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Synchronization of Atomic Quantum Transitions by Light Pulses

V.P. Chebotayev, V.A. Ulybin

Institute of Thermophysics, Siberian Branch, Academy of Sciences USSR
SU-630090 Novosibirsk-90, USSR

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Abstract. The synchronization of atomic quantum transitions with natural Raman oscillations by ultrashort light pulses has been investigated. This phenomenon may be observed if the duration of the perturbation pulse is less than the period of oscillations of the forbidden atomic transitions. The accuracy of the direct measurements of the quantum transition times for trapped particles can be of the same order as the ratio of the two-photon transition frequency to the homogeneous line width.

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Frequency measurements in atoms and molecules are currently made with an accuracy 10^{10}-10^{14} by using methods of super-high-resolution laser spectroscopy such as saturated absorption, Doppler-free two-photon absorption and other techniques. The known methods are based on resonant interaction between particles and an optical field, hence a highly monochromatic laser radiation is essential in these methods. The observation of resonances is carried out for times which are much longer then the oscillation periods of the quantum transitions under investigation.

A new spectroscopic method based on using pairs of pulses of radiation with a linewidth which is broader than the measured quantum-transition frequency interval, but with a stable interpulse time has been considered in [1]. The duration of the interaction between a particle and the single pulse is shorter than the oscillation period of the quantum transition. Hence, the result of the atomic interaction with two short pulses is defined by the phase of the free oscillations of a dipole moment at the time of arrival of the second pulse. In other words, the particle makes the transition from one energy level into the other one synchronously with the atomic oscillations. The synchronized quantum transitions are very accurately determined in time, and may be applied to direct precision measurements of time and frequency.

1. Qualitative Analysis

The phenomenon of synchronization of quantum transitions is not critically dependent on the nature of the pulse's perturbation. Hence, we shall explain it by a simple example of an atom interacting with two DC pulses of an electric field. We write the latter in the form

$$E(t) = Eg(t) + E'g(t-T)\,, \tag{1}$$

where g(t) is the pulse shape, and T is the interpulse time. The field-induced dipole moment of a 2-level atom may be written in the form

$$d(t) = a_{21} d_{21}^{*} \exp(-i\omega_{21} t) + \text{c.c.}\,, \tag{2}$$

where a_{21}, d_{21} and ω_{21} are the probability amplitude, the dipole moment and the frequency of the transition $|1\rangle \rightarrow |2\rangle$, respectively (the atom is considered to be in the state $|1\rangle$ when $t = -\infty$). Both d(t) and d_{21} are assumed to be projections onto the

field direction. According to [2] we have

$$a_{21} = (i/\hbar)d_{21}\int_{-\infty}^{t} dt' E(t')\exp(i\omega_{21}t') . \quad (3)$$

If the duration τ of the atomic interaction with a single pulse is much shorter than the period of atomic oscillations $2\pi/\omega_{21}$, then the function g(t) in (1) and (3) may be replaced by $\tau\delta(t)$, where $\delta(t)$ is Dirac's delta-function. It means that we probe the atomic dipole moment at times t = 0 and t = T. For t > T we find from (1-3) the dipole moment as the superposition of dipole moments excited at t = 0 and t = T

$$d(t) = id_{21}^{*}\tau\Omega_{21}\exp(-i\omega_{21}t) \times[1 + (E'/E)\exp(i\omega_{21}T)] + c.c. , \quad (4)$$

where $\Omega_{21} = Ed_{21}/\hbar$, and the probability of the transition $|1\rangle\rightarrow|2\rangle$ is

$$|a_{21}|^2 = (\tau|\Omega_{21}|)^2 [1 + (E'/E)^2 + \cos\omega_{21}T] . \quad (5)$$

As we see from (4 and 5), both d(t) and $|a_{21}|^2$ have maxima at times T that are integer multiples of the period $2\pi/\omega_{21}$. Thus, when an atom interacts with two short pulses, the quantum transition $|1\rangle \rightarrow |2\rangle$ may be synchronized with its natural oscillations. The effect is also shown to be very distinctive with equal pulse amplitudes (E'=E).

Instead of DC pulses of electric or magnetic fields, ac pulses may be used as well. The case of ac pulses is of interest for optical transitions. It is clear that a carrier frequency of ac pulses may take any value satisfying the inequality $\omega >> \tau^{-1}$, ω_{21}. There are only two pulse parameters of principal importance, i.e. the duration of a perturbation pulse, which should be shorter than an atomic-oscillation period, and the interpulse time, which should be stable and a multiple of the period.

The aim of the work is to consider the phenomenon of synchronization of quantum transitions by light pulses. At present, advanced methods of ultrashort laser pulse generation provide the possibility to obtain light pulses of 10-100 fs duraction [3]. This allows one to carry out experiments for synchronization of IR and FIR quantum transitions and direct measurements of time with an accuracy of the order 10^{12}-10^{13}. As the light frequency $\omega >> \omega_{21}$, the excitation of the quantum transition is realized by the two-photon Raman process. We start the consideration with an analysis of the Raman interaction between an atom and a single light pulse of any intensity. The analysis is of interest in its own right.

2. Interaction Between an Atom and Ultrashort Light Pulses

Let $|1\rangle$ and $|2\rangle$ be the ground and metastable atomic states obeying the selection rules for two-photon transitions. We consider the interaction between the atom and the light pulse

$$E(t) = Eg(t)\exp(-i\omega t) + c.c. ,$$

where 2E is the amplitude of the electromagnetic wave pulse. The frequency ω is nonresonant with respect to the intermediate transitions $|1\rangle\rightarrow|\alpha\rangle$ and $|2\rangle\rightarrow|\alpha\rangle$. The duration of the pulse is of the form

$$\tau = \int_{-\infty}^{\infty} dt|g(t)|^2$$

and obeys $\omega^{-1} << \tau \leq \omega_{21}^{-1}$. We assume that the light pulse is of a symmetric shape g(-t) = g(t).

The equations for density matrix elements which describe the stimulated Raman scattering in the field E(t) where reduced similarly to the equations for a two-level atom in an effective nonoscillating field [4]

$$\left\{\frac{d}{dt} + i[\omega_{21} - |g(t)|^2\Delta] + \Gamma\right\}\rho_{21} = \tfrac{1}{2}i\Omega|g(t)|^2(\rho_{11} - \rho_{22}) , \quad (6)$$

$$(d/dt + 2\Gamma)\rho_{22} = -\tfrac{1}{2}i\,\Omega|g(t)|^2\rho_{21} + c.c. ,$$

$$\rho_{11} + \rho_{22} = 1 ,$$

where 2Γ is the spontaneous decay rate of the upper state $|2\rangle$, $\Delta = E^2(D_{22} - D_{11})$ is the difference of the optical Stark shifts of the levels $|2\rangle$ and $|1\rangle$, $\Omega = 2E^2D_{21}$ is the effective (two-photon) Rabi frequency, $D_{ik} = D_{ik}(\omega)+D_{ik}(-\omega)$, $D_{ik}(\omega) = \Sigma_\alpha d_{i\alpha}d_{\alpha k}\hbar^{-2}(\omega_{\alpha 1} - \omega)^{-1}$ is a two-photon matrix element.

The following calculations will be carried out with $\Delta = 0$ in (6) because the Stark light shift Δ is small in comparison with the effective Rabi frequency Ω (for close atomic levels $|2\rangle$ and $|1\rangle$ $D_{22} - D_{11} << D_{21}$).

Before the interaction at time t = 0 with the light pulse an atom was in the ground state $|1\rangle$, hence using the initial condition $\rho_{11}(-\infty) = 1$ we find the coherence and the upper-level population probability

$$\rho_{21}(t) = \tfrac{1}{2}i\exp[-(\Gamma + i\omega_{21})t]\sin 2\theta , \quad (7)$$

$$\rho_{22}(t) = \exp(-2\Gamma t)\sin^2\theta , \quad (8)$$

where $t >> \tau$,

$$\theta = \tfrac{1}{2}\Omega G(\omega_{21}) ,$$

$$G(x) = \int_{-\infty}^{+\infty} dt|g(t)|^2 \exp(ixt) .$$

The parameter θ determines the power of the pulse perturbation: the case of $\theta << 1$ ($\Omega << \omega_{21} \leq \tau^{-1}$) corresponds to the weak perturbation and $\theta \simeq 1$ ($\Omega \simeq \tau^{-1} \geq \omega_{21}$) the strong one. The function $G(\omega_{21})$ is the value of the Fourier transform of a light-pulse shape at the frequency ω_{21}. It determines the atom-field interaction efficiency.

In the case of the Gaussian pulse $|g(t)|^2 = \pi^{-1/2} \times \exp(-t^2/\tau^2)$ we have $G(\omega_{21}) = \tau \exp[-(\omega_{21}\tau/2)^2]$. The atom-field interaction is shown to be efficient if $\tau \leq 2/\omega_{21}$. The increase of the pulse duration τ leads to a vanishing of spectral components at $\omega \pm \omega_{21}$ in a light pulse, so that two-photon interaction becomes impossible.

For the rectangular light-pulse shape we have $G(\omega_{21}) = \sin(\omega_{21}\tau/2)/(\omega_{21}/2)$. The efficiency is high for $\tau = \pi(2n-1)/\omega_{21}$, with n being integer here and elsewhere in the paper. The result of the atomic interaction with the light pulse of duration $\tau = \pi/\omega_{21}$ (n=1) is the same as in the case of a long pulse (n>>1). It means that only sharp fronts of the light pulse may give rise to the two-photon Raman process. The upper limit of τ will be defined by an accuracy, which is required for time localization of the atomic transition $|1\rangle \rightarrow |2\rangle$.

In this analysis we have neglected the one-photon excitation of the intermediate levels $|\alpha\rangle$, see Fig.1. This process reduces the number of atoms involved in the two-photon process. For $\omega_{\alpha 1} - \omega >> \tau^{-1}$ the α-level population is of the form

$$\rho_{\alpha\alpha} = (\Omega_{\alpha 1}^2/2)\, \gamma_{\alpha 1} \tau (\omega_{\alpha 1} - \omega)^{-2} ,$$

where $\Omega_{\alpha i}$ and $\gamma_{\alpha i}$ (i=1,2) are the one-photon Rabi frequency and the spontaneous decay rate of the transition $|\alpha\rangle \rightarrow |i\rangle$, respectively. The population ρ_{22} induced by the two-photon process in a weak field is $\rho_{22} \simeq \theta^2$, see (8). Thus, the inequality $\rho_{22} >> \Sigma_\alpha \rho_{22}$, which corresponds to suppressing the one-photon process, is reduced to

$$\Omega_{\alpha 2}^2 >> \tau \gamma_{\alpha 1} G^2(\omega_{21}) \sim \tau \gamma_{\alpha 1} \omega_{21}^2 ,$$

where α refers to the intermediate level which may be used to estimate the two-photon matrix element D_{21}.

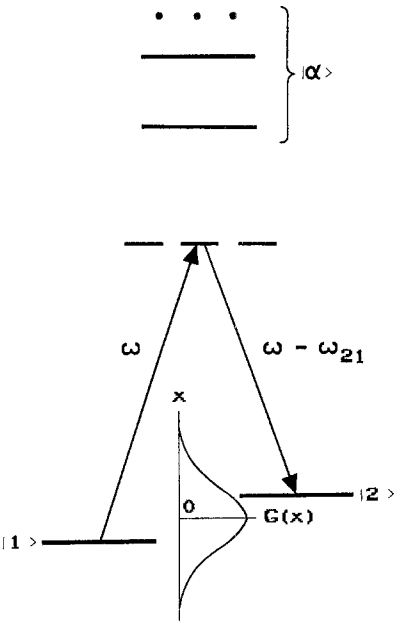

Fig.1. Two-photon Raman interaction between an atom and single ultrashort light pulse. G(x) is the Fourier transform of the pulse shape $|g(t)|^2$, x is a frequency

To conclude this discussion we note the following. First, the effect of the Stark light shift which we neglected above may be shown to be distinct in the strong field alone and lead to a decrease in the dynamical variation of the coherence amplitude $|\rho_{21}|$ and the probability ρ_{22} in proportion to the ratios Ω/Ω_0 and $(\Omega/\Omega_0)^2$, respectively, with $\Omega_0 = (\Omega^2+\Delta^2)^{1/2}$. Second, the formulas (7) and (8) may also be applied to the interaction between an atom and an ultrashort ($\tau \leq \omega_{21}^{-1}$) dc pulse of an electric field (for forbidden transitions). To use them one should equate the frequency ω in the two-photon matrix elements $D_{ik}(\omega)$ to zero.

3. Interaction Between an Atom and Two Light Pulses

Of the physical quantities that describe a light field, only the field power $\sim E^2|g(t)|^2$ appears in (6). So the interaction between an atom and a pair of the time-separated pulses will depend on the time delay T but not on the difference of their optical phases.

We have considered the excitation of an atomic natural oscillation with the frequency ω_{21} by one ultrashort light pulse, see (7). Another light pulse delayed by the time T (terminating pulse) will interact with the atom, if the phase of the oscillations is equal to $\omega_{21}T$. It is the phase that determines the result of the atomic interaction with the pair of ultrashort pulses.

Using (7) and (8) at time t = T as initial conditions for the interaction between the atom and the second light pulse we find from (6) the coherence and the upper-level population probability in the form

$$\rho_{21}(t) = \tfrac{1}{2} i\sin 2\theta \exp[-(\Gamma + i\omega_{21})(t - T)]$$
$$\times\{1 + \exp[-(\Gamma + i\omega_{21})T] - 2\sin^2\theta\exp(-\Gamma T)$$
$$\times[\exp(-\Gamma T) + \cos\omega_{21} T]\}, \quad (9)$$

$$\rho_{22}(t) = \sin^2\theta\exp[-2\Gamma(t - T)]$$
$$[1 + \exp(-2\Gamma T)\cos 2\theta$$
$$+ (1 + \cos 2\theta)\exp(-\Gamma T)\cos\omega_{21} T], \quad (10)$$

where $t - T >> \tau$. We see that the density-matrix variation for the small time τ of the terminating-pulse duration is synchronous with the atomic oscillations at the frequency ω_{21}. This is referred to as a synchronized stimulated quantum transition. There is a peculiarity of the transitions induced by the weak and strong perturbations, hence we consider them separately.

For the field which is weak to saturate the two-photon transition $|1\rangle \rightarrow |2\rangle$ ($\theta << 1$), we see from (7) and (8) that the starting light pulse induces the atomic coherence $\rho_{21} \sim E^2\tau$ and probability $\rho_{22} = |\rho_{21}|^2$. If the second pulse delayed in time arrives synchronously with the atomic oscillations ($T = 2\pi n/\omega_{21}$), then the amplitude of the previous ones is doubled (for $\Gamma T << 1$) and the upper level population probability is quadrupled, see (9) and (10), due to the interference of the probability amplitudes. If the delay time is equal to a half-integral multiple of the atomic period ($T = \pi(2n-1)/\omega_{21}$), the natural oscillations are annihilated and the atom returns into the ground state. The formulas (9,10) for a weak field are similar to (4,5).

For the strong light field ($\Omega \simeq \tau^{-1} \geq \omega_{21}$) we emphasize the case of a $\pi/4$-pulse ($\theta = \pi/4$) which yields the maximum amplitude of the synchronized density matrix variations. From (7 and 8) we see that the starting pulse induces the probability $\rho_{22} = \frac{1}{2}$ and the atomic oscillations with the amplitude $|\rho_{21}| = \frac{1}{2}$. The terminating pulse arriving synchronously with the atomic oscillations ($T = 2\pi n/\omega_{21}$) doubles the upper-level population probability and returns the atom into the ground state if $T = \pi(2n-1)/\omega_{21}$, with the natural oscillations being annihilated in both cases (Fig.2).

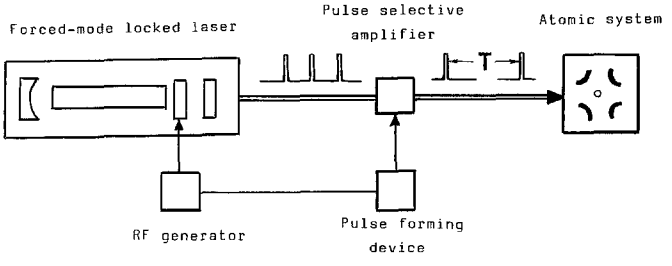

Fig.2. Interaction between an atom and two ultrashort light pulses ($\theta = \pi/4$)

4. Atomic Ensemble. Rarefied Gas

To find the upper-level population for an atomic ensemble we must take into account the atomic motion and average the population probability $\langle\rho_{22}\rangle$ over coordinates and velocities. Atomic motion is accounted for by replacing the value T on the right-hand side of (10) by the interpulse time $T_a = T + c^{-1}[z(T_a) - z_0]$ in the rest frame of the moving atom, where z_0 and $z(T_a)$ are the atomic coordinates along the light wave propagation direction at the times of the starting and terminating ultrashort pulses respectively. The ensemble-averaging of the upper-level population probability $\langle\rho_{22}\rangle$ is reduced to averaging the oscillating factor $\cos\omega_{21} T_a$.

For a free atom $z(T_a) - z_0 \simeq v_z T$, hence after the averaging over velocities v_z with the equilibrium distribution function we find

$$\langle\cos\omega_{21} T_a\rangle = \cos\omega_{21} T \exp[-(T\delta/2)^2], \quad (11)$$

where $\delta = \omega_{21} v_0/c$ is the Doppler shift at the Raman frequency, with v_0 being the thermal velocity. We see that for rarefied gas the observation of the quantum transitions synchronized with the atomic oscillations at the frequency ω_{21} is limited by the time interval $T \leq \delta^{-1}$ which yields an accuracy of time measurements of order $(\tau\delta)^{-1} \simeq c/v_0$.

The influence of the Doppler effect may be decreased by elastic collisions in a sufficiently dense gas [5]. In this case the diffusive distance which a particle covers during the interpulse time should be of the order of the wavelength associated with the atomic transition $|1\rangle \rightarrow |2\rangle$.

Note that in an atomic ensemble a spin-echo-like effect may be observed, i.e. the coherence $\rho_{21}(t)$ is seen to contain a term proportional to $\exp[-i\omega_{21}(t-2T)]\sin 2\theta \sin^2\theta$; see (9).

5. Ensemble of Trapped Atoms

During the interval between the starting and terminating pulses, an atom trapped in a finite volume may be displaced not more than the macroscopic

oscillation amplitude $v_{z\max}/\omega_z$, where ω_z is the frequency of atomic oscillations in a trap. Ensemble-averaging the factor $\cos\omega_{21}T_a$ we see that the parameter $\mu = \delta/\omega_z$ arises instead of $T\delta$ in (11). Furthermore, the Doppler effect does not limit the delay time T if the latter is a multiple of the macroscopic oscillation period. In this case the terminating pulse finds an atom at the same point as the starting one.

Let an atomic ensemble be trapped in a harmonic potential, with the principal axis of trap symmetry coinciding with the light direction Z. The equation of motion of a single atom is of the form

$$z(t) = z_0\cos\omega_z t + (v_{z0}/\omega_z)\sin\omega_z t\,.$$

Averaging over the initial coordinates z_0 and velocities v_{z0} we find

$$\langle\cos\omega_{21}T_a\rangle = \cos\omega_{21}T\exp[-\mu^2\sin^2(\omega_z T/2)]\,.$$

This confirms our above statements.

6. Laser Spectrometer Scheme

In the method under consideration the light source has a very large linewidth ($\Delta\omega\simeq 1/\tau$), and the value of its carrier frequency is unimportant. Only the stability of the pulse delay time is significant. There are at least two possibilities to realize the spectrometer. In the first case, an optical delay line may be used to form the terminating pulse. The delay time T = L/c (where L is the delay line length) may be changed continuously with a high accuracy. Unfortunatley, the absolute accuracy of the delay time measurement is limited here by the length measurement accuracy. If the transition frequency is known, then according to the new definition of the meter, the delay time T and the length L may be directly measured. The second possibility is given by the laser spectrometer scheme shown in Fig.3. This spectrometer allows one to measure the absolute values of both the delay time T and the transition frequency ω_{21}. The spectrometer is based on the use of ultrashort pulses generated by the forced-mode-locked laser. The pulse repetition frequency is determined by the frequency ν of the RF generator, which controls the intracavity amplitude modulator. The optical pulse amplifier locked to the rf generator allows one to form the time-separated pulses. Their delay time will be a multiple of the laser interpulse time, i.e. $T = n\nu^{-1}$. Usually $\nu \sim 10^8$ Hz. Time tuning may be realized by tuning the frequency ν. Such a system may serve as a standard for time and frequency simultaneously. As the frequency ω_{21} is stable, it is possible to stabilize the delay time T and consequently the rf generator frequency ν.

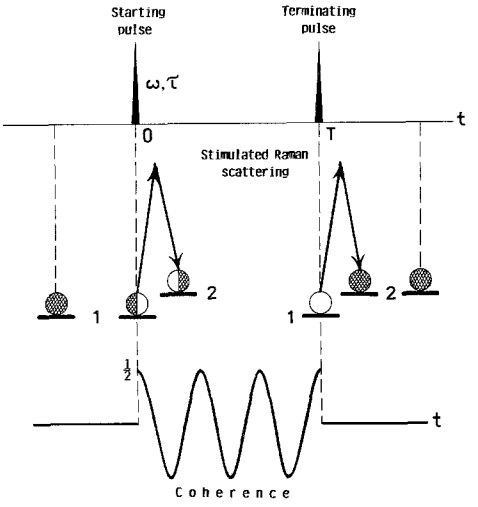

Fig.3. A possible spectrometer scheme for observing the synchronized quantum transitions

7. Conclusion

We have shown that for a single atom or an atomic ensemble the dynamics of the natural Raman oscillations may be investigated by a synchronization of atomic quantum transitions with these oscillations. This synchronization may be realized, for example, by time-separated ultrashort light pulses. The proposed effect may be used in super-high precision frequency measurement, developing new foundations for time standards and magnetometers, the measurement of atomic spectroscopy constants, the selective excitation of atoms and molecules, etc. The possibility of utilizing this phenomenon to develop high-speed atomic memory systems is also of interest.

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