Observation of the Formation of the 0π Pulse

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We have measured with 0.5-psec resolution the reshaping of small-area 7-psec laser pulses to 0π pulses due to their passage through an optically thick sodium cell. The reshaped pulses have a picosecond oscillatory structure characteristic of 0π pulses that extends over more than 100 psec. These observations are compared in detail with the predictions of theory and good agreement is obtained

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In coherent optics[1] the pulse area θ is defined as the total angle that the atomic state vector rotates around the electric field of the resonant driving pulse. This angle determines the atomic excitation which remains after passage of the pulse. For example, a $\pi/2$ pulse will put the atoms in a coherent superposition of the ground and excited states, a π pulse will put the atoms in the excited state, and a 2π pulse will take the atoms to the excited state and then back to the ground state. Another type of pulse which has generated considerable theoretical interest [2–8] but for which there has been only a few experimental investigations[9–13] is the zero-area (0π) pulse. In its simplest form, the initial part of the 0π pulse excites the atoms and then, as a result of a phase change of π in the electric field amplitude, the latter part of the pulse takes the atoms back to the ground state. Both strong and weak 0π pulses have been previously studied. The strong 0π pulse is made by electro-optically switching the phase by π of the second half of a pulse of arbitrary intensity, but it is difficult to get the 0π pulse area to the desired accuracy. The weak 0π pulse can be obtained to arbitrary accuracy by the automatic reshaping which occurs when a weak pulse is propagated through a resonant vapor. Early experimental work with the weak 0π pulses showed pulse reshaping on the nanosecond time scale,[10,11] and short pulse generation,[12] but no detailed comparison was made with theory. Recent experiments[13] demonstrating a new method of transient spectroscopy for molecular systems showed a large extension of the pulse wings and an oscillatory structure on the picosecond time scale.

In this paper we report the first subpicosecond experimental and theoretical study of weak-pulse propagation in a simple atomic vapor system and the concomitant formation of the 0π pulse. Our results show the strong oscillatory pulse-reshaping effects characteristic of 0π pulse formation. The reshaped envelope extends over many input pulse widths, and has time dependences shorter than the input pulse width and shorter than any relaxation time in the atomic vapor system. A detailed comparison between the observed reshaped pulses and our theoretical calculations gives good agreement. Our study shows that strong reshaping effects can occur when the absorption linewidth is negligible in comparison to the input pulse bandwidth, or equivalently, when the input pulses are much shorter than any system relaxation time. Because our experiments were performed in the linear regime, they have application to a wide range of physical systems, and can be explained by many different theoretical pictures.[1,4,14]

Many of the previous theoretical investigations[5–8] have been concerned with the strong 0π pulses, which must be described by the full coupled Maxwell-Bloch equations.[1] However, for the weak 0π pulses, the Bloch equations reduce to that of a harmonic oscillator and linear dispersion theory can be applied.[4] This fact simplifies the theoretical analysis considerably and eliminates the complications due to the intensity variation across the beam profile. For this case the pulse area decays exponentially with the on-resonance absorption coefficient α_0. This result is a special case of the area theorem of McCall and Hahn,[15]

$$d\theta(z)/dz = -\tfrac{1}{2}\alpha_0 \sin\theta(z), \tag{1}$$

i.e., for $\theta \ll 1$, $\sin\theta \approx \theta$, and then $\theta(z) = \theta(0) \times \exp(-\alpha_0 z/2)$, where z is the propagation distance. The weak-pulse area theorem is valid for any resonant pulse and applies to our experimental situation where we have 7-psec pulses with a spectral bandwidth ~ 20 times broader than the resonance linewidth of the atomic sodium vapor. Clearly, the pulse energy cannot decay exponentially. Instead the pulse area decays because of the reshaping of the pulse by its passage through the Na vapor. The electric field *amplitude* develops an oscillatory structure with periodic phase changes of π. The first oscillation of the field amplitude excites the atoms and then, as a result of a phase change of π, the

second oscillation deexcites them and so on. *This complicated excitation and deexcitation process remarkably leads to the simple exponential decay of the pulse area as the pulse propagates through the vapor.* For this reshaped pulse, the vapor becomes transparent to exponentially increasing accuracy.

The experimental configuration used is shown in Fig. 1. Pulses of 7-psec duration were obtained from a 4-MHz–rate cavity-dumped synchronously pumped dye laser. The laser was tuned to the 5890-Å transition of Na and had a linewidth of ~ 2 cm^{-1}. A 10%-90% beam splitter provided two beams. The strong beam was sent through an optical-fiber pulse compressor[16] to produce a train of probing pulses of width less than 0.5 psec. The weak beam was sent through a compensating delay line and directly into a Na vapor cell. The peak input power of the weak beam was ~ 100 W, and the beam diameter in the cell was 0.3 cm, corresponding to an input pulse area of $\pi/30$ rad. The 50-cm long, 2.5-cm-diam Pyrex glass cell contained an excess of 99.95% pure Na metal transferred under vacuum from a sealed ampoule. The peak absorption α_0 was calculated from the atomic number density corresponding to the measured cell temperature.

The output pulse shapes from the sodium cell were measured with better than 0.5-psec resolution by cross correlation with the compressed pulses by noncollinear generation of second-harmonic light in a 0.3-mm-long potassium dihydrogen phosphate crystal. The second-harmonic light was monitored with a photomultiplier connected directly to a signal averager which was synchronized with the delay setting of the probe pulses. A typical scan of the probe delay took 100 secs and usually ten scans were averaged.

In Fig. 2 we show a series of our measurements (solid curves) of the output pulse shapes for different absorptions $\alpha_0 l$ of the sodium cell. The input pulse, normalized to unity, is given by the top curve with $\alpha_0 l = 0$. The observed pulse reshaping extends over many pulse widths but shows time dependences faster than the 7-psec input pulse and significantly faster than the relaxation times of the Na atoms, $T_1 = 16$ nsec, $T_2 = 32$ nsec, and $T_2^* \cong 100$ psec. As $\alpha_0 l$ is increased by increasing the Na density, the number of oscillations in the output pulse envelope increases, the oscillations come closer together, and the duration of the pulse envelope increases. In accordance with theory, the positions of the zeros in the oscillatory envelopes

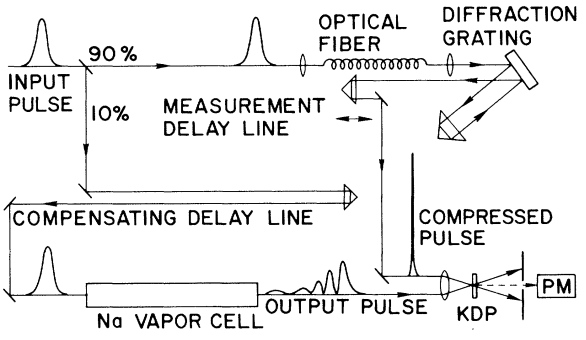

FIG. 1. Experimental arrangement used to measure the reshaped-pulse intensity.

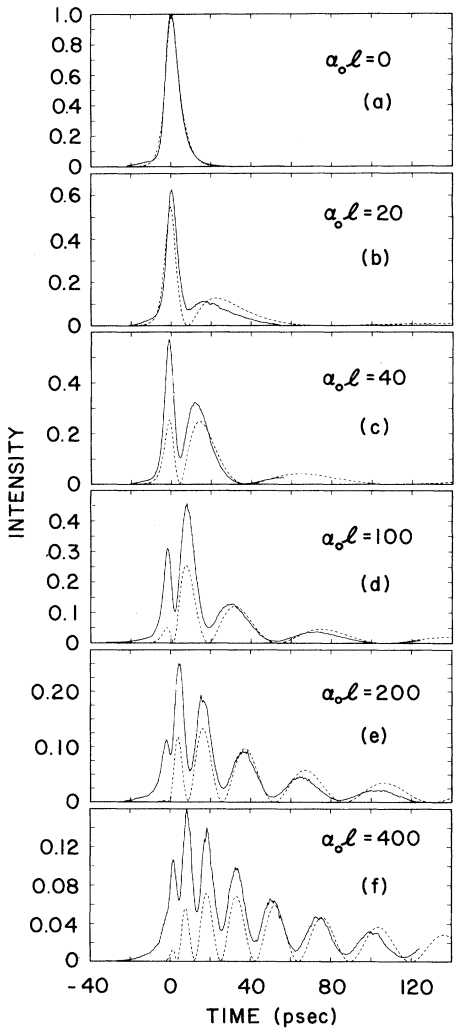

FIG. 2. Observed (solid curves) and theoretical (dashed curves) reshaped-pulse intensity after resonant propagation through sodium vapor of optical density $\alpha_0 l$. Theory is for transform-limited pulses tuned exactly to the Na absorption line and involves no adjustable parameters.

are proportional to a fundamental frequency multiplied by the square root of the time. As can be seen, the total energies of the pulses remain essentially constant as $\alpha_0 l$ is increased, while the intensities decrease because of the increased pulse duration. In fact, consistent with the area theorem, once the absorption profile was saturated ($\alpha_0 l > 5$) the measured small amount of energy absorption showed little change. For $\alpha_0 l = 5$ ($\alpha_0 l = 400$) the pulse energy decreased by only 7% (11%) as a result of passage through the cell. Consequently, the main pulse-reshaping mechanism is the dispersion of the index of refraction n of the Na vapor and not the absorption α.

The numerical calculations (dashed curves) are based on linear dispersion theory following the procedure of Crisp.[4] We used the Voigt profile for the absorption coefficients and the plasma dispersion function for the indices of refraction of the two hyperfine components of the 5890-Å line. Thus, with the electric field of the input pulse given by

$$E(z=0,t) = \mathrm{Re}\{\mathscr{E}(0,t)\exp(-i\omega_0 t)\}, \tag{2}$$

and

$$\tilde{\mathscr{E}}(0,\Omega) = (1/2\pi)\int_{-\infty}^{\infty} e^{i\Omega t}\mathscr{E}(0,t)\,dt, \tag{3}$$

the output field is given by

$$E(z,t) = \mathrm{Re}\{\exp(-i\omega_0 t)\int_{-\infty}^{\infty}\tilde{\mathscr{E}}(0,\Omega)\exp(-i\Omega t)\exp(-\alpha z/2)\exp[inz(\Omega+\omega_0)/c]\,d\Omega\}, \tag{4}$$

where α and n are frequency dependent, and ω_0 is the resonant laser carrier frequency. From these expressions the area theorem is then immediately apparent (μ is the dipole moment of the atomic transition):

$$\theta(z) = (\mu/\hbar)\left|\int_{-\infty}^{\infty}\mathscr{E}(z,t)\,dt\right| = (\mu/\hbar)\left|\tilde{\mathscr{E}}(z,0)\right| = (\mu/\hbar)\left|\tilde{\mathscr{E}}(0,0)\exp(-\alpha_0 z/2)\right| = \theta(0)\exp(-\alpha_0 z/2). \tag{5}$$

In Fig. 2 we compare our experimental results for the pulse envelopes with the theoretical predictions. The transform-limited input pulse used in our calculations was obtained by fitting the experimental input pulse ($\alpha_0 l = 0$) as shown in Fig. 2(a). The data of Fig. 2 show excellent agreement with the theory, considering that the comparison between theory and experiment extends over many input pulse widths and that the calculations involve no adjustable parameters. However, there are two discrepancies. Firstly, the theory predicts much smaller signal intensities at small delays than is actually observed. We believe that this occurs because the bandwidth of the pulses exceeds the transform limit by a factor of ~ 1.5. The resulting excess of spectral energy far from the Na absorption increases the signal intensity at $t = 0$. Secondly, the minima of the oscillations in the data do not go to zero as predicted. We note that our time resolution was more than adequate to resolve these minima and that the linearity of the system was verified by attenuation of the input pulse by a factor of 10. One possible explanation is that there may have been a small pulse-to-pulse frequency jitter. To check these concepts, we introduced a linear chirp and frequency jitter (whose combined effects were consistent with the observed spectrum) in the input pulses used in our calculation. As shown in Fig. 3, this approach gives nearly quantitative agreement.

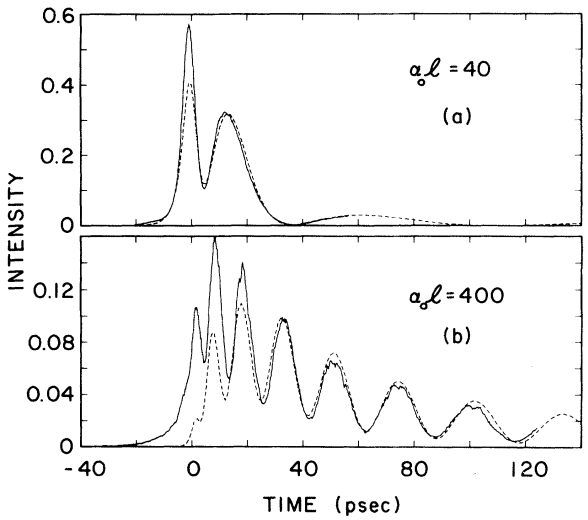

FIG. 3. Reshaped-pulse intensity, experiment (solid lines) and theory (dashed lines). Theory is for linearly chirped pulses with a triangular distribution of center frequencies consistent with the observed pulse spectrum.

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