# $2N\pi$ Ultrashort Light Pulses

Thomas W. Barnard

The Perkin-Elmer Corporation, Wilton, Connecticut 06897

(Received 11 September 1972)

A method for obtaining analytical expressions for  $2N\pi$  pulses in an unbroadened resonant medium is derived by a systematic application of the Bäcklund transformation. The resulting large-area pulses display the expected property of pulse breakup into separate  $2\pi$  hyperbolic secant pulses.

#### INTRODUCTION

A distinctive property of ultrashort-light-pulse propagation in resonant media is the conservation of pulse area for multiple  $2\pi$  pulses. This result, which applies to an ideal attenuator with inhomogeneously broadened line shape, was first stated in an area theorem by McCall and Hahn.<sup>1</sup> A similar result holds for unbroadened ideal attenuators.<sup>2</sup> In this communication, we shall derive analytical expressions for multiple  $2\pi$  pulses to any order which may propagate in unbroadened media without change of pulse area. As these pulses propagate in the resonant medium, they evolve into separate  $2\pi$  hyperbolic secant (hs) pulses characteristic of self-induced transparency (SIT). A similar result has been obtained for multiple  $2\pi$  pulses in broadened media by numerical integration of the equations of motion.<sup>1,3</sup>

## EQUATIONS OF MODEL

The equations of motion for ultrashort-pulse propagation in resonant media have been discussed in several papers<sup>1,2,4</sup> and so they will only be reviewed here. The classical field serves as a perturbation to the two-level nondegenerate resonant medium which is treated in the dipole approximation according to the Bloch formalism.<sup>5</sup> The field is assumed to be a linearly polarized plane wave propagating in the z direction and is approximated by a carrier frequency  $\phi = \omega_0(t - z/c)$  and a slowly varying amplitude  $\epsilon(z, t)$  and phase  $\theta(z, t)$ :

$$E(z,t) = \epsilon(z,t) \cos[\phi - \theta(z,t)] .$$
(1)

If we assume that the atomic transition frequency is equal to the field-carrier frequency, the equations of motion  $reduce^2$  to

$$\left(\frac{\partial}{\partial t} + c \; \frac{\partial}{\partial z}\right) \tilde{\epsilon} = \frac{2\pi n_0 \omega_0 p^2}{\hbar} S \;, \tag{2}$$

$$C=0$$
, (3)

$$\dot{\mathbf{S}} = \tilde{\boldsymbol{\epsilon}} \boldsymbol{\eta} \,, \tag{4}$$

$$\dot{\eta} = -\tilde{\epsilon} S , \qquad (5)$$

where C and S are defined in terms of the macro-

scopic polarization P(z, t),

 $P(z,t) = n_0 p[C(z,t)\cos\phi + S(z,t)\sin\phi], \qquad (6)$ 

and  $\eta(z, t)$  is the normalized atomic population inversion

 $\eta(z, t) = |a(t)|^2 - |b(t)|^2$ .

a(t) and b(t) are the coefficients of upper and lower energy states in the atomic wave function, p is the atomic dipole matrix element,  $n_0$  is the density of active atoms, and  $\boldsymbol{\xi} = (p/\hbar)\boldsymbol{\epsilon}$ .

Equations (4) and (5) may be integrated in terms of the partial field area

$$\psi(z,t) = \int^{t} \boldsymbol{\xi}(z,t) dt \tag{7}$$

to give

$$S(z, t) = \sin\psi(z, t) ,$$
  

$$\eta(z, t) = -\cos\psi(z, t) .$$
(8)

Equation (2) may then be expressed as

$$\frac{\partial^2 \psi}{\partial \xi \partial \tau} = -\sin\psi \tag{9}$$

in terms of the variables  $\tau = \alpha(t - z/c)$  and  $\xi = \alpha z/c$ , where

$$\alpha = (2\pi n_0 \omega_0 p^2 / \hbar)^{1/2}$$
.

We wish to find solutions of Eq. (9) for which  $\psi(z, +\infty)$  is an integer multiple of  $2\pi$ . As may be seen by integrating Eq. (9) from  $-\infty$  to  $+\infty$ , these solutions propagate without change in area. A useful technique for obtaining solutions to Eq. (9) has been pointed out by Lamb<sup>6</sup> and consists of the Bäcklund transformation.<sup>7</sup> This method permits generating a second solution of Eq. (9) from a known one by solving the coupled pair of first-or-der partial-differential equations

$$\frac{\partial}{\partial \xi} \frac{1}{2} (\psi_1 - \psi_0) = a \sin \frac{1}{2} (\psi_1 + \psi_0) ,$$

$$\frac{\partial}{\partial \tau} \frac{1}{2} (\psi_1 + \psi_0) = -\frac{1}{a} \sin \frac{1}{2} (\psi_1 - \psi_0) ,$$
(10)

where a is an arbitrary constant. We denote the solution of Eqs. (10) for  $\psi_1$  in terms of  $\psi_0$  by the

.

373

7





FIG. 1. Notation convention for pulses linked by Bäcklund transformation.

expression  $\psi_1 = B_a \psi_0$ . If four solutions of Eq. (9) are linked by Bäcklund transformations in terms of two constants, as indicated below,

$$\begin{split} \psi_1 &= B_{a_1} \psi_0 \ , \\ \psi_2 &= B_{a_2} \psi_0 \ , \\ \psi_3 &= B_{a_2} \psi_1 = B_{a_1} \psi_2 \ , \end{split}$$

then the four solutions obey a simple trigonometric relationship

$$\tan\left(\frac{\psi_3 - \psi_0}{4}\right) = \frac{a_1 + a_2}{a_1 - a_2} \tan\left(\frac{\psi_1 - \psi_2}{4}\right) \quad . \tag{11}$$

New solutions of the second-order partial-differential equation [Eq. (9)] may thus be expressed as algebraic combinations of known solutions.

#### $2N\pi$ PULSES

In this section a recursion relation is derived for generating  $2N\pi$  pulses, where N is any integer. A diagrammatic representation of the Bäcklund transformation suggested by Lamb<sup>2</sup> will be useful in visualizing the analytical technique (see Fig. 1). Figure 1 also defines the notation convention for the four solutions of Eq. (9),  $\psi_{i,j}$ , etc., and the two constants  $a_i$  and  $a_j$ , which are related by the trigonometric form of the Bäcklund transformation [Eq. (11)].

The pulse area  $\theta_{i,j} = \psi_{i,j}(z, +\infty)$  is given by

$$\theta_{i,j} = \theta_{i+1,j-1} + \theta_{i,j-1} - \theta_{i+1,j} , \qquad (12)$$

if  $(a_i + a_j)/(a_i - a_j) > 0$  and if  $\theta_{i+1,j-1}$ ,  $\theta_{i,j-1}$ , and  $\theta_{i+1,j}$  are integer multiples of  $2\pi$ . This may be demonstrated by following the evolution of

$$\psi_{i,j}(z,t) = \psi_{i+1,j-1}(z,t) + 4 \tan^{-1} \left[ (a_i + a_j) / (a_i - a_j) \tan \frac{\psi_{i,j-1} - \psi_{i+1,j}}{4} \right]$$
(13)

across the branch points of the tangent function as t goes from  $-\infty$  to  $+\infty$ . The field amplitude is also obtained from this expression by differentiation with respect to time:

$$\tilde{\boldsymbol{\epsilon}}_{i,j} = \tilde{\boldsymbol{\epsilon}}_{i+1,j-1} + K_{ij} \frac{1 + g_{ij}^2}{1 + K_{ij}^2 g_{ij}^2} (\tilde{\boldsymbol{\epsilon}}_{i,j-1} - \tilde{\boldsymbol{\epsilon}}_{i+1,j}) , \quad (14)$$

where

$$K_{ij} = (a_i + a_j)/(a_i - a_j)$$

and

$$g_{ij} = \tan \left[ \frac{1}{4} (\psi_{i,j-1} - \psi_{i+1,j}) \right]$$

The simplest solution of Eq. (9) is the trivial  $\psi = 0$ . If we let  $\psi_{i+1,i} = 0$  for all i > 0, then it follows from Eq. (10) that  $\psi_{i,i} = 4 \tan^{-1} e^{u_i}$ , where  $u_i = a_i \alpha (t - z/v_i) + \gamma_i$ , where  $\gamma_i$  is a constant and  $1/v_i = (1/c)(1 + 1/a_i^2)$  for all i > 0. The corresponding electric field amplitude is  $\xi_{i,i} = 2\alpha a_i \operatorname{sech} u_i$ . Since  $\psi_{i,i}(z, +\infty) = \pm 2\pi$  for  $a_i \ge 0$ ,  $\psi_{i+1,i}$  and  $\psi_{i,i}$  can be used as a basis for building multiple  $2\pi$  pulses by repeated use of Eq. (13). Figure 2 shows a sequence of transformations that may be extended systematically to obtain analytical expressions for arbitrary large-area (or negative-area) pulses. The pulse areas are indicated in the circles which designate the pulse functions. The pulse area is alternately  $+2M\pi$  and  $-2M\pi$  across each row, where M is the number of rows from the bottom.

The transformation constants must be constrained by a set of inequalities in order to obtain  $2N\pi$ pulses. Since the second row of pulses has alternately  $+2\pi$  and  $-2\pi$  area, the constants must be alternately positive and negative.

Thus,



FIG. 2. Sequence of Bäcklund transformations for generation of  $12\pi$  pulse.



FIG. 3.  $12\pi$  pulse (a) before and (b) after pulse breakup for the constants  $a_1 = 3.0$ ,  $a_2 = -2.5$ ,  $a_3 = 2.2$ ,  $a_4 = -1.6$ ,  $a_5 = 1.0$ , and  $a_6 = -0.4$ .

$$(-1)^i a_i < 0$$
 . (15)

Also, the inequality

$$(a_i + a_j)/(a_i - a_i) > 0$$
 (16)

must be preserved for all j > i. It may readily be seen that both Eqs. (15) and (16) are satisfied if

$$(-1)^{i}a_{i} < (-1)^{j}a_{i}, \quad j > i$$
 (17)

which yields a self-consistent set of inequalities for the transformation constants. The expressions which define the  $2N\pi$  pulses are summarized below:

$$\begin{array}{c} \psi_{i+1,i} = 0 , \\ \psi_{i,i} = 4 \tan^{-1} e^{u_i} , \\ (-1)^i a_i < 0 , \end{array} \right\} \qquad i > 0 ; \\ \psi_{i,j} = \psi_{i+1,j-1} + 4 \tan^{-1} \left[ K_{ij} \tan \left( \frac{\psi_{i,j-1} - \psi_{i+1,j}}{4} \right) \right] , \\ \tilde{\epsilon}_{i,j} = \tilde{\epsilon}_{i+1,j-1} + K_{ij} \frac{1 + g_{ij}^2}{1 + K_{ij}^2 g_{ij}^2} (\tilde{\epsilon}_{i,j-1} - \tilde{\epsilon}_{i+1,j}) \\ \theta_{i,j} = (-1)^{i+1} (j-i+1) 2\pi , \\ (-1)^i a_i < (-1)^j a_i , \end{array} \right) j > i .$$

In order to generate the analytical expression for one of the higher-order pulses, it is necessary first to obtain the expressions for the constituent pulses. This corresponds in Fig. 2 to finding the expressions for all solutions within the triangle whose apex is the solution in question and whose base is the set of trivial  $0\pi$  pulses.

### DISCUSSION

It has been noted previously<sup>1</sup> that the only stable SIT pulse is the  $2\pi$  hs pulse and that, consequently, a multiple  $2\pi$  pulse will break up into separate  $2\pi$ hs pulses. This fact may be verified by calculating the field envelope according to Eq. (14). The result is also expected from the manner in which higher-order pulses are constructed by means of the Bäcklund transformation.

In order to obtain a  $2N\pi$  pulse, one must combine N hs pulses each with a different velocity. For large values of z, only one retarded-time variable  $u_i = a_i \alpha (t - z/v_i) + \gamma_i$  will be near zero for a given value of t. Since the pulse is centered about  $u_i = 0$  and has significant amplitude only in a limited region around zero, it is as though only one nontrivial pulse were present in the hierarchy of Bäcklund transformations for a given value of z, t. This is equivalent to requiring that all  $a_i$  be zero except for one such as  $a_i$ , where *i* depends on z, t. It is clear from Eqs. (13) and (14) that any multiple  $2\pi$ pulse will consist of separate  $2\pi$  hs pulses.

A plot of a  $12\pi$  pulse is shown in Fig. 3 before and after it has evolved into six  $2\pi$  hs pulses. The pulses are ordered according to their velocities  $c[a_i^2/(1+a_i^2)]$ , their widths  $1/\alpha a_i$ , and their amplitudes  $\alpha a_i$ , as would be expected.

# ACKNOWLEDGMENTS

The author wishes to thank E. A. Mishkin for suggesting this problem and B. A. Lippmann for many helpful discussions.

JANUARY 1973

<sup>1</sup>S. L. McCall and E. L. Hahn, Phys. Rev. Letters <u>18</u>, 908 (1967); Phys. Rev. <u>183</u>, 457 (1969).

<sup>2</sup>G. L. Lamb, Jr., Rev. Mod. Phys. <u>43</u>, 99 (1971). <sup>3</sup>F. A. Hopf and M. O. Scully, Phys. Rev. <u>179</u>, 399 (1969).

<sup>4</sup>A. Icsevgi and W. E. Lamb, Jr., Phys. Rev. <u>185</u>,

PHYSICAL REVIEW A

#### 517 (1969).

 ${}^{5}$ R. P. Feynman, F. L. Vernon, Jr., and R. W. Hell-warth, J. Appl. Phys. <u>28</u>, 49 (1957).

<sup>6</sup>G. L. Lamb, Jr., Phys. Letters <u>25A</u>, 181 (1967).

<sup>7</sup>A. V. Bäcklund, Math. Ann. <u>9</u>, 297 (1876).

# Hartree-Fock Third-Harmonic Coefficient of Neon

VOLUME 7, NUMBER 1

Ralph Klingbeil

Institute for Fluid Dynamics and Applied Mathematics, University of Maryland, College Park, Maryland 20742 (Received 20 July 1972)

This is a note concerned with a previous publication by R. Klingbeil, V. G. Kaveeshwar, and R. P. Hurst [Phys. Rev. A 4, 1760 (1971)] giving specific results for neon.

Recently Klingbeil, Kaveeshwar, and Hurst<sup>1</sup> (KKH) derived the Hartree-Fock expression for the third-harmonic coefficient (THC) of an atomic system by a method employing Chung's time-dependent perturbation theory.<sup>2</sup> KKH also obtained the Hartree-Fock THC (HFTHC) of helium. It is the purpose of this note to present the results of a calculation of the HFTHC of neon.

It is necessary to obtain zeroth-, first-, and second-order wave functions in order to evaluate the expression for the THC given by Eq. (17) of KKH. The zeroth-order wave function chosen is that of Clementi.<sup>3</sup> First-order wave functions are obtained by the method of Kaveeshwar, Chung, and Hurst,<sup>4</sup> and agreement with their calculation of the linear dynamic polarizability is excellent. Secondorder wave functions are obtained by the method of

TABLE I. Hartree-Fock third-harmonic coefficient of neon.

ω (a.u.)	λ(Å)	$\chi_{zzzz}(-3\omega; \omega, \omega, \omega)$ (a.u.)
0.000	∞	54.0 <sup>a</sup>
0.025	18238	54.9
0.050	9119	57.9
0.075	6079	63.2
0.100	4559	72.0
0.125	3648	86.1
0.150	3040	109.6
0.175	2605	152.5
0.200	2280	246.7
0.225	2026	570.6
0.247	1846 <sup>b</sup>	100939.1
0.248	1838	-16333.3

 ${}^{2}\chi_{\text{szeg}}(-3\omega; \omega, \omega, \omega) = 54.0 \text{ a.u. at zero frequency.}$ 

 $b_{1}^{1}(1846 \text{ Å}) = 615 \text{ Å}$  is the calculated value of the first transition wavelength. The experimental value is 736 Å.

KKH.

The results of the calculation of the HFTHC  $X_{zzzz}(-3\omega; \omega, \omega, \omega)$  of neon are presented in Table I and plotted in Fig. 1 along with the HFTHC of helium<sup>1</sup> for comparison. At zero frequency the HFTHC of neon is 54.0 a.u. Surprisingly, this result is significantly larger than the Hartree-Fock static hyperpolarizability result of 42 a.u. obtained by Sitter and Hurst.<sup>5</sup> Both results are considerably below the experimental static hyperpolarizability



FIG. 1. Hartree-Fock third-harmonic coefficient  $\chi_{xxxx}(-3\omega; \omega, \omega, \omega)$  of helium (Ref. 1) and neon (this work).