High Intensity Pulse Propagation in the Extreme Sharp-Line Limit

M. Matusovsky, B. Vaynberg, and M. Rosenbluh

Resnick Institute for Advanced Technology and Department of Physics, Bar-Ilan University, Ramat-Gan 52900, Israel

(Received 19 August 1996; revised manuscript received 7 November 1996)

We observe the propagation and reshaping of intense pulses, with a pulse area greater than π, through a highly absorbing medium. Depending on pulse and absorber parameters, both pulse breakup and undistorted pulse propagation are observed. Using a theoretical description of pulse propagation in the sharp-line limit and a multilevel system possessing unequal transition dipole moments, we demonstrate good agreement with the observations. [S0031-9007(96)01918-7]

PACS numbers: 42.50.Gy

Self-induced transparency (SIT) [1], characterized by changes of the pulse velocity, pulse reshaping, and under special conditions a lossless, soliton, propagation of the pulses through a resonant medium, has been extensively studied. The theoretical description of SIT for a two-level system is well established [2] and has been confirmed by experiments [3,4]. The investigation of SIT in the sharp-line limit (SLL), where the pulse spectrum is much broader than the width of the atomic absorption line, was reported by Gibbs and Slusher [4] (ratio of pulse width to linewidth $\sim$4) for a two-level system possessing a unique dipole moment.

The theoretical description of SIT for a multilevel system, even when the multiple levels are degenerate, is much more complicated, since the various allowed transitions all have different dipole moments. In spite of this, SIT in a degenerate multilevel system has been observed [5,6] in alkali-metal vapors, molecular gases, and solids, and has also been explained theoretically [6,7]. In these experiments the dipole moments of the multiple, degenerate transitions cluster strongly around a particular value, and thus the system could be described as a two-level system with a unique dipole moment with perturbations. These results were obtained, however, for pulses with nanosecond or longer duration, so that the pulse spectrum was narrow compared with the absorption line shape. Here we extend the observation and theoretical description of SIT, to the case of a multilevel system, with many unequal transition dipole moments, interacting with a short, high-intensity laser pulse. In our experiments the extreme SLL is reached where the ratio of pulse width to linewidth is much greater than one. These experiments are thus a generalization of short pulse propagation to multilevel systems, and we show that undistorted pulse propagation occurs in such systems at high enough intensities. Our numerical calculations and experiments show that similar results must be observed for any multilevel absorber.

The experiments were performed in an atomic K vapor cell, with the pulse center frequency in resonance with the D_1 line of K. The power of the laser in our experiments was sufficient to produce pulses with an area exceeding 4π even for the transition with the smallest dipole moment. At high K densities and for the highest optical powers used [8], other nonlinear effects, such as self-focusing and filamentation, are observed to become important and make it difficult to isolate the SIT features of the pulse propagation. Here we report on pulse propagation for parameters that lie below the onset of these nonlinearities.

It has been previously shown [9–11] that, in the SLL, low intensity 0-π pulses are strongly reshaped due to the dispersion of the medium. One might thus expect that for a given optical absorption depth, as the pulse area is increased, dispersion will continue to play a role and result in the reshaping and breakup of the pulse. In this Letter we show, however, that, contrary to this expectation, reshaping occurs only for the low area pulses, and, with increasing pulse area, the pulse propagates undistorted. Calculations indicate that the pulse eventually breaks up only if the optical density, αl, becomes sufficiently large ($>$300).

To interpret the experimental results we numerically solve the Maxwell-Bloch equations as they apply to the D_1 line of K. Transitions from either of the $F = 1,2$ ground states to either of the $F' = 1,2$ excited states are allowed, and, in addition, we consider that each of the F and F' states has a set of degenerate m_F levels. This results in a highly degenerate (or very nearly degenerate) system with many different transition moments. We assume in the calculation a complete energy degeneracy in both the lower and upper states, since the hyperfine energy differences (460 MHz for the $F = 1,2$ states and 50 MHz for the $F' = 1,2$ states) are negligible compared to the spectral width of the pulse (typically 50 GHz).

Considering an input pulse, whose electric field is given by $\mathcal{E}(z,t) = E(z,t)\cos\omega_L t$, at central frequency, ω_L, and assuming the validity of a slowly varying wave approximation, the equation which describes the propagation of the pulse envelope in the extreme SLL can be rewritten to include hyperfine and Zeeman degeneracy. For resonance between ω_L and the atomic transition frequency, ω_0, the

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Maxwell-Bloch equation is

$$\frac{\partial E}{\partial z} = -\frac{\partial E}{c\,\partial t} - \frac{2\pi\omega_L}{(2J+1)(2I+1)c} N \times \sum_{F,F',m_F,m_{F'}} p^q_{F,F',m_F,m_{F'}} \times \sin\left(k^q_{F,F',m_F,m_{F'}} \int_{-\infty}^{t} E(z,t')dt'\right), \qquad (1)$$

with $p^q_{F,F',m_F,m_{F'}} = k^q_{F,F',m_F,m_{F'}}/\hbar$ the dipole moment matrix element for a $\{F, m_F\}$ to $\{F', m_{F'}\}$ transition, q is an index corresponding to the polarization of the incident light ($q = 0$, for linear π polarization and $+1$ and -1 for $\sigma+$ and $\sigma-$ polarizations), J and I are the atomic quantum numbers, and N is the atomic density.

For low-intensity 0-π pulses, the integral in Eq. (1) is much less than 1, and by expanding the sin appearing in the integral and using the methods of Crisp [9], Eq. (1) can be solved in the SLL to obtain a pulse envelope given by

$$E(z,t) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} \varepsilon(\omega,0) \times \exp\left[-i\omega\left(t-\frac{z}{c}\right) + \frac{\alpha_0}{i\omega}\right]d\omega\,, \qquad (2)$$

where the Fourier components of the field are

$$\varepsilon(\omega,z) = \varepsilon(\omega,0)\exp\left[i\left(\frac{\omega z}{c} - \frac{\alpha_0}{\omega}z\right)\right], \qquad (3)$$

and whereby,

$$\alpha_0 = 2\pi\omega_0 N p^2/c\hbar\,,$$

$$p^2 = \frac{1}{(2J+1)(2I+1)} \sum_{F,F',m_F,m_{F'}} p^2_{F,F',m_F,m_{F'}}\,.$$

Since absorption in the SLL is negligible, it is reasonable that we obtain an imaginary exponent in Eq. (3) indicating that pulse reshaping is due to media dispersion and is not a consequence of the absorption.

For high-intensity pulses the small area approximation, $k\int_{-\infty}^{t} E(z,t')\,dt' \ll \pi$ is no longer valid, and Eq. (1) has to be solved numerically. It is clear from Eq. (1) that stable solitonlike solutions, such as those which were found for propagation through a medium with a unique dipole moment, do not exist for multiply degenerate levels, as is the case for the potassium $D1$ line. The numerical evaluation of Eq. (1), for the case of the K $D1$ line with varying pulse area and optical depth, was therefore performed.

In our experiment, a mode-locked Ti-sapphire laser produced nearly transform limited, 6 ps pulses, at a rate of 82 MHz. The central wavelength of the laser was adjusted to be at the K resonance wavelength of 769.9 nm. Up to 1.5 W of average laser power could be propagated through a 1 cm long Pyrex cell containing K vapor and maintained at a variable temperature between 30 °C and 250 °C. (Higher temperatures were avoided due to the initiation of self-focusing and conical emission.) The focused laser beam had a diameter of $\sim$80 μm and a confocal parameter of 11 mm, close to the 10 mm length of the K cell. The pulses before and after passing through the cell were measured with a scanning autocorrelator and simultaneously monitored with a high resolution spectrometer.

We used the autocorrelation technique for measuring the time dependent pulse shape. This is appropriate if we assume that the pulse envelope, $\mathcal{E}(z,t)$, is real and is uniquely connected to the time dependent pulse intensity by $I_{\rm ac}(\tau) \sim \int I(t)I(t-\tau)\,dt$. Thus our results are most conveniently presented as an autocorrelation trace which can be transformed into a time dependent pulse intensity. We have checked that sensitivity to changes in the pulse shape are not lost due to the indirect measurement of the autocorrelation function. We have previously shown [11] that even small ($\sim$10%) changes in the pulse intensities give rise to significant changes in the autocorrelation trace.

Experimental and theoretical autocorrelations for low-intensity 0-π pulses propagated through high-density K are shown in Fig. 1. The theoretical predictions are based on Eq. (2), with the calculation having no adjustable parameters. The maximum value of the calculated autocorrelation is normalized to the maximum of the autocorrelation of a low-intensity laser pulse propagated through a room temperature K cell, providing a very low value of optical depth, shown in the inset of Fig. 1. All subsequent autocorrelations are normalized to this low-intensity trace and can thus result in large numerical values on the y axis of the autocorrelations. Pulse reshaping for 0-π pulses was found to be independent of the polarization of the incident pulse. This is consistent with Eq. (3) since the sum of the square of the dipole moments for linear and circular polarizations are identical.

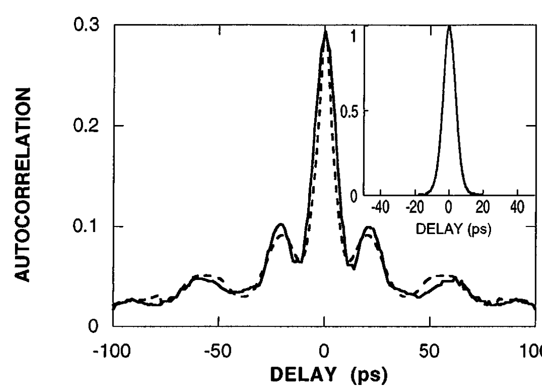

FIG. 1. Normalized autocorrelations for 6 ps low-intensity pulses after propagation through potassium vapor with $\alpha l = 170$. The solid line is the experiment and the dashed line is the calculation based on Eq. (1). The autocorrelation of the input pulse is shown in the inset.

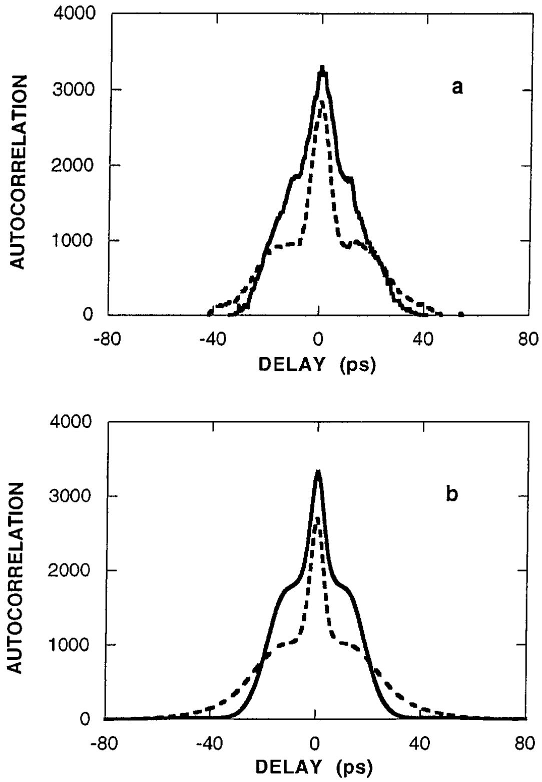

FIG. 2. Normalized autocorrelations for 6 ps intermediate-intensity pulses (pulse area in the range of π to 2.45π) after propagation through potassium with $\alpha l = 170$. The solid line is the data for linear polarization and the dashed line is the data for circular polarization: (a) experiment and (b) calculation based on Eq. (1).

In Fig. 2 we show the measured and calculated autocorrelations for higher-intensity laser pulses. Here the small area approximation is no longer valid with the pulse area lying in the range of π to 2.45π, depending on the particular transition dipole moment. For such pulses we still observe a significant breakup, however, the results for circular and linear polarizations appear different. This can be understood from Eq. (1) by noting that when the small area approximation fails we have to consider the next terms in the expansion of the sin. The linear term is proportional to the sum of the squares of the dipole moments while the next term is proportional to the sum of the dipole moments to the fourth power, the third term to the sum of the dipole moments to the sixth power, etc. As we noted above, the sum rule for dipole moments

$$\sum_{F,F',m_F,m_{F'}} p^2_{F,F',m_F,m_{F'}}\Big|_{q=1} = \sum_{F,F',m_F,m_{F'}} p^2_{F,F',m_F,m_{F'}}\Big|_{q=-1} = \sum_{F,F',m_F,m_{F'}} p^2_{F,F',m_F,m_{F'}}\Big|_{q=0}$$

explains why there is no difference between circular and linear polarization for small area pulses. However,

$$\sum_{F,F',m_F,m_{F'}} p^4_{F,F',m_F,m_{F'}}\Big|_{q=1} = \sum_{F,F',m_F,m_{F'}} p^4_{F,F',m_F,m_{F'}}\Big|_{q=-1} \neq \sum_{F,F',m_F,m_{F'}} p^4_{F,F',m_F,m_{F'}}\Big|_{q=0}$$

with similar inequalities for higher order terms, and therefore propagation of more intense pulses is expected to depend on polarization.

With increasing input pulse area, reshaping decreases and disappears all together. The measured and calculated autocorrelations for high-intensity pulses are shown in Fig. 3 with the pulse area in the range of 2π to 4.9π. Since no reshaping occurs, the results for circular and linear polarization for such big area pulses are approximately the same, with only a small hint of a difference remaining in the wings of the pulse. Further increase of the input pulse intensity results in a totally negligible difference between the two polarizations and gives undistorted pulse propagation, as shown in Fig. 4, for the pulse area between 4π and 9.8π. We have not been able to measure any attenuation for these high-intensity pulses.

These results indicate that for values of optical depth, $\alpha l < 200$, pulse breakup, such as was observed [3,4] in the breakup of a 4π pulse into two 2π pulses, does not

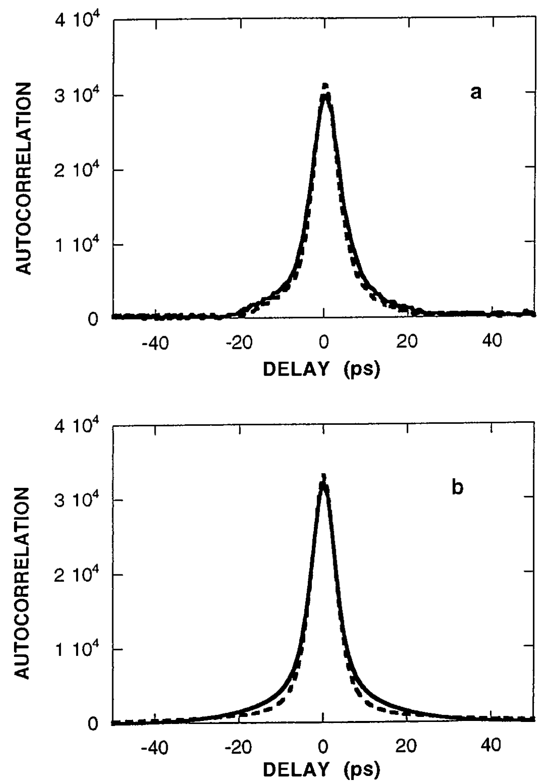

FIG. 3. Normalized autocorrelations for 6 ps high-intensity pulses (pulse area in the range of 2π to 4.9π) after propagation through potassium with $\alpha l = 170$. The solid line is the data for linear polarization and the dashed line is the data for circular polarization: (a) experiment and (b) calculation based on Eq. (1).

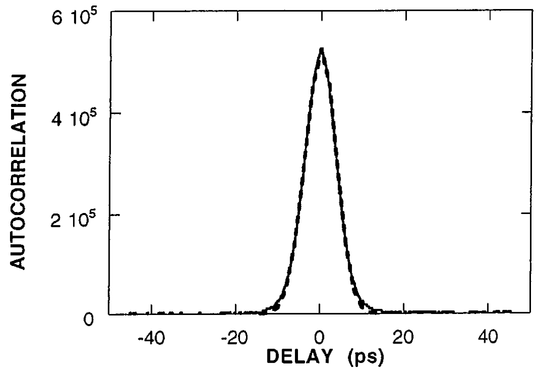

FIG. 4. Normalized autocorrelations for 6 ps very high-intensity pulses (pulse area in the range of 4π to 9.8π) after propagation through potassium with $\alpha l = 170$. The solid line is the experimental pulse autocorrelation after propagation and the dashed line is the autocorrelation of the initial pulse. The calculation based on Eq. (1) gives identical results to the observation and is not shown.

occur. Numerical estimates of energy losses to the atomic medium show that these results cannot be explained as being due to the incoherent bleaching of the atomic transition, which would lead to a nondispersive medium. From our numerical evaluation of the Maxwell-Bloch equations, we found that no significant population remains in the excited state over the time between pulses (12.2 ns). Rather, we interpret the absence of pulse breakup as due to two reasons. The first has to do with the multiplicity of levels and consequent unequal dipole moments which tend to reduce the sensitivity to pulse breakup. This is evident from the last term in Eq. (1) which is a sum of many incommensurate oscillators that have to be summed to produce the polarization of the medium.

The second reason for the cessation of pulse breakup with increasing pulse area is that, even for a two level system possessing a unique dipole moment, pulse breakup becomes less significant at pulse areas exceeding 2π. Thus, even in this simpler case, the breakup of a 4π pulse into two 2π pulses occurs only for very large αl. When translated to an effective optical depth [4] $\alpha_{\mathrm{eff}} l = \alpha l \frac{\tau}{T^*}$, where τ is the pulse duration and T^* is the inhomogeneous broadening time, our observations and calculations show that the significant pulse breakup of high intensity pulses occurs only if $\alpha_{\mathrm{eff}} l \geq 2$. This is due to the sin in front of the integral in Eq. (1), which does not permit the polarization of the medium to grow indefinitely as the pulse area is increased. In other words, for larger pulse areas, saturation sets in and the index of the medium stops playing its normal dispersive role.

In summary, we have investigated, theoretically and experimentally, large area pulse propagation through a multilevel degenerate absorber in the extreme SLL. We examine the transition from small to large area pulses, and obtain that, for $\alpha_{\mathrm{eff}} l \sim 1$, strong reshaping occurs only for low energy pulses, in agreement with dispersion theory. For a larger pulse area the sensitivity of the pulse to breakup is reduced, and the propagation is shown to be transition dipole moment dependent as evidenced by the different behavior for circular and linear polarizations. For very high pulse intensities the propagation is undistorted until $\alpha_{\mathrm{eff}} l > 2$.

The authors wish to acknowledge the helpful comments of A. Wilson-Gordon and J. Rothenberg, and partial support from the Israel Academy of Sciences. M. M. acknowledges the support of an Eshkol fellowship.

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