from the combination of the elementary fields radiated by the individual dipoles. The relevant signal intensity is averaged over a large number of realizations. It reads $\langle|\sum\rho_{ab}(n)|^2\rangle$, where the sum runs over the individual atoms and where $\langle\,\rangle$ stands for the statistical average over signal realizations. The dominant contribution to this sum arises from the averaged cross products $\langle\rho_{ab}(n)\rho_{ba}(m)\rangle$. Thus the quantity to be calculated is $\langle\sum(\mathbf{r},\mathbf{r}',t)\rangle=\langle\boldsymbol{\sigma}(\mathbf{r},t)\otimes\boldsymbol{\sigma}(\mathbf{r}',t)\rangle$, where $\otimes$ represents an outer product. From Eq. (3) one obtains[11]

$$\left\langle\sum(\mathbf{r},\mathbf{r}',t)\right\rangle=\int^t dt'\langle\mathbf{M}(\mathbf{r},\mathbf{r}',t)\mathbf{M}(\mathbf{r},\mathbf{r}',t')\sum(\mathbf{r},\mathbf{r}',t')\rangle\,, \quad (6)$$

where $\mathbf{M}(\mathbf{r},\mathbf{r}',t)=\mathbf{m}(\mathbf{r},t)\otimes\mathbf{I}+\mathbf{I}\otimes\mathbf{m}(\mathbf{r}',t)$. A solution to this equation is readily obtained without further characterization of the driving field in either of the three following situations[2]: (i) The weak-field condition $(\tau_L/T\ll1)$ is satisfied by both Ω_1 and Ω_2. (ii) The fields Ω_1 and Ω_2 are *uncorrelated* and they both satisfy the short correlation time condition. The correlation time τ_m then coincides with τ_c and the condition reads $\tau_c/T\ll1$. (iii) The fields Ω_1 and Ω_2 are *correlated* and the short correlation time condition now reads $\tau_c/T\ll1$ *and* $t_{12}/T\ll1$.

In the first case, the quantity $\sum(\mathbf{r},\mathbf{r}',t')$ on the left-hand side of Eq. (6) is replaced by its initial nonfluctuating value. In the other two cases, the variations of the atomic quantities may be decorrelated from the fluctuations of the driving field. Then Eq. (6) reads

$$\left\langle\sum(\mathbf{r},\mathbf{r}',t)\right\rangle=\int^t dt'\langle\mathbf{M}(\mathbf{r},\mathbf{r}',t)\mathbf{M}(\mathbf{r},\mathbf{r}',t')\rangle\left\langle\sum(\mathbf{r},\mathbf{r}',t)\right\rangle. \quad (7)$$

It results that the quantities $Y_i(\mathbf{r},\mathbf{r}',t)=\langle\sigma_i(\mathbf{r},t)\times\sigma_{-i}(\mathbf{r}',t)\rangle$ verify the equation

$$\dot{\mathbf{Y}}(\mathbf{r},\mathbf{r}',t)=-\mathbf{A}(\mathbf{r},\mathbf{r}',t)\mathbf{Y}(\mathbf{r},\mathbf{r}',t)\,, \quad (8)$$

where

$$\mathbf{A}(\mathbf{r},\mathbf{r}',t)=\frac{1}{2}\begin{bmatrix}a & b & 0\\ b & a+a^* & b^*\\ 0 & b & a^*\end{bmatrix} \quad (9)$$

and

$$\begin{aligned}a&=\int_{-\infty}^t dt'\langle\chi^*(\mathbf{r},t)\chi(\mathbf{r},t')+\chi(\mathbf{r}',t)\chi^*(\mathbf{r}',t')\rangle\,,\\ b&=-\int_{-\infty}^t dt'\langle\chi^*(\mathbf{r}',t)\chi(\mathbf{r},t')+\chi(\mathbf{r},t)\chi^*(\mathbf{r}',t')\rangle\,.\end{aligned} \quad (10)$$

The probe field is scattered on the population grating described by $Y_0(\mathbf{r},\mathbf{r}',t)$. To first order in Ω_1, it is an easy matter to extract the term $y_0^{(\mathbf{k}_1)}(\mathbf{r},\mathbf{r}')$ which originates the scattering in direction $\mathbf{k}_1$. When $\Omega_2(t)$ is strong [i.e., when $\langle|\int dt\,\Omega_2(t)|^2\rangle\gg1$], and starting with the initial condition $Y_i(\mathbf{r},\mathbf{r}',0)=\delta_{i0}$, one obtains

$$y_0^{(\mathbf{k}_1)}(\mathbf{r},\mathbf{r}')=\tfrac{2}{9}(\Omega_1^0)^2\tau_L\tau_c e^{i\mathbf{K}\cdot(\mathbf{r}-\mathbf{r}')} \quad (11)$$

in the case when Ω_1 and Ω_2 are *uncorrelated*, and

$$y_0^{(\mathbf{k}_1)}(\mathbf{r},\mathbf{r}')=\tfrac{1}{9}(\Omega_1^0)^2\tau_L\tau_c[2-\beta(2-\beta)]e^{i\mathbf{K}\cdot(\mathbf{r}-\mathbf{r}')} \quad (12)$$

when Ω_1 and Ω_2 are *correlated*, where $\beta=\tau_c^{-1}\times\int_{t_{12}}^{\infty}|g(\tau)d\tau$ and $(\Omega_i^0)^2\tau_L\tau_c=\langle|\int dt\,\Omega_i(t)^2\rangle$. As soon as $t_{12}>\tau_c$, Eq. (12) coincides with Eq. (11). This suggests that strong-field excitation prevents atoms from keeping memory of the field phase and amplitude over a time interval larger than τ_c. Atoms are then no longer able to detect the long-time correlation inside the composite field. The t_{12} dependence of the signal intensity is expected to conform to that of $y_0^{(\mathbf{k}_1)}(\mathbf{r},\mathbf{r}')$ in the region where $t_{12}<T$. This is verified on the experimental profile (Fig. 3) where $T=0.1$ ns.

Even if $\tau_c\ll T$, the short-correlation-time condition is violated when t_{12} is larger than the characteristic time of the atom evolution, T. For that case of long correlation time, no general method exists to solve Eq. (6). Standard techniques require that the driving field is Markovian, while, in the present problem, the composite field $\chi(\mathbf{r},t)$ is a non-Markov process, irrespective of the stochastic properties of Ω_1 and Ω_2. Indeed, let t' be a time such that $t-t_{12}<t'<t$. Then, the conditional probability density at time t, given the value at time t', is not uniquely determined, since it is affected by the knowledge of the value at an earlier time $t-t_{12}$.[2] We have developed a diagrammatic method which has enabled us to calculate $\langle\sum(\mathbf{r},\mathbf{r}',t)\rangle$ for $t_{12}>T$, despite the non-Markov character of the driving field.[12,13] When Ω_2 is strong, and to first order in Ω_1, the resulting expression for $y_0^{(\mathbf{k}_1)}(\mathbf{r},\mathbf{r}')$ confirms that the signal keeps no trace of the correlation between Ω_1 and Ω_2 as soon as $t_{12}>\tau_c$. It confirms that under strong-field conditions the excitation memory is no longer kept by the atoms over time intervals longer than τ_c.

The obliteration of memory over a time scale τ_c in this specific non-Markov intense excitation is the main result of this paper. It should be emphasized that in analog experiments it has been previously suggested that the memory could be kept over a time scale T.[8,9] This conjecture relied on the Bloch picture of atomic motion. In that picture, the atomic system is represented by a vector which keeps the memory of its initial heading over the time scale T.[9] It was then inferred that the memory of correlation between the excitation pulses would be kept as long as $t_{12}<T$, and that the signal induced by corre-

lated pulses would differ from the signal in an uncorrelated-pulse situation as long as $t_{12} < T$. Our present results clearly question this argument. Complementary experiments that we have performed on sodium vapor indicate that the TDFWM signal is very sensitive to the atomic-level structure. The two-level picture conveniently describes the considered transition in strontium. It fails to predict observed features on the sodium D lines, which connect levels with complex Zeeman and hyperfine structure. Finally, the field Ω_2 can be varied experimentally out of the region where $\tau_c \ll T$. Then, additional wings to the sharp dip appear as $\langle | \Omega_2(t) | ^2 \rangle \tau_c^2$ is increased. This feature is being investigated on a theoretical ground, together with the signal formation when both Ω_1 and Ω_2 are strong fields.

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