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Coherent accumulation of excitation in the electromagnetically induced transparency of an ultrashort pulse train

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We investigate the excitation of an atomic degenerate-Λ system by a train of ultrashort pulses. An iterative analytic solution to the optical Bloch equations is found describing the temporal evolution of the density matrix elements. We show that electromagnetically induced transparency of the excitation pulses develops through the coherent accumulation of excitation (population and coherence) between the degenerate lower states. The influence of the pulse train parameters (area, repetition period, and phase between successive pulses) on establishing the transparency is investigated.

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I. INTRODUCTION

In electromagnetically induced transparency (EIT) [1], an atomic medium is made transparent to a resonant probe field by means of a coupling field tuned to a linked transition. The physical mechanism behind this induced transparency is coherent population trapping [2]: the two excitation fields create a dark superposition state, decoupled from the fields, to which the atomic population is transferred. EIT has found a variety of important applications, such as the production of slow and frozen light [3–5], giant Kerr nonlinearities [6], few-photon nonlinear optics [7], and picotesla magnetometers [8], among others. Most applications explore EIT in the steady-state regime.

But the transient behavior of EIT, which may be important for quantum-information storage, has been theoretically [9–11] and experimentally [12,13] investigated as well. It is known that the onset of EIT occurs on a time scale proportional to the square of the coupling field's Rabi frequency divided by the excited-state relaxation rate. And for a sudden switching on of a saturating cw coupling field, Rabi oscillations can occur before the system reaches the steady-state regime. These oscillations have been detected in the transmission signal of a cw probe field propagating through a sample of cold rubidium atoms in a magneto-optical trap [12,13].

EIT in the short [14,15] and ultrashort [16,17] pulsed regimes has also been investigated. And it was theoretically predicted by Kocharovskaya and Khanin [18] in 1986 that a train of ultrashort pulses may also induce coherent population trapping when interacting with a three-level Λ system. They showed, in the steady-state regime, that if the pulse repetition frequency is matched to a subharmonic of the frequency splitting of the lower atomic states, a sufficiently intense pulse may result in population trapping. More recently, Sautenkov and colleagues [17] observed EIT of an ultrashort pulse train produced by a mode-locked diode laser propagating through a sample of rubidium atomic vapor.

In this article, we investigate EIT of an ultrashort pulse train interacting with a degenerate-Λ system in the transient regime. The repetition period of the pulse train to be considered here is shorter than the excited-state lifetime. In this regime, we show that a dark superposition of the lower states results from the coherent accumulation of excitation in these states, leading to EIT of the pulse train. The results discussed here also apply to nondegenerate systems, such as those of Refs. [18,19], if one considers the restriction of having to match the pulse repetition rate to the frequency splitting of the lower levels.

In Sec. II of the article, we describe the atomic Λ system, present the optical Bloch equations that describe the interaction of the atomic system with an ultrashort pulse train, and introduce an analytic iterative solution to the Bloch equations. Sec. III contains a discussion of the temporal evolution of the atomic coherence between the lower states of the Λ system, as well as of the dark superposition of these lower states, as a function of the number of driving pulses in the excitation train. And Sec. IV concludes the article, summarizing the main results.

II. ATOMIC SYSTEM AND ITS EQUATIONS OF MOTION

The investigated atomic system is shown in Fig. 1. It consists of a closed, three-level, degenerate-Λ system interacting with the classical electric field $E(t)=E_0 f(t)\exp(-i\omega_L t)+\text{c.c.}$ of an ultrashort pulse with carrier frequency ω_L, real ampli-

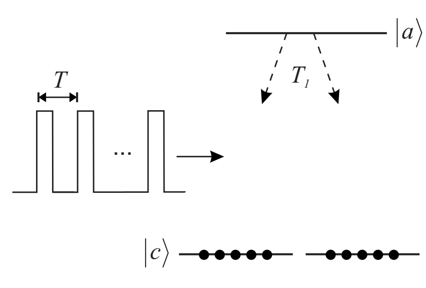

FIG. 1. The model atomic system: a degenerate-Λ system interacting with a train of ultrashort pulses of arbitrary shape and repetition period T. The atomic population is initially equally distributed between the lower b and c states, but there is no initial coherence between these states. The driving pulses couple both lower states equally to the excited state, which has a lifetime of T_1.

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tude E_0, and slowly varying envelope $f(t)$. The excited a state has a spontaneous lifetime of T_1 and decays to both lower b and c states at equal rates; we neglect any decay between b and c. We also assume that the dipole matrix elements d_{ij} between the lower and excited states are real and aligned parallel to the field polarization vector, and $d=d_{ba}=d_{ca}$.

The Hamiltonian describing the interaction of the atomic system with the ultrashort pulse is given by $\hat{H}=\hat{H}_0+\hat{H}_{int}+\hat{H}_{sp}$, where

$$\hat{H}_0 = \hbar\omega_0|a\rangle\langle a| \tag{1}$$

is the Hamiltonian of the free atom,

$$\hat{H}_{int} = -dE(t)(|b\rangle\langle a| + |c\rangle\langle a|) + \text{H.c.} \tag{2}$$

is the atom-field interaction Hamiltonian, and

$$\hat{H}_{sp} = -i(\hbar/T_1)|a\rangle\langle a| \tag{3}$$

is a term added phenomenologically to account for spontaneous decay from the excited state.

A. Optical Bloch equations

In the rotating-wave approximation, the optical Bloch equations describing the temporal evolution of the density matrix elements are then

$$\dot{\rho}_{cc} = (1/2T_1)\rho_{aa} + 0.5[i\alpha^*(t)\rho_{ca} + \text{c.c.}],$$

$$\dot{\rho}_{aa} = -(1/T_1)\rho_{aa} + 0.5[i\alpha(t)(\rho_{ab} + \rho_{ac}) + \text{c.c.}],$$

$$\dot{\rho}_{ab} = -(1/2T_1)\rho_{ab} - 0.5i\alpha^*(t)(1 + \rho_{cb} - \rho_{cc} - 2\rho_{aa}),$$

$$\dot{\rho}_{ac} = -(1/2T_1)\rho_{ac} + 0.5i\alpha^*(t)(\rho_{aa} - \rho_{cc} - \rho_{bc}),$$

$$\dot{\rho}_{bc} = -0.5i\alpha(t)\rho_{ac} + 0.5i\alpha^*(t)\rho_{ba}, \tag{4}$$

where $\alpha(t)=(2dE_0/\hbar)f(t)$ is a time-dependent Rabi frequency and "c.c." denotes the complex conjugate. The ultrashort pulse is tuned to the atomic resonance ($\omega_L=\omega_0$). The population in level b was eliminated from Eqs. (4) by the normalization condition $\rho_{aa}+\rho_{bb}+\rho_{cc}=1$.

When the atomic system is interacting with a train of identical ultrashort pulses, the slowly varying envelope of the field becomes

$$f_T(t) = \sum_{n=0}^{\infty} f(t - nT)e^{in\Delta\varphi}, \tag{5}$$

where T is the repetition period of the pulse train and $\Delta\varphi$ is the phase difference between two consecutive pulses [20]. The pulse width is assumed to be much shorter than the pulse repetition period such that the pulses are well separated in time.

We want to investigate the role of the pulse train parameters T, $\Delta\varphi$, and $\theta=\int_{-\infty}^{\infty}\alpha(t)dt$ (the pulse area) in establishing EIT. We are particularly interested in situations in which the pulse repetition period is shorter than the radiative lifetime of the excited state. This would be the case, for example, for excitation of the $5S_{1/2}\rightarrow 5P_{1/2}(D_1)$ transition in ^{87}Rb by a mode-locked Ti:sapphire laser. The lifetime of the $5P_{1/2}$ is $T_1=28$ ns, while typical repetition periods of Ti:sapphire lasers range from $T=1$ ns (1-GHz laser cavity) to $T=13$ ns (80-MHz cavity). For a 360-MHz laser cavity, about ten pulses can excite such an atomic system in the excited state's lifetime. Under such a condition, population and coherence can accumulate in the atomic system from pulse to pulse. Coherent accumulation of excitation has been previously studied in connection with the excitation of two-level [21] and three-level [22–24] cascade atoms, and significant differences from incoherent accumulation processes were observed. Here, the first excitation pulse finds the atom with population equally distributed between its two lower states $\rho_{bb}(0)=\rho_{cc}(0)=0.5$, but without any coherence: $\rho_{bc}(0)=0$. Since the atomic system will not have had enough time to completely relax before the next excitation pulse, each subsequent pulse will find the atom in a different arbitrary state induced by the previous pulse.

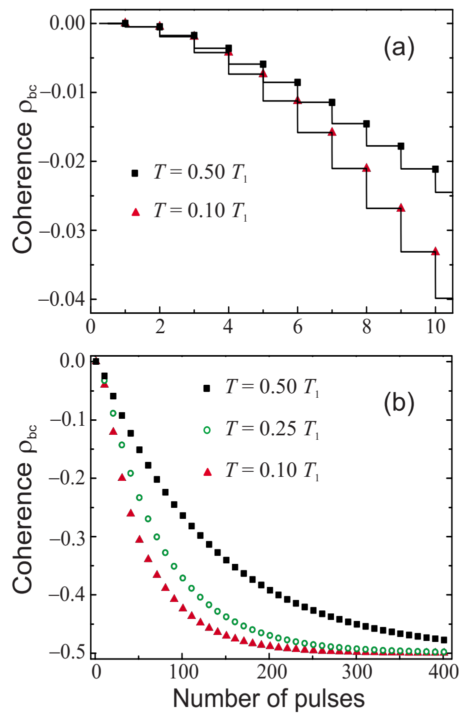

FIG. 2. (Color online) (a) The lower states' coherence term ρ_{bc} evaluated with the iterative solution (symbols) and the numerical solution (solid lines) of the optical Bloch equations for a small number of excitation pulses. The plot of the numerical solution actually corresponds to the evolution of ρ_{bc} as a function of time in units of the pulse repetition period T. (b) The coherence term ρ_{bc} as a function of the number of excitation pulses evaluated using the iterative solution. For clarity, the results are plotted for only one out of each ten excitation pulses. In both (a) and (b), $\theta=\pi/50$.

B. Analytic iterative solution

Although numerically solving the Bloch equations could provide a complete description of the temporal evolution of the atomic system excited by such an ultrashort pulse train, this procedure can easily become computationally intense for a large number of pulses. Recently, Felinto and colleagues [22] developed an analytic iterative procedure to study the excitation of a three-level, cascade atomic system by an ultrashort pulse train. We followed their approach and developed an iterative solution that allowed us to efficiently calculate the density matrix elements of our Λ system. The two basic assumptions made are that the pulses have a small area and excitation occurs in the quasi-impulsive regime. The former assumption means that each individual ultrashort pulse is capable of transferring only a small amount of population and coherence between the atomic levels. And the latter means that the temporal width of each of the individual pulses (hundreds of femtoseconds to picoseconds long) is orders of magnitude shorter than all atomic relaxation times (tens of nanoseconds long) and the laser repetition time (a few nanoseconds long). Therefore, the density matrix elements do not evolve during the short action of a single pulse.

We formally integrate Eqs. (4) and, then, perturbatively solve the integral equations by keeping only terms up to θ^2. Denoting $\rho_{ij}^{(n)}$ and $\rho_{ij}^{(n+1)}$ as the density matrix elements before excitation by the nth and $(n+1)$th pulses, respectively, we find

$$\rho_{cc}^{(n+1)} = \rho_{cc}^{(n)} + \frac{1}{2}\rho_{aa}^{(n)} + \frac{1}{2}[ie^{in\Delta\varphi}(\rho_{ab}^{(n)} - \rho_{ac}^{(n)}) + \text{c.c.}]\theta + \left[1 + \frac{3}{2}(\rho_{bc}^{(n)} + \rho_{cb}^{(n)}) - 4\rho_{aa}^{(n)} + \rho_{cc}^{(n)}\right]\theta^2$$
$$- \frac{1}{2}\{\rho_{aa}^{(n)} + [ie^{in\Delta\varphi}(\rho_{ab}^{(n)} + \rho_{ac}^{(n)}) + \text{c.c.}]\theta + 2[1 + \rho_{bc}^{(n)} + \rho_{cb}^{(n)} - 3\rho_{ba}^{(n)}]\theta^2\}e^{-T/T_1},$$

$$\rho_{aa}^{(n+1)} = \frac{1}{2}\{\rho_{aa}^{(n)} + [ie^{in\Delta\varphi}(\rho_{ab}^{(n)} + \rho_{ac}^{(n)}) + \text{c.c.}]\theta + 2[1 + \rho_{bc}^{(n)} + \rho_{cb}^{(n)} - 3\rho_{ba}^{(n)}]\theta^2\}e^{-T/T_1},$$

$$\rho_{ab}^{(n+1)} = \left\{\rho_{ab}^{(n)} + ie^{-in\Delta\varphi}[2\rho_{aa}^{(n)} + \rho_{cc}^{(n)} - \rho_{cb}^{(n)} - 1]\theta + \frac{1}{2}[3\rho_{ab}^{(n)} + \rho_{ca}^{(n)} - 2e^{-2in\Delta\varphi}(\rho_{ba}^{(n)} + \rho_{ca}^{(n)})]\theta^2\right\}e^{-T/2T_1},$$

$$\rho_{ac}^{(n+1)} = \left\{\rho_{ac}^{(n)} + \frac{1}{2}ie^{-in\Delta\varphi}[\rho_{aa}^{(n)} - \rho_{cc}^{(n)} - \rho_{bc}^{(n)}]\theta + \frac{1}{8}[2e^{-2in\Delta\varphi}(\rho_{ba}^{(n)} + \rho_{ca}^{(n)}) - (\rho_{ab}^{(n)} + 3\rho_{ac}^{(n)})]\theta^2\right\}e^{-T/2T_1},$$

$$\rho_{bc}^{(n+1)} = \rho_{bc}^{(n)} + \frac{1}{2}i[e^{-in\Delta\varphi}\rho_{ba}^{(n)} - e^{in\Delta\varphi}\rho_{ca}^{(n)}]\theta + \frac{1}{8}[3\rho_{aa}^{(n)} - 2\rho_{bc}^{(n)} - 1]\theta^2. \tag{6}$$

Equations (6) consist of an iterative solution for the density matrix elements. The atomic state just before the $(n+1)$th pulse is determined as a function of the atomic state before the nth pulse. The iterative solution depends on the pulse area θ, but not on the pulse shape. It is also dependent on the phase difference $\Delta\varphi$ between pulses, as well as on the pulse repetition period T. Successive application of the above set of equations to an initial state provides the temporal evolution of the atomic system evaluated at time intervals of the pulse repetition period.

III. COHERENT ACCUMULATION OF EXCITATION

A. Atomic coherence evolution

To check the validity of the iterative solution, we compared it to the numerical integration of Eqs. (4) for a small number of pulses. We considered a Gaussian test pulse $\alpha(t) = (\theta/\tau)\exp(-\pi t^2/\tau^2)$, where $\tau=0.001T$ is the pulse width and $\theta=\pi/50$. The phase difference between consecutive pulses $\Delta\varphi$ was arbitrarily set to zero. Figure 2(a) shows the iterative and numerical solutions evaluated for two different pulse repetition periods ($T=0.50T_1$ and $T=0.10T_1$) and same pulse area. For $T=0.50T_1$, two pulses excite the atomic system during the excited state's lifetime, while for $T=0.10T_1$, the system is excited by ten pulses in that same lifetime. While the iterative solution evaluates the coherence ρ_{bc} between the lower atomic states at discrete time instants, the numerical solution yields the full temporal evolution of the atomic coherence. We see an excellent agreement between the iterative and numerical solutions. Due to its small area, each individual pulse excites only a small coherence ρ_{bc} between the degenerate lower states. But at every new pulse that excites the atom, this coherence builds up. Extending the iterative solution over excitation by a much larger number of pulses (longer time scales), we see in Fig. 2(b) that the atomic system eventually reaches a stationary state of full coherence: $|\rho_{bc}|=0.5$. The coherence accumulation depends on the pulse repetition period: the shorter the repetition period T, the faster the accumulation since the atomic system will have less time to relax between pulses.

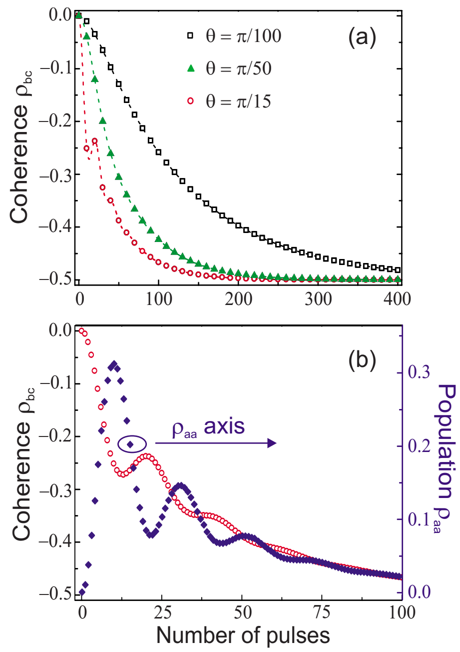

FIG. 3. (Color online) (a) The coherence between the lower atomic states as a function of the number of excitation pulses evaluated using the iterative solution for different pulse areas. For clarity, we show only the results for one out of each ten driving pulses; the dashed lines are a visual guide. (b) The atomic coherence ρ_{bc} and population ρ_{aa} in the excited state for a smaller number of pulses and $\theta=\pi/15$. In both figures, $T=0.10T_1$.

For a fixed period $T=0.1T_1$, we investigated the effect of pulse area on the coherent accumulation process. Figure 3(a) shows the dynamics of the lower states' coherence evolution evaluated for three different pulse areas. If the system is excited by a large enough number of pulses, the lower levels will eventually reach full coherence, even for a pulse area as small as $\theta=\pi/100$. The pulse area is not determinant in establishing a coherence between the lower states [25], but rather of how fast this coherence develops. The larger the pulse area is, the faster is the coherent accumulation effect leading to full coherence. Figure 3(b) shows the coherence ρ_{bc} and the population ρ_{aa} in the excited state over a short time scale for $\theta=\pi/15$. Initially, population in the excited state tends to grow with the number of excitation pulses reaching up to 32% of the atomic population. But after excitation by a large number of pulses, population in state a decreases, tending to zero. It is possible to observe oscillations in ρ_{bc} and ρ_{aa} occurring in the accumulation process associated with Rabi oscillations of population between the lower and excited states. These oscillations are of a different nature of the ones usually associated with excitation by cw fields and large-area single pulses [26]. Each individual pulse here cannot induce Rabi oscillations by itself due to its small area. Furthermore, the time separation between adjacent pulses is three orders of magnitude greater than their temporal widths, so the driving pulses do not overlap. The Rabi oscillations shown in Fig. 3(b) are a result of the combined effect of excitation by many pulses and the coherent accumulation of excitation between these pulses.

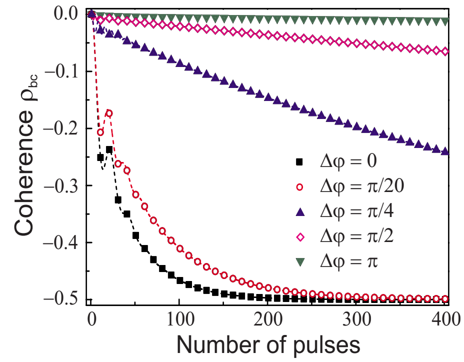

FIG. 4. (Color online) The lower states' coherence ρ_{bc} as a function of the number of excitation pulses for various pulse phase difference $\Delta\varphi$. Results are plotted for only one out of each ten pulses, and the dashed lines are a visual guide. We took $T=0.10T_1$ and $\theta=\pi/15$.

If the phase difference $\Delta\varphi$ between successive pulses is not zero or an integer multiple of 2π, the dynamics of accumulation may be significantly modified. Even if $\Delta\varphi$ is small, after a large number n of pulses, the nth pulse may have a phase very different from that of the first excitation pulse. In Fig. 4 we show how the coherence accumulation depends on $\Delta\varphi$. For $\Delta\varphi=0$ (or $2\pi k$, where k is an integer), a stationary state of full atomic coherence is achieved after excitation by about 300 pulses. But even for a phase difference as small as $\Delta\varphi=\pi/20$, the dynamics towards full coherence is already modified. For a given number of excitation pulses, as the phase difference increases, the final ground-state coherence decreases. For $\Delta\varphi=\pi$, the atomic system is barely excited and no coherence is developed between the lower atomic states. In this case, since the atomic system does not have enough time to relax between pulses, each pair of consecutive pulses effectively acts as a "zero-area pulse." These results indicate that in order to achieve full coherence between the lower states with the smallest possible number of excitation pulses, the pulse train must be stabilized to a phase difference of zero or an integer multiple of 2π.

B. Bright- and dark-state evolution

With full coherence established between the lower states, absorption of the excitation pulses should cease. According to Sautenkov *et al.* [19], a dark state—formed by a linear superposition of the lower states—is created that is decoupled from the excitation pulses. A bright state, which interacts with the excitation pulses, is also created. Population in the bright state is transferred to the dark state where it remains trapped. When all the atomic population is in the dark state, the excitation pulses are no longer absorbed and one observes an electromagnetically induced transparency of these pulses.

The bright and dark states are defined as

$$|B\rangle = \frac{1}{\sqrt{2}}(|b\rangle + |c\rangle),$$

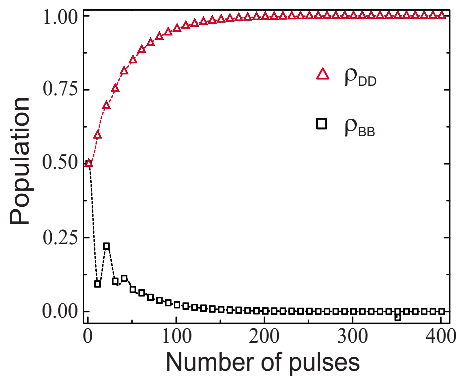

FIG. 5. (Color online) Population in the bright (ρ_{BB}) and dark (ρ_{DD}) states plotted as a function of the number of driving pulses. For clarity, we plot the results for only one out of each ten driving pulses, and the dashed lines are a visual guide. Here, $\Delta\varphi=0$, $T=0.10T_1$, and $\theta=\pi/15$.

$$|D\rangle = \frac{1}{\sqrt{2}}(|b\rangle - |c\rangle), \tag{7}$$

for equal transition strengths between the lower and excited states. Population in the bright and dark states is then $\rho_{BB}=0.5\rho_{bb}+0.5\rho_{cc}+\text{Re}(\rho_{bc})$ and $\rho_{DD}=0.5\rho_{bb}+0.5\rho_{cc}-\text{Re}(\rho_{bc})$, respectively. With the iterative set of solutions of Eqs. (6), it is straightforward to evaluate the temporal evolution of ρ_{BB} and ρ_{DD}.

Figure 5 shows the population in the bright and dark states as a function of the number of pulses exciting the atomic system. Here, we took $\Delta\varphi=0$, $T=0.1T_1$, and $\theta=\pi/15$. Both dressed states start out equally populated, but as the number of driving pulses grows, population is optically pumped from the bright to the dark state. Each individual pulse transfers a small amount of population between those two states, but an accumulation effect between successive pulses results in a complete population transfer to the dark state. Rabi oscillations between the bright and excited states are seen. Since the dark state is decoupled from the driving fields, no oscillations occur during its evolution. Simultaneously, the population in the excited state goes to zero, as shown in Fig. 3(b). The ultrashort pulses are then no longer absorbed, and electromagnetically induced transparency takes place.

IV. CONCLUSIONS

To conclude, we studied the occurrence of electromagnetically induced transparency in the excitation of a degenerate-Λ system by a train of ultrashort pulses. We showed that the coherent accumulation of excitation (population and coherence) between the lower states of the atom plays a significant role in establishing EIT. Each driving pulse creates a small coherence between the lower states that accumulates between successive pulses until a steady state of full coherence is reached and absorption of the pulse train is canceled. An ultrashort pulse train can be an effective tool to experimentally study transient EIT. Coherent accumulation of population in a two-photon transition in atomic rubidium was recently experimentally observed by Stowe and colleagues [24]. The experimental study of the formation of dark states and the onset of coherent population trapping by coherent accumulation of excitation should be equally possible by monitoring the excited state's fluorescence as a function of the number of excitation pulses, for example. In future work, we will use the iterative solution developed here to study the propagation of an ultrashort pulse train along an extended atomic collection under electromagnetically induced transparency.

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