Wave Equation for Dark Coherence in Three-Level Media

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We report the derivation of a wave equation for coherence in "dark state" two-photon-resonance spectroscopy. One of its consequences is a dark state area theorem. The dark area theorem is a single ordinary differential equation which is globally equivalent, in a way we describe, to the full set of five coupled nonlinear partial differential equations that govern space-time evolution of two-pulse coherence in a lambda medium. The predictions of the dark area theorem are open to test via laser spectroscopy in dilute vapors and inhomogeneously broadened solids.

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The so-called "dark state" of double-resonance spectroscopy arises from optical pumping relaxation that puts quantum amplitudes into a specific coherent superposition of ground states that is immune to further excitation. The dark state appears as an extremely sharp hole in the double-resonance absorption line [1]. It has played a role in studies of lasing without inversion (LWI) and electromagnetically induced transparency (EIT) [2], as well as of coherent population transfer [3], including effects recently associated with optical information storage [4–7].

Our goal is a theoretical formulation of coherence evolution in three-level media similar to the "area theorem" discovered by McCall and Hahn [8], which is an ordinary differential equation for spatial evolution of coherence in two-level single-resonance media. The coherence parameter identified by McCall and Hahn, and the key to global understanding of the coupled Maxwell-Bloch partial differential equations for two-level media, is the pulse "area," i.e., the dimensionless time integral of the envelope of the propagating pulse: $A(z) \equiv \int_{-\infty}^{+\infty} \Omega(z,t')\,dt'$, where the Rabi frequency $\Omega(z,t) \equiv 2\,\mathrm{ex}_{12}\mathcal{E}(z,t)/\hbar$ is the dimensionally scaled real envelope. Here ex_{12} is the real dipole matrix element of the two-level transition.

The area A is conventionally written as an angle θ because in two-level quantum systems it has a second interpretation as the tipping angle of the Bloch vector for the system. We will demonstrate for three-level double resonance that the pulse integral definition as well as the tipping angle definition fails. Only the definition of a "dark area," arising from dark state considerations, gives access to a global understanding of coherence, in the same sense as in the McCall-Hahn area theorem, i.e., in the sense that while details of pulse shape are sacrificed in a time integral that spans the duration of the physical pulses, the integral sharply clarifies important features of asymptotic spatial evolution.

The McCall-Hahn area theorem is written

$$\frac{d}{dz}A = -\frac{\alpha}{2}\sin A\,. \qquad (1)$$

Its first unexpected feature is that it is "merely" an ordinary differential equation (ODE) able to represent a set of three coupled nonlinear partial differential equations (PDE's) [8], and its second is that it makes accurate predictions about pulse propagation despite its highly nonlocal temporal character. For example, the area theorem explains easily, in fact it first predicted, the effect of self-induced transparency (SIT), in which a pulse can coherently alter a resonant attenuating medium in such a way as to be transparent, but only to the pulse itself.

An apparent obstacle in seeking an overall propagation theory for three-level absorbers is that exact integrability for the relevant PDE's has never been established, whereas exact integrability in the two-level case is closely linked to the solitons associated with SIT. It is interesting that our theory does not need the property of exact integrability, while it still admits known special-case solutions for solitons within its domain.

The key to approaching pulse propagation in three-level media is to change the focus from the physical pulses themselves. Our derivation is based on the dressing transformation in Hilbert space that connects the two lower states of a lambda system with its bright and dark "dressed" states of three-level spectroscopy [1], as indicated in Fig. 1(a) and expressed by the matrix relation between the corresponding probability amplitudes:

$$\begin{bmatrix} B \\ D \end{bmatrix} = \begin{bmatrix} \cos\theta/2 & \sin\theta/2 \\ -\sin\theta/2 & \cos\theta/2 \end{bmatrix} \begin{bmatrix} c_g \\ c_{g'} \end{bmatrix}. \qquad (2)$$

The dressing angle θ measures the induced dark coherence of the atom-field interaction and is defined as a function of z and t in terms of the Rabi frequencies of both physical field envelopes assumed to be real: $\cos(\theta/2) \equiv \Omega_g/\Omega_B$ and $\sin(\theta/2) \equiv \Omega_{g'}/\Omega_B$, where the auxiliary parameter $\Omega_B \equiv \sqrt{\Omega_g^2 + \Omega_{g'}^2}$ is called the bright Rabi frequency. The well-known equations for the original three-level two-photon-resonant Schrödinger amplitudes are

$$i\,\frac{\partial}{\partial t}\,c_g = -\frac{1}{2}\,\Omega_g c_a\,, \qquad (3)$$

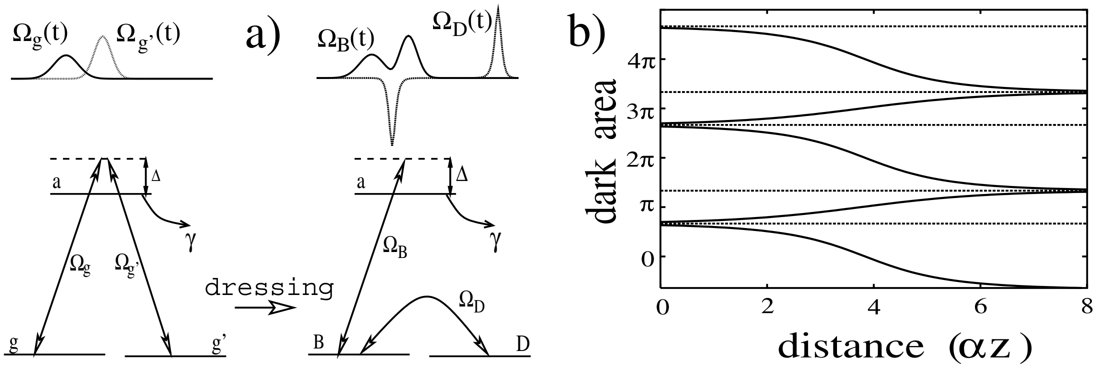

FIG. 1. Two pulses, a lambda atom, and the solution branches for Eq. (6). (a) Above: physical pulse shapes Ω_g and $\Omega_{g'}$ and their corresponding dressed pulse shapes Ω_B and $-i\Omega_D$ are plotted vs time. Note the strikingly unphysical appearance of $-i\Omega_D$, with its strong peak in the far tail of both physical pulses. Below: bare-state versus dressed-state sketches indicating lambda-atom physics. The common detuning is denoted Δ, and γ is the homogeneous decay rate of the upper level (out of the system). (b) Multibranched behavior of the dark area θ_D with distance, as predicted by Eq. (6). Asymptotes are determined by $\theta_D + \phi = 2n\pi$ and $\tan(\theta_D/2)\tan(\phi/2) = \alpha/\alpha'$. The values used here are $\phi = 2\pi/3$ and $\alpha' = 3\alpha$.

$$i\frac{\partial}{\partial t}c_{g'} = -\frac{1}{2}\Omega_{g'}c_a, \tag{4}$$

$$i\frac{\partial}{\partial t}c_a = (\Delta - i\gamma)c_a - \frac{1}{2}(\Omega_{g'}c_{g'} + \Omega_g c_g). \tag{5}$$

Remarkably, the same dressing angle θ has another definition, as the time integral of an auxiliary function, which is not a dimensionally scaled pulse envelope but a nonlinear combination of scaled pulse envelopes, a combination first used as a "dark Rabi frequency" by Fleischhauer and Manka [9], and involving time derivatives:

$$\Omega_D \equiv 2i(\Omega_{g'}\dot{\Omega}_g - \Omega_g\dot{\Omega}_{g'})/\Omega_B^2.$$

That is, one also has $\theta(z,t) = i\int_{-\infty}^{t} dt'\,\Omega_D(z,t')$. As the plots in Fig. 1(a) illustrate, because of the derivatives in the definition of the dark Rabi frequency, it may have almost no obvious relation to the two physical Rabi frequencies.

We next define $\lim(t\to\infty)\theta(z,t) \equiv \theta_D(z)$ as the total dark area. The surprise is that $\theta_D(z)$, a time integral over a complex nonlinear combination of two propagating fields, evolves spatially according to a relatively simple single ODE, a "dark area theorem":

$$\frac{d\theta_D}{dz} = -\frac{\alpha + \alpha'}{4}\sin(\theta_D + \phi) + \frac{\alpha - \alpha'}{4}(\sin\theta_D + \sin\phi), \tag{6}$$

for which we sketch the derivation in the following paragraphs. The clear similarity to the McCall-Hahn equation (1) is evident. The infinite set of trigonometric branch solutions to (6) are shown in Fig. 1(b). The constant ϕ is defined by $\tan(\phi/2) \equiv \lim(t\to -\infty)c_g/c_{g'}$; i.e., it is determined by the initial state of coherence of the medium before the pulses arrive. The Beer's law absorption coefficients are $\alpha = \pi\mu F(0)$ and $\alpha' = \pi\mu' F(0)$ with $F(\Delta)$ as the inhomogeneous detuning distribution function and Δ is shown in Fig. 1(a). Here μ and μ' are the coupling coefficients for the a-g and a-g' transitions.

The five partial differential equations governing propagation of Ω_g and $\Omega_{g'}$ and evolution of coherence in three-level systems have been long known [10–15]. In addition to (3)–(5) one has the propagation equations

$$\left(\frac{\partial}{\partial z} + \frac{\partial}{\partial ct}\right)\Omega_g = i\mu\langle c_a c_g^*\rangle, \tag{7}$$

$$\left(\frac{\partial}{\partial z} + \frac{\partial}{\partial ct}\right)\Omega_{g'} = i\mu'\langle c_a c_{g'}^*\rangle, \tag{8}$$

where angular brackets denote inhomogeneous averaging taken below as a Gaussian with characteristic time T^*. Then, by combining the dressing definition of θ with the propagation equations for the two physical envelopes, one can find a wave equation for dark coherence, i.e., a wave equation obeyed by the Hilbert space rotation angle:

$$\left(\frac{\partial}{\partial z} + \frac{\partial}{\partial ct}\right)\theta(z,t) = \frac{2i}{\Omega_B}[\mu_+\langle c_a D^*\rangle - \mu_-\langle c_a B^*\rangle], \tag{9}$$

where $\mu_+ \equiv \frac{1}{2}(\mu + \mu')$ and $\mu_- \equiv \frac{1}{2}(\mu - \mu')$. Equation (9) has not previously been derived, so far as we know, although a related equation without inhomogeneous broadening, and with $\mu_- = 0$, was used by Fleischhauer and Manka [9] to study quasiadiabatic pulse propagation. We note that our derivation did not involve assumptions common in lambda physics such as adiabatic elimination, fast homogeneous damping, weak probe pulses, or near-cw pump pulses. To go further we require only a relatively broad inhomogeneous line. This is commonly available in Doppler-broadened vapors and in low-temperature solids such as the rare-earth doped insulator Y_2SiO_5 (Pr:YSO), which has been used recently for EIT studies [7]. Other relevant materials are those for which hole-burning or echo effects are observable, and several are noted in [16].

We next need the equation for the upper-level amplitude, and when expressed in terms of the dressed state amplitudes, it is found to be

$$i\frac{\partial}{\partial t}c_a = (\Delta - i\gamma)c_a - \frac{1}{2}\Omega_B B, \tag{10}$$

and by formally integrating this equation, with $c_a(-\infty) = 0$, we get expressions for the terms required in (9):

$$\langle c_a D^* \rangle = \frac{i}{2}\frac{T^*}{\sqrt{2\pi}}\int_{-\infty}^{\infty} D^*\left[\int_{-\infty}^{t} e^{(\gamma + i\Delta)(\tau - t)}(\Omega_B B)\,d\tau\right] \times e^{-(1/2)(T^*\Delta)^2}\,d\Delta, \tag{11}$$

and similarly for $\langle c_a B^* \rangle$. We extend the upper limit in the time integral to a "final" time T_f in the far tail of the physical pulses. The resulting double integrals can then be evaluated [17] in the regime of rapid inhomogeneous relaxation ($T^* \ll \tau, \tau', \gamma^{-1}$, where τ and τ' are durations of the pulses). One finds at time T_f the compact formulas: $i\langle c_a D^* \rangle \approx -\frac{\pi}{2}F_0\Omega_B D_0 B_0$ and $i\langle c_a B^* \rangle \approx -\frac{\pi}{2}F_0\Omega_B B_0^2$, where the subscript 0 denotes $\Delta = 0$. One also finds $D(T_f) = \cos\frac{1}{2}(\theta_D + \phi)$ and $B(T_f) = \sin\frac{1}{2}(\theta_D + \phi)$. These factors simplify the right-hand side of the wave equation for θ; it loses its t dependence and becomes an equation for θ_D in z alone. The dark area theorem, i.e., the single nonlinear ODE given in (6), is the immediate consequence.

A transparency of a lambda absorber (cf. [2]) can be induced by a pair of pulses working together to create a dark state—the state in which destructive quantum interference cancels absorption. Prior work has already led to a consensus about an asymptotic feature: the final pulse envelopes must be matched [18] in their temporal shape, i.e., $\lim(z \to \infty)\Omega_g(z,t)/\Omega_{g'}(z,t) = K$. The dark area theorem is in agreement with this, but it also predicts both the value of the constant K and the rate of spatial evolution toward the matched state. It also gives the first formula for the spatial development of the dark and bright state amplitudes that form in the wake of the pulses:

$$Q(z, T_f) \equiv \frac{D^2}{B^2} = \frac{D_{\text{in}}^2}{B_{\text{in}}^2}\exp[\alpha(z - z_{\text{in}})], \tag{12}$$

where D_{in} and B_{in} are values at an arbitrary early spatial position z_{in}. We put $\mu = \mu'$ here only for the sake of simplicity. Conservation of probability, $B^2(z, T_f) + D^2(z, T_f) = 1$, gives the dark and bright amplitude solutions separately. Monotonic growth of $Q(z, T_f)$ is equivalent to monotonic conversion of population into the dark state, as illustrated in Fig. 2. This is a conclusion also reached in our earlier mostly numerical study of the distinction between SIT-type and EIT-type propagation [19].

Given the complicated form of Ω_D in terms of the physical Rabi frequencies [recall Fig. 1(a)], spatial evolution of the dark area is remarkably straightforward. Starting from any initial value, $\theta_D(z)$ will monotonically evolve along its branch toward the asymptotically stable value. The dark area theorem says that the tangent of half this value determines the ratio of the final Rabi frequencies $\Omega_{g'}$ and Ω_g, or of the final field envelopes. This is illustrated in Fig. 3. In strict analogy with the McCall-Hahn theorem [8], the dark area theorem (6) makes a global statement about the pulses and provides no details about their evolving shapes [20].

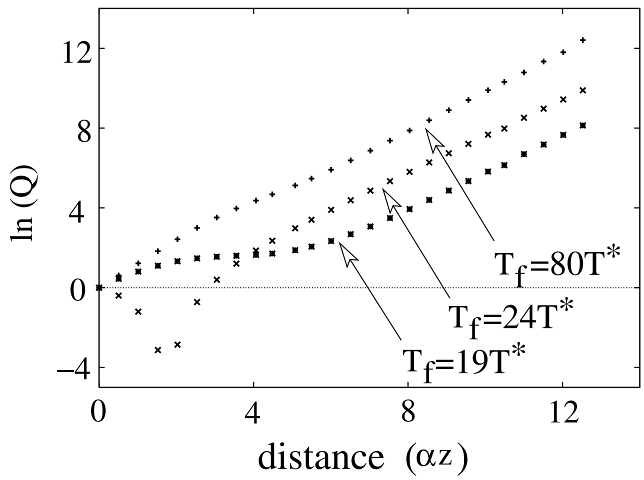

FIG. 2. The specific prediction of Eq. (12) is confirmed by the straight-line behavior shown. We have taken data from the same numerical integration used to obtain example (a) of Fig. 3. We plot $\ln Q$ vs αz for three different times: $T_f = 19T^*, 24T^*$, and $80T^*$. The asymptotically straight line for $T_f = 80T^*$ has approximately unit slope, in good agreement with dark area propagation theory. The superior agreement at the longest time, $T_f = 80T^*$, is expected, as (12) is strictly valid only after the pulses have completely passed the position z.

To confirm that the simple expressions presented here are accurate, we directly integrated the full system of five fundamental lambda PDE's for fields and probability amplitudes and show in Figs. 3(a) and 3(b) two space-time pulse-propagation scenarios obtained in this way. The captions mention several comparisons with predictions of the dark area theorem. Our formulas and equations are applicable to initially matched or unmatched pulse pairs and remain valid even if the pulses change substantially in either time or space. As remarked above, they do not require assumptions such as adiabatic elimination, fast damping, weak probe pulses, or cw pump pulses, but they rather require only a relatively broad inhomogeneous line [7,16].

Despite its connections to the McCall-Hahn case, and a wealth of data from many lambda experiments for guidance, the theory of two-pulse propagation [10–15] in lambda systems has many gaps. Although one link between theory and experiment in coupled-pulse propagation dynamics has been noted, in which observation [21] of predicted adiabaton pulses [22] was reported, no theoretical approach has been found, apart from purely numerical solution methods, that (a) deals nonperturbatively with the nonlinearities of the equations, (b) permits both pulses to change their shapes significantly while propagating,

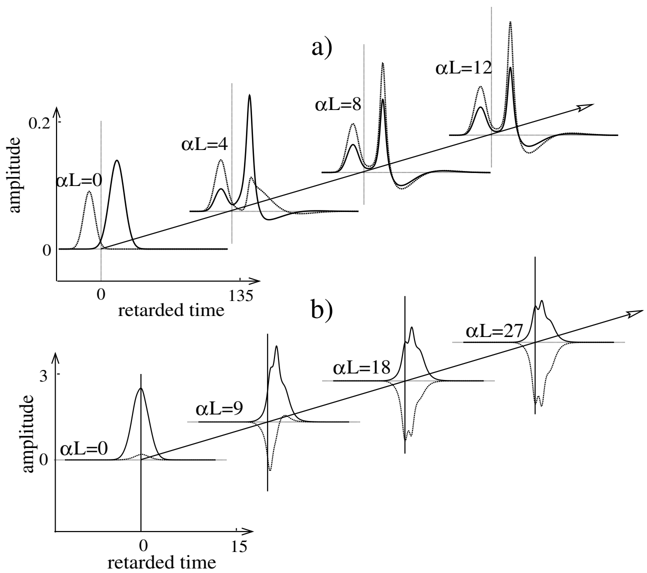

FIG. 3. Coupled evolution of a pair of pulses initially Gaussian in shape. Two-pulse propagation is shown, as governed by the five PDE's for c_a, c_g, $c_{g'}$ and Ω_g and $\Omega_{g'}$, obtained by exact numerical solution, for comparison with the predictions of the single dark area ODE (6). (a) Initial areas 3.1π and 1.4π, propagating through a lambda medium with $\phi = -2\pi/3$. At $\alpha z = 12$ the pulses are almost matched, with $\Omega_{g'}/\Omega_g \approx 1.6$, in good agreement with the final amplitude ratio predicted by the branch value of θ_D, i.e., $\tan(-\phi/2) = \tan(\pi/3) = \sqrt{3}$. (b) Pulses initially exactly matched with areas 6.2π and 0.5π, and $\phi = 2\pi/3$. Near-final matching is achieved at $\alpha z = 27$, with greatly altered pulse shape and with $\Omega_{g'}/\Omega_g \approx -1.6 \approx -\sqrt{3}$. In all numerical examples, $\alpha = \alpha'$, local time is in units of T^*, and Rabi frequencies in $1/T^*$. To be clear that the effects discussed here do not rely in some way on adiabatic following arising from the presence of the upper-state damping coefficient γ, the numerical integrations were made with $\gamma \equiv 0$.

(c) accounts for the sizable difference almost always found between the oscillator strengths of the atom's two optical transitions, and (d) allows both pulses to be shorter than the relaxation time of the absorptive transitions. The dark coherence wave equation (9) and the dark area theorem (6) address all four points (a)–(d) successfully.

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