Synchronization of quantum transitions

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The interaction of light pulses with a quantum-mechanical particle is analyzed. When the length of the pulses is shorter than the period of the oscillations of the transition, the probability of the excitation of the particle is a rapidly oscillating function of the delay time between pulses. The period of the oscillation corresponds to the transition frequency.

1. We know quite well that after a linear oscillator is excited by two short pulses, whose length is much shorter than the oscillation period, it will execute sinusoidal oscillations. These oscillations can be represented as a superposition of two oscillations, with amplitudes and phases which correspond to independent excitation by two pulses:

$$C(t) = A\sin\omega t + B\sin\omega(t+T), \tag{1}$$

where $C(t)$ is the resultant oscillation, of amplitude C; A and B are the amplitudes of the oscillations corresponding to the first and second pulses; and T is the pulse delay time. The amplitude C is given by the expression:

$$C = (A^2 + B^2 + 2AB\cos\omega T)^{1/2}, \tag{2}$$

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from which it follows that the resultant amplitude C depends directly on the phase of the oscillation of the oscillator at the time at which the second pulse arrives. In the present letter we show that the behavior of a quantum-mechanical particle as it is excited by two short pulses is also determined by the phase of the dipole moment of the particle at the time of excitation by the second pulse. In particular, in the interaction of a two-level (or three-level) particle with two very short light pulses, whose length τ is shorter than the oscillation period of the corresponding transition ($\tau \ll \omega_{21}^{-1}$, where ω_{21} is the transition frequency), the excitation of a level will be determined by the phase $\omega_{21} T$.

The features which we see here, while simple, may prove important for solving various scientific and applied problems.

2. We consider the interaction of a short light pulse, of length τ and carrier frequency ω, with a three-level particle. In accordance with the discussion above, we assume that the transition frequency ω_{21} satisfies the condition $\omega_{21} \ll \tau^{-1}$. The equations for the probability amplitude take the form

$$\dot{a}_0 = a_1 G_{01} e^{-i\Omega t} + a_2 G_{02} e^{-i\Omega' t} \qquad \dot{a}_1 = a_0 G_{10} e^{i\Omega t} \qquad \dot{a}_2 = a_0 G_{20} e^{i\Omega t} - \gamma a_2$$
$$G_{01} = i p_{01} E/\hbar, \qquad G_{02} = i p_{02} E/\hbar \tag{3}$$

Here p_{01} and p_{02} are matrix elements of the dipole moment of the transition $0 \to 1$ and $0 \to 2$, respectively; $\Omega = \omega - \omega_{01}$ and $\Omega' = \omega - \omega_{02}$ are the deviations of the field frequency from the resonant frequencies of transitions $0 \to 1$ and $0 \to 2$; γ is the damping of level 2; and E is the field amplitude of the first pulse.

At $t = 0$ we have $a_1 = 1$ and $a_2 = a_0 + 0$. After the first pulse has acted, the probability amplitude is given by $a_2^{(1)} = (-1/2 G_{02}^* G_{01} \tau^2$. After the interaction with the second pulse, delayed a time T, the probability amplitude $a_2^{(2)}$ is

$$a_2^{(2)} = - G_{01} G_{02}^* (e^{-\gamma T} + \beta e^{i\omega_{21} T}) \tau^2 / 2, \tag{4}$$

where β is the ratio of the amplitudes of the pulses. Correspondingly, the probability for finding the particle in level 2 is

$$|a_2^{(2)}|^2 = |G_{02}|^2 |G_{01}|^2 \tau^4 (1 + \beta^2 e^{-2\gamma T} + 2\beta e^{-\gamma T} \cos\omega_{21} T) 1/4 . \tag{5}$$

The probability for finding a particle in level 2 is thus a rapidly oscillating function of the delay T. The excitation maximum corresponds to a delay time which is a multiple of the oscillation period of the $1 \to 2$ transition. At a low damping rate, the delay ($T \sim 1/\gamma$) can be quite large and can be measured within a relative error better than γ / ω_{21}, which can have a value of 10^{-12}–10^{-13}. If there is a fine structure in the population of level 2, we will observe beats as the time T is varied. The period of these beats will depend on the difference between the transition frequencies. This circumstance can also be utilized in ultrahigh-resolution spectroscopy. The method may thus be thought of as a supplement to other time-varying methods of coherent spectroscopy.

3. Tunable sources with a narrow output line have been the sources used most

commonly in ultrahigh-resolution spectroscopy. The frequency of such a source is tuned to the center of the transition. In the method which we are considering here, the source has a very broad line ($\Delta\omega \sim 1/\tau$), and the value of its carrier frequency is not important. The only point of fundamental importance is that the delay time T be stable. There are at least two possibilities for designing a spectrometer. First, one could use an optical delay line to form a delayed pulse. The duration of the delay would be $T = L/c$, where L is the length of the delay line, and c is the velocity of light. This delay time can be adjusted highly precisely. Unfortunately, the absolute accuracy of the measurement of the delay time is limited here by the accuracy with which the length can be measured. If the absolute transition frequency is known, the delay time T and the length L can be measured at once in accordance with a new definition of the meter. Second, one could carry out both an absolute measurement of the delay time and an absolute measurement of the frequency of the $1 \to 2$ transition. In this case, one would use ultrashort pulses from a mode-locked laser. With a forced mode locking the pulse repetition frequency would be determined by the frequency (Ω) of the generator which powers the amplitude modulator inside the resonator. An optical pulsed amplifier synchronized with this generator would make it possible to select and shape individual pulses. The delay of these pulses would be a multiple of the distance between pulses; i.e., we would have $T = n\Omega^{-1}$, where n is an integer, and Ω is the generator frequency, in hertz. We would usually have $\Omega \approx 10^8$ Hz. The delay T can be adjusted smoothly over a narrow interval by varying the frequency Ω. A system of this sort could double as a time and frequency standard. If the frequency Ω_{21} is stable, it could be used for a stabilization of the delay time T and thus of the frequency Ω.

The features of the interaction of ultrashort light pulses with particles which we have discussed here open up some new possibilities for ultrahigh-resolution spectroscopy, for developing new principles for developing time standards and magnetometers, for measuring transition frequencies, and for a selective excitation of levels. The possibility of utilizing these processes to develop fast memory systems based on the use of atoms and molecules deserves study. Already, the attainment[1] of a pulse length of 10^{-14} s makes it possible to carry out experiments at wavelengths to 10 μm.

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[1]Proceedings of PSA Topical Meeting, Snowmass, Colorado, June 16–19, 1986; G. R. Fleming and A. E. Siegmann (editors), *Ultrafast Phenomena*, Vol. 5, Springer-Verlag, New York.

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