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NONLINEAR OPTICAL PHENOMENA

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Population difference gratings produced by unipolar subcycle pulses in a resonant medium

R.M. Arkhipov, M.V. Arkhipov, I. Babushkin, A.V. Pakhomov, N.N. Rosanov

Abstract. **We demonstrate the possibility of inducing, erasing and extra rapidly controlling population difference gratings resulting from coherent interaction of unipolar subcycle pulses with a resonant medium. Gratings can be produced without overlap of pulses in the medium, which is an important distinction of the proposed approach from the traditional one, in which gratings are produced using interference of two or more overlapping quasi-monochromatic light beams. The use of unipolar subcycle pulses ensures faster control over gratings in comparison with bipolar pulses studied by us previously.**

Keywords: *unipolar optical pulses, population difference grating, resonant optical media.*

1. Introduction

Advances in the generation of femto- and attosecond few-cycle pulses (FCPs) [1–3] paved the way for the advent of attosecond science and made it possible to gain insight into a number of previously unapproachable light–matter interaction processes. For example, it became possible to control ultrafast processes in matter, including control over the dynamics of wave packets in matter, which provided insight into fundamental aspects of the structure of matter [4, 5] and enabled control over electron beams on a subcycle timescale [6]. The FCPs thus obtained are bipolar, i.e. the time integral of the electric field strength at a given point of space (the electrical area of the pulse) is zero. Recent years have seen special interest in the generation of so-called unipolar pulses (UPs), i.e. pulses that have a nonzero electrical area (see reviews [7, 8] and research papers [9–18]). Since there is a static field component, such pulses have a unidirectional action on electric charges: unlike in the case of a bipolar pulse, here a nonzero mechanical momentum can be imparted to a charged particle, and the charge can continue its motion after the action of the UP. This makes UPs a unique tool for controlling the dynamics of charged particles and accelerating them by such a field.

It is traditionally thought that UPs cannot be obtained because the source of a field being emitted – acceleration of a system of coupled charges – is always bipolar. Analysis shows however that, in a number of situations, including a one-dimensional case, UPs can be produced, e.g., when a single-cycle pulse is reflected from a thin metallic (or dielectric) film [19]. There are also soliton solutions to equations of nonlinear optics in the form of UPs (see Ref. [7] and references therein).

Recent work [20, 21] predicted one unusual aspect of interaction of bipolar FCPs with a resonantly absorbing medium: the possibility of inducing and erasing polarisation and population difference gratings and multiplication (ultrafast control) of their spatial frequency using FCPs that do not overlap in the medium. An essential point is that overlap of pulses is unnecessary only in the case of coherent interaction of FCPs with resonant media, when the pulse duration τ_p is shorter than the population difference relaxation time T_1 and the polarisation relaxation time T_2, $\tau_p \ll T_1, T_2$ [22]. In a traditional approach, spatial overlap of two or more interfering beams is required for producing gratings [23]. Diffraction of light by photoinduced gratings thus produced finds wide application in spectroscopy and nonlinear optics [23]. In the case of coherent interaction, control and fabrication of gratings are the result of interaction with the wave of resonance polarisation produced by the preceding pulse [20, 21]. Note that the possibility of producing population gratings via coherent interaction with a resonant medium using a train of pulses that do not overlap in the medium was demonstrated experimentally in the nanosecond range as early as the first photon echo experiments [24–26], but the 'coherent mechanism' of grating formation was abandoned and found no optical applications.

In previous work [20, 21], excitation pulses were taken to be bipolar. In addition to the above-mentioned benefits, UPs shorter than the field period (subcycle pulses) contain one field half-wave and are shorter than bipolar FCPs. This circumstance will allow one to produce gratings and control them on timescales of the order of half the light wave period, which is considerably faster than in the case of bipolar pulses. However, in the case of subcycle pulses the concept of carrier

R.M. Arkhipov, M.V. Arkhipov St. Petersburg State University, Universitetskaya nab. 7/9, 199034 St. Petersburg, Russia; e-mail: arkhipovrostislav@gmail.com;
I. Babushkin Max Born Institute, 10117, Berlin, Germany; Institute of Quantum Optics, Leibniz University Hannover, 30167, Hannover, Germany;
A.V. Pakhomov Samara National Research University, Moskovskoe sh. 34, 443086 Samara, Russia; P.N. Lebedev Physical Institute (Samara Branch), Russian Academy of Sciences, Novo-Sadovaya ul. 221, 443011 Samara, Russia;
N.N. Rosanov S.I. Vavilov State Optical Institute, Kadetskaya liniya 5/2, 199053 St. Petersburg, Russia; St. Petersburg National Research University of Information Technologies, Mechanics and Optics, Kronverkskii prosp. 49, 197101 St. Petersburg, Russia; Ioffe Physical Technical Institute, Russian Academy of Sciences, Politekhnicheskaya ul. 26, 194021 St. Petersburg, Russia

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frequency is not defined, and typical views of resonant coherent light–matter interaction turn out to be inapplicable [27–31]. For example, the McCall–Hahn area theorem is violated and the concept of pulse area becomes inapplicable. This led us to examine the feasibility of producing gratings via coherent interaction of unipolar subcycle pulses with a resonantly absorbing medium. We demonstrate that such gratings can exist in spite of apparent limitations. The effect under study can be used for the fabrication of ultrafast laser beam deflectors and ultrafast optical switches and furthers a recently proposed [32] new concept of coherent photonic devices, i.e. nonlinear photonic devices operating in the coherent light–matter interaction regime.

2. Numerical simulation results

To examine the feasibility of producing polarisation and population difference gratings in a resonant medium by a train of unipolar subcycle pulses, we used the Maxwell–Bloch system of equations. Given the short duration of excitation pulses, neither the slowly varying envelope approximation nor the rotating wave approximation was used. The equations have the following form [22, 33, 34]:

$$\frac{d\rho_{12}(z,t)}{dt} = -\frac{\rho_{12}(z,t)}{T_2} + i\omega_0\rho_{12}(z,t) - \frac{i}{\hbar}d_{12}E(z,t)n(z,t), \quad (1)$$

$$\frac{dn(z,t)}{dt} = -\frac{n(z,t)-n_0(z)}{T_1} + \frac{4}{\hbar}d_{12}E(z,t)\,\mathrm{Im}\rho_{12}(z,t), \quad (2)$$

$$\frac{\partial^2 E(z,t)}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2 E(z,t)}{\partial t^2} = \frac{4\pi}{c^2}\frac{\partial^2 P(z,t)}{\partial t^2}, \quad (3)$$

$$P(z,t) = 2N_0 d_{12}\,\mathrm{Re}\rho_{12}. \quad (4)$$

The medium was considered in the two-level approximation. Equations (1) and (2) describe the evolution of the off-diagonal density matrix element ρ_{12} and that of the difference between the diagonal density matrix elements, n, which has the meaning of the population difference (inversion) between the ground and excited states of the two-level system. The resonant transition is thought to be homogeneously broadened. Numerical calculations demonstrate that taking into account additional levels of the medium [10–13] or inhomogeneous broadening [11, 13, 35–38] causes no drastic changes in the key features of the coherent propagation of few-cycle pulses in resonant media.

The propagation of pulses with an electric field $E(t)$ in the medium is described by the wave equation (3), which includes the polarisation of the medium, $P(t)$, defined by (4). The polarisation is related to the off-diagonal density matrix element ρ_{12}. In addition, the system of equations includes the following parameters: ω_0 is the frequency of the resonant transition in the medium; d_{12} is the transition dipole moment; n_0 is the population difference in the absence of an electric field ($n_0 = 1$ for an absorbing medium); c is the speed of light in vacuum; $\hbar$ is the reduced Planck constant; and N_0 is the concentration of two-level atoms. The Maxwell–Bloch system of equations (1)–(3) was integrated numerically. The Bloch equations for the density matrix (1) and (2) were solved by the fourth-order Runge–Kutta method, and the wave equation (3) was solved by a finite-difference method.

The train of unipolar excitation pulses had a Gaussian shape:

$$E_i(t) = \sum_{i=1}^{N} E_0 \exp\left(-\frac{[t-\tau_i]^2}{\tau_p^2}\right). \quad (5)$$

Here τ_i is the delay of the ith pulse relative to the first pulse and N is the number of excitation pulses. The delays between the pulses, τ_i, were adjusted so that the pulses did not overlap in the medium. The amplitude of all the pulses was E_0.

Figure 1 shows the variation in the polarisation and population difference induced by a train of UPs of the form (5) with negligible overlap in the medium. The number of excitation pulses is $N = 3$. The pulse amplitude was adjusted so that the first pulse acted as a $\pi/2$ pulse. After such a pulse, the medium has zero inversion. The following parameters were used in the simulation: resonant transition wavelength $\lambda_0 =$

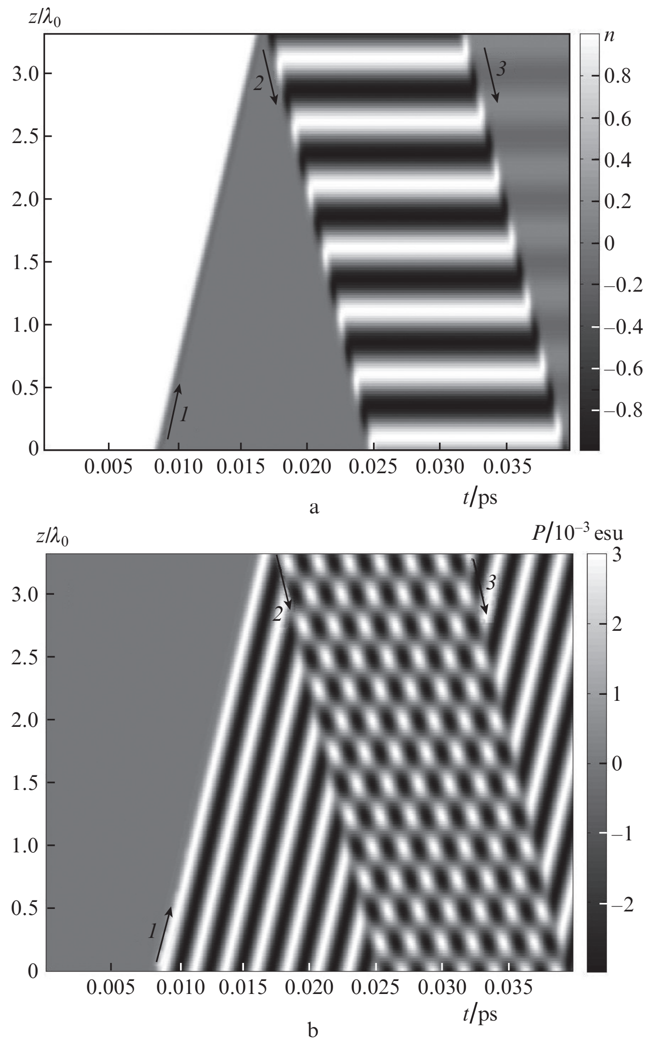

Figure 1. Variations in (a) inversion and (b) polarisation under the effect of a train of three unipolar subcycle pulses (*1* – *3*). The arrows indicate the propagation direction. The parameters used in the simulation are specified in text.

700 nm (natural oscillation period $T_0 = \lambda_0/c = 2.33$ fs), $d_{12} =$ 20 D, $N_0 = 1.5\times 10^{14}$ cm^{-3}, length of the medium of the order of $4\lambda_0$, $T_1 = T_2 = 1$ ns, unipolar pump pulse amplitude $E_0 =$ 9.55×10^4 esu and pulse duration $\tau_p = 0.38$ fs ($T_0/6$). Dipole moments of tens of debyes are typical of quantum dots. The relaxation time T_2 in quantum dots can also reach several nanoseconds at low temperatures [39].

The numerical simulation results demonstrated the following dynamics of the system: The first $\pi/2$ pulse propagated from left to right in the medium, with zero inversion (Fig. 1a) and a travelling polarisation wave (Fig. 1b) left behind. The second pulse was opposite in polarity to the first and was sent to the medium from right to left with a delay, after the first pulse had left the medium. The delay time is substantially shorter than the relaxation times in the medium, so the polarisation induced by the preceding pulse does not damp out before the next pulse arrives (see Fig. 1, which shows the instants when a pulse appears in the medium). The interference of the second pulse with the polarisation wave produced by the first pulse led to the formation of a population difference grating with a period equal to half the resonant transition wavelength, $\lambda_0/2$. The polarisation of the medium had the form of a standing wave (Fig. 1b). A third pulse, propagating from right to left after the second pulse with some delay, erases the grating (Fig. 1a), bringing the system back to the zero inversion state. The process can be continued by increasing the number of excitation pulses. The effect in question is observed in wide ranges of parameters of the system and medium. The only limitation on grating formation is imposed by the relaxation times T_1 and T_2 of the medium.

3. Conclusions

The possibility of inducing and erasing population difference gratings in a resonantly absorbing medium by a train of unipolar subcycle pulses has been predicted for the first time. The interaction of UPs with the medium is coherent, and gratings can be produced without overlap of pulses in the medium, unlike in the traditional approach.

The effect studied here can be used for the fabrication of ultrafast laser beam deflectors, optical switches and Bragg gratings. Travelling polarisation waves can be thought of as relativistic mirrors and serve to convert the frequency of reflected pulses [40], because such mirrors can move at relativistic speeds, in contrast to 'material' mirrors. The effect can be observed in gases and quantum dots. The latter are characterised by large dipole moments of transitions (tens of debyes) and long polarisation relaxation times at low temperatures (several nanoseconds) [39].

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