

Transient coherent effects in two-level systems

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The topic of atomic coherence is reviewed. Conditions necessary for observing coherence are discussed. The McCall-Hahn area theorem is addressed and the transient coherent effects of self-induced transparency, free induction decay, photon echo, and angular momentum orientation are described. Recent applications of these effects are cited.

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I. INTRODUCTION

The recent availability of high intensity, ultrashort (< 200 fs) pulses has led to a large number of experimental investigations of transient optical coherence effects. Transient coherent effects are now reproducible enough to be used as tools for measuring relaxation rates, molecular structure, and chemical reaction dynamics. This paper reviews the topic of coherence as it relates to atom-field interactions, outlines the conditions necessary for observing coherence, describes the most common transient coherent effects, and mentions some of their recent applications.

II. COHERENCE

The term coherence is normally associated with quantities that exhibit oscillatory behavior. In this paper, two or more objects are coherent if their phase of oscillation can be related in a traceable, non-random way. As examples, consider optical temporal and optical spatial coherence. Optical temporal coherence exists if, at the same position, the phase of a passing electric field at two different times can be related. Optical spatial coherence exists if, at the same time, the phase of the electric field at two different positions can be related. The oscillating quantities do not have to be identical—coherence can exist between an atom and an electric field. In this case the electric field drives the atom's electrons and creates an oscillating dipole moment. Coherence between the field and dipole moment implies that the phase of the dipole moment is related to the phase of the electric field. Atomic coherence occurs when the dipole moment of one atom is related to the dipole moment of another atom as when a group of atoms is exposed to the same electric field [1]. Atomic coherence is the source of the effects described in this paper.

The dipole moment induced by an electric field plays a key role in the mathematical description of atomic coherence. The interaction between the field and an individual atom, as described by the dipole moment, can be calculated quantum mechanically. Assuming the response of the single atom represents the response of an entire group, one can use the quantum mechanical dipole moment to obtain an expression for the macroscopic polarization density. The polarization density acts as a source term in the Maxwell wave equation, contributing back to the electric field that originally induced the dipole moment [2].

The quantum expectation value of the dipole moment is given by

$$\langle \boldsymbol{\mu} \rangle = \langle \Psi | e \mathbf{x} | \Psi \rangle = \int \Psi e \mathbf{x} \Psi d^3x, \quad (1)$$

... where $\boldsymbol{\mu} = e \mathbf{x}$ is the classical value of the dipole moment and Ψ is the state vector describing the molecular system while under the influence of the external electric field. The state vector must be a solution to the Schrödinger equation given by

$$i\hbar \frac{\partial \Psi}{\partial t} = H \Psi. \quad (2)$$

In Eq. (2), H is the system Hamiltonian given by $H = H_0 + H_\mu$. H_0 describes the internal energy of the isolated molecule and H_μ describes the interaction energy between the molecule and the external field: $H_\mu = -\boldsymbol{\mu} \cdot \mathbf{E}$. The coherence lies in the $\boldsymbol{\mu} \cdot \mathbf{E}$ expression where the phase and direction of $\boldsymbol{\mu}$ are assumed to remain linked to the phase and polarization direction of \mathbf{E} . Coherence is also assumed when the same $\boldsymbol{\mu}$ is taken to represent all atoms in the group. Under these conditions, a non-zero dipole moment density \mathbf{P} is created which contributes energy back to the incident electric field.

\mathbf{E} is assumed to be weak compared with the internal fields that bind the molecule together such that $|\boldsymbol{\mu} \cdot \mathbf{E}| \ll \hbar\omega_0$ where $\hbar\omega_0$ characterizes the transition energy between two H_0 eigenstates. Because the contribution of H_μ to the energy of the system is small compared with that of H_0 , a solution to Eq. (2) can be composed from a time-dependent superposition of the stationary eigenstates associated with H_0 . The assumed state vector for the system is given by

$$\Psi = c_1(\mathbf{t}) \varphi_1(\mathbf{r}) + c_2(\mathbf{t}) \varphi_2(\mathbf{r}) + \dots + c_n(\mathbf{t}) \varphi_n(\mathbf{r}), \quad (3)$$

where $\phi_n(r)$ are the time-independent eigenstates of H_0 and $c_n(t)$ are coefficients whose square magnitudes represent the probability of the system being in state n . The c -coefficients can also be interpreted as representing the probability of transitions between states. For example, if a molecule is initially in state $n = 1$, $|c_2(t)|^2$ represents the probability of a transition from $n = 1$ to $n = 2$.

Substituting Eq. (3) into Eq. (1), one can find an expression for $\langle \mu \rangle$ in terms of the c -coefficients. The same arrangement of c -coefficients that describes the dipole moment also appears in the elements of the system's density matrix. The connection between the density matrix and the dipole moment permits one to use the density matrix in place of the dipole moment in calculations. By using the density matrix, one can introduce incoherent relaxation mechanisms (like collisions) that cannot be practically incorporated into Eq. (2) [3].

III. OBSERVING COHERENCE

Observing a coherent interaction in the laboratory requires considerable effort as the electric field must meet several stringent criteria. The two most important conditions involve the field's temporal and spatial character. The simplest solution to the equations in section II assumes a cw or constant amplitude electric field. The constant field creates a population inversion that oscillates between 0 and 100 percent at the well known Rabi frequency $\Omega_R = \mu E/\hbar$. If, instead of a cw field, the field is pulsed, Ω_R is a function of time given by $\Omega_R(t) = \mu E(t)/\hbar$. Unlike cw electric fields that are characterized by a Rabi frequency, pulsed fields are better described by a pulse area A . The pulse area is the time integral of $\Omega_R(t)$:

$$A(z) = \int \Omega_R(t') dt' = \frac{\mu}{\hbar} \int E(z, t') dt'. \quad (4)$$

To observe coherent effects, the observation time for a cw field or the pulse duration for a pulsed field must be shorter than the time constant of any decay mechanisms. In addition, the experimental field must not vary too much from the plane wave field assumed in theory. When the field has a non-uniform spatial profile, Ω_R or A may vary widely across the field's cross section, making a clear observation of coherence nearly impossible.

Besides arranging a suitable electric field one must also prevent noisy processes from destroying the coherence. The two chief incoherent processes are radiative decay and collisions. Spontaneous radiative transitions disrupt coherence because their contribution to the electric field is random in direction, phase,

and frequency. To arrange an observable coherent interaction, one must ensure that the interaction time τ between the field and the atoms is shorter than the radiative decay constant T_1 . For transitions between metastable states, this condition is easily met.

Preventing collisions is more difficult. Collisions can reorient an atom's alignment with the electric field or change the atom's velocity so that the atom is Doppler shifted out of resonance with the field. Collisions interrupt the phase of the atom's dipole moment so that μ is no longer in step with \mathbf{E} or the dipole moments of the atoms around it. Collisions can remove the atom from the interaction by promoting it to a different state. Of the collision types listed, phase interrupting collisions are the most prevalent. To observe coherence when collisions are present, τ must be shorter than the mean time between collisions T_2' . This mean time can be estimated from the inverse of the homogeneous linewidth $\Delta\nu_H$ such that $T_2' = 1/\Delta\nu_H$ [4]. $\Delta\nu_H$ is a half width at half maximum (HWHM) value.

Even without radiative losses and collisions, a coherent interaction may be unobservable because of destructive interference. Interference between two states results in a sinusoidal oscillation in the population inversion much like the interference between two sinusoidal signals results in a beat pattern that is itself sinusoidal. Any additional state, even a degenerate one, brings with it its own dipole moment that also contributes to the interference. The inclusion of a third state reduces the amplitude of the state population oscillations and disrupts the once pure sinusoidal character of the oscillations. As even more states are added the coherent interaction becomes indistinguishable from an incoherent one [5]. In certain cases, a degenerate system can be reduced to an ideal two-state system. The R(0) or P(1) transitions between the rotational and vibrational levels of a diatomic molecule in its electronic ground state are one such case. By using a circularly polarized field, the Δm_j selection rule (+1 for one circular polarization, -1 for the other) bypasses the degeneracy and changes what could be a four-state system back into a two-state system.

The two-state limit is not as restrictive as one might at first think since the word "state" does not necessarily mean bound state. A recent paper has predicted coherent oscillations between dissociative molecular states [6]. An initial laser pulse excites molecules into a low-energy dissociative state. A second pulse then promotes molecules from the low-energy state into a higher-energy dissociative state. Coherence between the two dissociative states causes upper state number density to oscillate as a function of the second pulse's energy. Because the upper and lower levels of the system lie in repulsive electrical potentials, the energy gap between the states changes with time as the atoms that compose the molecule begin to separate. This requires the duration of the second pulse to be extremely short—10 to

20 fs—so that significant separation does not occur while the pulse is present. The states in a coherent interaction need not even be those of a conventional atom or molecule: Rabi oscillations have been directly observed between exciton states in a semiconductor [7] as has exciton self-induced transparency [8].

Even given an ideal two-state system, destructive interference can still make coherence unobservable. If the electric field spectrum is broad enough to excite atoms across a transition's entire bandwidth, the interference caused by the continuum of dipole oscillation frequencies will become completely destructive in a time T_2^* . The value of T_2^* , also called the inhomogeneous dephasing time, can be estimated from the inverse of the HWHM inhomogeneous linewidth $\Delta\nu_1$ such that $T_2^* = \sqrt{\ln(2)} / \Delta\nu_1$. Conditions for observing coherent effects are optimal when $\tau < T_1, T_2', T_2^*$ (sharp line limit) and when $T_2^* < \tau < T_1, T_2'$ (broad line limit). The terms sharp and broad refer to the transition's spectral linewidth compared with the spectral linewidth of the field. In the sharp line limit, the electric field interacts with all portions of the absorption linewidth, but because τ is much less than T_2^* , the coherent interaction is finished long before destructive interference sets in. In the broad line limit, the field interacts with only a fraction of the absorption linewidth associated with T_2^* so that τ and the inhomogeneous dephasing time of the atoms responding to the field are similar.

IV. TRANSIENT COHERENT EFFECTS

The coherence effects associated with a pulsed electric field are more numerous and more diverse than the simple Rabi flopping associated with a cw field. Given a pulsed system, the simplest observable coherent effect is the oscillation of the final excited state population with pulse area. This oscillation can be described with the area theorem. Other coherent effects include self induced transparency, free induction decay, photon echoes, and orientation.

A. Area Theorem

In the late 1960s, S. L. McCall and E. L. Hahn solved the Schrödinger and Maxwell wave equations simultaneously for a non-degenerate, decay-free, two-level system and derived and experimentally verified what is known as the McCall-Hahn area theorem [9][10]:

$$\frac{dA}{dz} = -\frac{\alpha}{2} \sin A, \quad (5)$$

where A is the pulse area defined in Eq. (4), and α is the inverse Beer's absorption length. The area theorem shows that for a pulse with an area less than π , the change in area with distance is always negative and that the medium will absorb the pulse. For a pulse whose area is greater than π , the change in area with distance is positive so that A increases until it reaches a value of 2π . When $A = 2\pi$, the area no longer changes with distance suggesting absorption free propagation. Beyond 2π , the pattern repeats with 3π acting as the next threshold between decreasing and increasing area.

The oscillation in absorbed energy with pulse area shows that in any regime where coherence plays a role, greater pulse energy or longer exposure to the pump field does not necessarily mean greater absorption. An illustration of this can be found in a recently proposed experiment [11] whose goal is to produce a complete inversion in an excited electronic state. A positively chirped laser pulse pumps molecules out of the ground electronic state and into the repulsive potential region of an excited electronic state (Franck-Condon principle). As the repulsive potential forces the atoms apart, the energy gap between the excited and ground states shrinks. Simultaneously the photon energy associated with the positively chirped pulse increases. The short resonant interaction time needed to create the inversion is achieved over the limited overlap time between the pulse and energy gap spectrums. Calculations show that without the chirp, the interaction time is too long, resulting in a less than complete inversion.

McCall and Hahn showed that a self-consistent solution to the combined Maxwell-Schrödinger equations and the area theorem exists only for electric fields with a pulse envelope given by

$$\mathbf{E} = \mathbf{E}_0 \operatorname{sech}\left[(t - z/v) / \tau\right], \quad (6)$$

where v is speed of the pulse through the medium. Pulses with envelope functions other than the one described by Eq. (6) were shown to undergo reshaping through the coherent interaction with the medium so that the steady state pulse shape was always consistent with Eq. (6). However, experiments with high intensity, ultra short pulses showed that reshaping did not occur, calling into question the validity of the area theorem. Recent simulations [4] have reconfirmed the accuracy of the area theorem by showing that reshaping takes place through the mechanism of free induction decay (described later). The area theorem does break down when the pulse duration is so short that the slowly varying envelope approximation, upon which the area theorem is based, no longer holds. In this case, Rabi flopping occurs at the carrier wave frequency and high frequency components not predicted by the area theorem are added to the transmitted pulse [12].

B. Self induced transparency

Self-induced transparency (SIT) refers to an $A = 2\pi$ pulse's ability to travel through a medium without being absorbed. SIT was the first demonstration of atomic coherence at optical frequencies [8]. For a conceptual understanding of SIT considers the 2π pulse as being made up of two π pulses. The medium absorbs the first half of the 2π pulse while the second half of the pulse extracts it. SIT can occur in a medium with decay mechanisms provided the energy exchange takes place faster than the T_1 , T_2' , and T_2^* time constants. Some decay inevitably occurs, and the pulse loses energy. To maintain the 2π area while losing energy, the pulse broadens in time. Eventually the pulse temporal width becomes comparable to the incoherent decay time at which point the pulse is rapidly absorbed [13]. The distance traveled before absorption sets in, called the critical length, is given by

$$Z_B = \frac{3T_B}{\alpha \tau}, \quad (7)$$

where Z_B is the critical length and T_B is a characteristic time given by $1/T_B = 1/T_1 + 2/T_2'$.

A characteristic feature of SIT that distinguishes it from the more common saturation transparency is the substantial slowing of the pulse's propagation speed through the medium. The continuous cycling of the energy between the medium and the field during SIT can reduce the phase velocity by a factor of 100. This is not so, however, for SIT in the sharp line limit. In the sharp line limit, a large portion of the field energy lies outside the absorber's bandwidth so that most of the photons pass undisturbed through the medium at the normal c/n speed [13].

An interesting application of SIT is its potential use as a passive mode locking mechanism [14]. When a π pulse passes through an excited gain medium, it induces a 100% depletion of the inversion. (This is consistent with conceptual description of SIT given earlier.) The cross-sectional dimension of the pulse must then be adjusted so that when it reaches the passive mode locker—an unpumped section of the gain medium—its new area is 2π . The 2π pulse passes unabsorbed through the passive medium while the medium attenuates all other field configurations.

C. Free induction decay

When a laser pulse creates an incoherent excited state, the excited atoms radiate energy away through spontaneous emission (fluorescence). The fluorescence is randomly polarized and emitted in all directions. When a laser pulse creates a coherent excited state, the excited atoms still radiate but they do

so in phase and in the same direction. Because this fluorescent signal is coherent, destructive interference rapidly sets in, causing the fluorescence to disappear much faster than the fluorescence from an incoherently excited state. The rapid decay of the coherent signal is called free induction decay (FID). FID is the result of the finite bandwidth over which oscillation occurs and is linked to the inhomogeneous dephasing constant T_2^* . The addition of a FID signal to the transmitted high intensity, ultra-short pulses mentioned in section A allowed the pulses to meet the requirements of the area theorem despite the absence of pulse reshaping. FID can be practically used to gain insight into the dynamics of molecules. One group of researchers used a 50 fs pulse at 3 μm to pump two vibrational modes in solute water molecules. The combined FID signal from the two modes showed quantum beating, allowing the researchers to obtain structural and dynamical information [15].

D. Photon echoes

Photon echoes are the product of at least two pump pulses. The initial pump pulse, usually with a $\pi/2$ area, creates an excited state population. The excited state begins re-radiating the absorbed energy, but because of FID, the radiation stops almost immediately. The energy remaining in the upper state is essentially trapped because the destructive interference caused by inhomogeneous dephasing prevents the energy from being released. This is analogous to the trapping and guiding of energy in a phased array radar. An individual element of the radar, if isolated, radiates its energy in all directions. When placed in a group of radar elements oscillating coherently but with different frequencies and phases, interference channels the energy of each element into a single beam that sweeps across the horizon. A second sweep occurs because the discrete frequencies at which the elements oscillate eventually return to a common phase. For coherently excited atoms, radiation occurs across a continuous bandwidth. A continuum of frequencies initially starting in phase will take an infinite amount of time to come back into phase. Assuming no incoherent dephasing (collisions), the only way to access the trapped energy is to reverse the direction of dephasing. This can be accomplished with a second pump pulse that has π area. The π pulse reverses the dephasing direction so that the oscillating excited state atoms return to their initially common phase. The result is a third pulse called the echo pulse [16]. The delay time τ_{12} between the first and second pump pulses determines when the echo pulse appears. When incoherent decay mechanisms are present, a long τ_{12} results in a weak echo pulse since decay processes remove energy from the excited state. Pulse echo measurements then become a tool for determining incoherent relaxation rates [17]

E. Orientation

While coherence determines the final excited state population created by a pump pulse, it does not affect the rate at which atoms or molecules leave the excited state once the pulse has passed. Coherence will however affect the *measurement* of such a rate. As an example, consider pump pulse passing through a diatomic gas. The pulse encounters ground state molecules whose rotational angular momentum vector (\mathbf{J}) is distributed uniformly in all directions. Assuming the pump pulse is linearly polarized, it will preferentially interact with those molecules whose bond axis lies in the plane of polarization. If the absorption process increases the magnitude of \mathbf{J} without significantly altering its direction, the excited state will be composed of molecules whose \mathbf{J} vectors lie along the same axis. Without collisions, the excited molecules will remain in an aligned configuration. If the absorption of a probe beam is used to assess the excited state population, the measured absorption will depend on how well the probe beam's polarization is aligned with preferred orientation of the excited state molecules. The change of probe beam absorption with time caused by collisions allows one to separate elastic from inelastic relaxation rates [5].

V. CONCLUSION

Coherent effects are becoming more common and more visible as access to lasers with pulse durations shorter than typical collisional relaxation times increases. Transient coherent effects such as SIT, FID, photon echo, and orientation are now common enough to be used as laboratory tools for examining rapid dynamical processes. A familiarity with atomic coherence and the conditions under which it appears will therefore prove highly useful to the contemporary experimentalist.

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