All-optical control of unipolar pulse generation in a resonant medium with nonlinear field coupling

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We study the optical response of a resonant medium possessing nonlinear coupling to an external field driven by a few-cycle pump pulse sequence. We demonstrate the possibility of directly producing unipolar half-cycle pulses from the medium possessing an arbitrary nonlinearity, by choosing the proper pulse-to-pulse distance of the pump pulses in the sequence. We examine various ways of shaping the medium response using different geometrical configurations of nonlinear oscillators and different wavefront shapes for the excitation pulse sequence. Our approach defines a general framework to produce unipolar pulses of controllable form.

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I. INTRODUCTION

Generation of ultrashort pulses is a field of active research due to emerging opportunities for real-time control of ultrafast processes [1–4]. Since optical pulses of few-cycle or even subcycle duration became available, the number of applications for them increases continually, enhancing our understanding of fundamental phenomena in atoms, molecules, and condensed matter. The shortest possible pulse duration opens up new opportunities for the direct measurement of extremely highspeed dynamical processes important for various branches of physics, chemistry, medicine, and biology. The development of new techniques for generation and control of ultrashort pulses is therefore crucial for the advancement of modern optical and material science.

Compared with few-cycle pulses, half-cycle optical pulses possess an exceptional feature of being unipolar, that is, the electric field does not change its sign throughout the pulse. Such a field shape gives rise to light-matter interactions that are not possible using conventional light pulses. In particular, this property can be an advantage if one wants to control the ultrafast charge motion in pump-probe experiments. Specifically, unipolar pulses can efficiently deliver a kinetic momentum to the charged particles in order to control their motion, for instance, to ionize the atoms or ions in the medium [5-7] or to measure the quantum dynamics of electron and ionic wave packets [8–11]. Unipolar pulses can efficiently accelerate charge particles and thus be used for producing coherent beams for particle injectors and charge-particle accelerating devices [12,13]. Over the last years, unipolar pulses have also been proposed for the generation of isolated highly intense attosecond pulses [14-16]. It was recently shown that the coherent light-matter interaction with the use of unipolar pulses enables the control of resonant medium properties (population inversion and polarization gratings) on a subcycle

time scale [17,18]. Therefore, finding effective means of unipolar pulse production and control is needed. It is important to note, though, that outlined applications do not require pulses that are strictly unipolar. Effectively one can consider pulses with a central part of constant sign and long tails on each side that are opposite in sign, which allows us to avoid the difficulties of dealing with zero-frequency component.

To date, subcycle pulses have been generated in the visible, near-infrared, terahertz, and x-ray ranges [19–23] and, recently, in the midinfrared [24]. Half-cycle pulses have been experimentally obtained from laser-driven plasma in a solid target [25] and from a double-foil target irradiated with intense few-cycle laser pulses [26]. Theoretically unipolar pulses were predicted when an initially bipolar ultrashort pulse propagates in a nonlinear resonant medium [27–30] and in a Raman-active medium in a self-induced transparency regime [31,32] or under excitation by few-cycle pulses [33–35].

Ultrashort laser pulse shaping technologies find everwidening applications in the fields of coherent control, high-field laser-matter interactions, nonlinear microscopy and spectroscopy, biotechnology, and optical data processing [36–39]. All these applications prove to be highly sensitive to the maximal flexibility in ultrashort-pulse profiling. Today's mainstream research in the generation of ultrabroadband pulses is developing towards subcycle waveform synthesis, which is the coherent combination of pulses generated from different sources covering separate spectral regions [20,22,40-44]. The synthesis of ultrashort pulses calls for careful manipulation of the parameters of the individual pulses, namely, the spectral phase, carrier phase, and relative delay between the sources. The ultrabroad spectrum also requires a highly accurate spectral phase and amplitude control to process ultrashort pulses, what implies the usage of complicated pulse shaping systems to manipulate components extended over large bandwidths. Therefore, the possibilities of arbitrary optical waveform generation appear to be technically challenging and are naturally limited by the shaping device processing characteristics. The possibility of ultrashort and, particularly,

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unipolar pulse shaping directly during the generation process thus seems to be an extremely inviting prospect. We devise in this paper an alternative approach for the generation of ultrashort waveforms, which we hope will make a contribution towards this direction.

In Ref. [45] the optical response of a one-dimensional string made of two-level oscillators with a periodically varying density excited at superluminal velocity by an incident ultrashort pulse was studied. It was shown that such a system allows the generation of Cherenkov radiation of an unusual shape; namely, an additional resonant frequency appeared in the radiation spectrum. Furthermore, in [46] this consideration was extended to the case where the coupling of the oscillators to the field is nonlinear; a concept of unipolar pulse generation was proposed when the oscillators are excited by a train of few-cycle pulses.

In this paper, we generalize this approach to the case of an arbitrary shape of nonlinear coupling of the oscillators to the external field as well as arbitrarily curved wavefronts and different geometries of the oscillator arrangement. Special consideration is devoted to the role of the carrier-envelope phase (CEP) of the pump pulses in unipolar pulse generation. We show that the CEP value can have a profound impact on the medium optical response and its proper adjustment can be important to obtain unipolar pulses.

As we show, combined into spatially extended arrays, such oscillators can allow direct generation of unipolar pulses of a controllable waveform. This is possible because the excitation velocity varies upon propagation along the oscillator array, in contrast to the case of a constant excitation velocity [33–35]. We study in detail several particular spatial arrangements of the oscillators, for instance, located on a planar string or on a disk. Our findings show that this approach has great possibilities for unipolar pulse shaping.

The paper is organized as follows. In Sec. II we derive the general condition defining the possibility of unipolar pulse generation in our system. In Secs. III and IV we discuss the most promising examples for unipolar pulse shaping. Finally, in Sec. V we give a brief overview of our findings and present concluding remarks.

II. GENERAL CONDITION FOR HALF-CYCLE PULSE GENERATION

We consider in the following a model of a resonant optical oscillator excited by a few-cycle pump pulse. We assume the excitation field to be linearly polarized, which gives us the equation for the evolution of medium polarization P(t),

$$\ddot{P} + \gamma \dot{P} + \omega_0^2 P = g[E(t)]E(t), \qquad (1)$$

where ω_0 is the oscillator resonant frequency, γ is the damping rate, and the function g[E(t)] describes the coupling strength of the oscillator to the field. Considering that the field coupling can be anisotropic, Eq. (1) should be written for each component of the polarization vector, but with a linearly polarized electric field these equations have a form analogous to Eq. (1). Hence, without loss of generality we can restrict ourselves to the scalar case described by Eq. (1).

The coupling-field function g[E(t)] can be arbitrary. Physically, it can be implemented by several nonlinearly coupled oscillators with strongly different parameters such as effective mass and resonant frequency. For instance, we may consider just two such oscillators; the first is excited by a high-frequency external field and induces slow motion of the other oscillator through nonlinear bonding. By excluding the slow motion, we arrive at a nonlinear equation like Eq. (1). Such nonlinear field coupling can be expected, for instance, for nonlinearly coupled localized plasmonic resonances in metallic nanostructures [47,48], hybrid aggregates of organic supramolecular assemblies and inorganic nanocrystals [49,50], coupled semiconductor microcavities [51], doublequantum-dot heterostructures [52,53], and other hybrid optical materials. Natural examples of such field coupling include also Raman-active media [54,55].

We suppose that the pump pulse is short compared to the natural period of oscillations: $\omega_0 \tau_p \ll 1$. We also assume that the oscillation decay rate $\gamma \ll \omega_0$ and consider oscillations in the time interval of the order of the natural period, so that oscillator damping can be neglected. Then, setting $u(t) = \dot{P}(t) + i\omega_0 P(t)$, we obtain the following equation:

$$\dot{u} - i\omega_0 u = g[E(t)]E(t). \tag{2}$$

The assumption of a short pulse duration allows us to suggest that the oscillator is affected by instantaneous forcing followed by the free oscillations. With that Eq. (2) yields, for the induced polarization dynamics right after the pump pulse action for t > 0,

$$u(t) = e^{i\omega_0 t} \left\{ u_0 + \int_{-\infty}^{+\infty} g[E(t')]E(t')e^{-i\omega_0 t'}dt' \right\}.$$
 (3)

The integral on the right-hand side of Eq. (3) is considered to be taken over the whole pulse duration, which is indicated by the infinite integration limits. Keeping in mind that the electric field is real and splitting the complex exponent under the integral sign into real and imaginary parts, Eq. (3) gives

$$P(t) = P_0 \sin(\omega_0 t + \phi_0) + \frac{\Pi_1}{\omega_0} \sin(\omega_0 t) - \frac{\Pi_2}{\omega_0} \cos(\omega_0 t),$$
(4)

$$\dot{P}(t) = \omega_0 P_0 \cos(\omega_0 t + \phi_0) + \Pi_1 \cos(\omega_0 t) + \Pi_2 \sin(\omega_0 t),$$
(5)

where P_0 and ϕ_0 correspond to the oscillation amplitude and phase at the moment of excitation pulse arrival and Π_1 and Π_2 are given by

$$\Pi_{1} = \int_{-\infty}^{+\infty} g[E(t')]E(t')\cos(\omega_{0}t')dt',$$

$$\Pi_{2} = \int_{-\infty}^{+\infty} g[E(t')]E(t')\sin(\omega_{0}t')dt'.$$
(6)

The oscillator is supposed to be initially at a standstill, i.e., $P_0 = 0$. Thus, as we have shown in recent papers [33–35], to emit unipolar pulses, the sine response in Eq. (4) is required. In this case the oscillator emission induced by one few-cycle pulse can be stopped by another identical pulse at the half-period delay in such a way that the emitted field maintains a constant sign. The emitted field then has the form of a half-cycle optical pulse. This result holds since the far field

emitted by the oscillator in between excitation pulse action at an arbitrary point \overline{r}' is given as

$$\overline{E}(\overline{r}',t) \sim \overline{\overline{P}}(\overline{r},t-|\overline{r}-\overline{r}'|/c) \sim \omega_0^2 \overline{P}(\overline{r},t-|\overline{r}-\overline{r}'|/c),$$
(7)

and thus, except for the constant factor, the emitted field is proportional to the polarization itself. For this reason, the presence of a nonzero cosine term in Eq. (4) necessitates a nonunipolar emitted field due to a cosine function that has a varying sign during the first oscillation half-period.

Thus, from Eqs. (4) and (5) one gets the necessary criterion for unipolar half-cycle pulse production by means of the proposed method:

$$\Pi_2 = \int_{-\infty}^{+\infty} g[E(t')]E(t')\sin(\omega_0 t')dt' = 0.$$
(8)

Analysis of Eq. (8) reveals some rather significant results. Let us consider an excitation pulse possessing a symmetric envelope with respect to the middle of the pulse (e.g., of a Gaussian shape) and an arbitrary phase shift of the carrier:

$$E(t) = E_0 e^{-t^2/\tau_p^2} \cos(\Omega t + \vartheta_{\rm CE}), \qquad (9)$$

where Ω is the central frequency and ϑ_{CE} stands for the carrierenvelope phase. Suppose first that $\vartheta_{CE} = \pm \frac{\pi}{2}$, implying that the driving electric field is an odd function with respect to the middle of the pulse. In this case Eq. (8) means that the field coupling function g[E(t)] must necessarily be odd as well. For example, the field coupling strength may be proportional to the odd degree of the driving field, as shown in [46] for the simplest linear dependence.

However, for another CEP, $\vartheta_{CE} \neq \pm \frac{\pi}{2}$, the situation turns out different. Assume the most important case, $\vartheta_{CE} = 0$. As can be seen from Eqs. (8) and (9) the field coupling function g[E(t)] in this case can be of arbitrary power-law dependence on the driving field E(t). This means that even the simplest linear oscillator with $g[E(t)] = g_0 = \text{const}$ can be driven to exhibit a sine response and thus to allow halfcycle pulse generation with accurate adjustment of the CEP. Equations (6) yield the dependence of the linear oscillator response amplitudes Π_1 and Π_2 on the carrier-envelope phase ϑ_{CE} ,

$$\Pi_{1} = \sqrt{\pi} E_{0} g_{0} \tau_{p} e^{-(\Omega^{2} + \omega_{0}^{2})\tau_{p}^{2}/4} \cosh\left(\Omega\omega_{0}\tau_{p}^{2}/2\right) \cos\vartheta_{\text{CE}},$$

$$\Pi_{2} = -\sqrt{\pi} E_{0} g_{0} \tau_{p} e^{-(\Omega^{2} + \omega_{0}^{2})\tau_{p}^{2}/4} \sinh\left(\Omega\omega_{0}\tau_{p}^{2}/2\right) \sin\vartheta_{\text{CE}},$$
(10)

which are illustrated in Fig. 1. Since $\Omega \gg \omega_0$ due to the short excitation pulse duration, it is seen from Eq. (10) that the cosine term amplitude Π_2 is equal to 0 for $\vartheta_{CE} = 0$ and $\vartheta_{CE} = \pm \pi$ only, but except in the vicinity of $\vartheta_{CE} = \pm \frac{\pi}{2}$, the sine term amplitude Π_1 is much greater. Considering that $\omega_0 \tau_p \ll 1$ and $\Omega_0 \tau_p \sim 1$, the observed relation between response amplitude Π_1 and response amplitude Π_2 is provided by

$$\max_{\vartheta_{\rm CE}} \Pi_2 / \max_{\vartheta_{\rm CE}} \Pi_1 = \tanh\left(\Omega \omega_0 \tau_p^2 / 2\right) \approx \Omega \omega_0 \tau_p^2 / 2 \ll 1.$$

The resulting oscillator response P(t) is depicted in Fig. 2 (red lines) for different CEP values together with the appropriately scaled electric field of the excitation pulses (black lines).



FIG. 1. Dependence of the linear oscillator response amplitudes Π_1 and Π_2 , (10), on the carrier-envelope phase ϑ_{CE} ; $\omega_0 \tau_p = 0.04$, $\Omega/\omega_0 = 100$.

According to Eq. (7), the emitted pulse appears to be unipolar when $\vartheta_{CE} = 0$ and $\vartheta_{CE} = \pm \pi$ and contains a certain portion of opposite sign for other CEP values. Some high-frequency oscillations distorting the emitted pulse shape are inevitably observed during the action of exciting pulses, but we suppose them to be effectively cut off by the appropriate low-pass filter.

It should be noted that the oscillation amplitude delivered to the linear oscillator by the excitation pulse is nevertheless substantially lower than can be achieved for the oscillator with nonlinear field coupling. The oscillator response amplitudes Π_1 and Π_2 for the field coupling function $g[E(t)] = g_1E(t)$



FIG. 2. The linear oscillator response P(t) (red line) and the electric field of the exciting pulses, (9) (black line), for different values of the carrier-envelope phase: (a) $\vartheta_{CE} = 0$; (b) $\vartheta_{CE} = \frac{\pi}{3}$; (c) $\vartheta_{CE} = \frac{7\pi}{15}$; (d) $\vartheta_{CE} = \frac{\pi}{2}$. $\omega_0 \tau_p = 0.04$, $\Omega/\omega_0 = 100$. The time delay between two excitation few-cycle pulses is equal to $T_0/2$.

are given as follows:

$$\Pi_{1} = \frac{1}{2} \sqrt{\frac{\pi}{2}} E_{0}^{2} g_{1} \tau_{p} e^{-\omega_{0}^{2} \tau_{p}^{2}/8} \\ \times \left[1 + e^{-\Omega^{2} \tau_{p}^{2}/2} \cosh\left(\Omega \omega_{0} \tau_{p}^{2}/2\right) \cos 2\vartheta_{\text{CE}} \right], \\ \Pi_{2} = -\frac{1}{2} \sqrt{\frac{\pi}{2}} E_{0}^{2} g_{1} \tau_{p} e^{-(4\Omega^{2} + \omega_{0}^{2}) \tau_{p}^{2}/8} \\ \times \sinh\left(\Omega \omega_{0} \tau_{p}^{2}/2\right) \cdot \sin 2\vartheta_{\text{CE}}.$$
(11)

Equations (10) and (11) yield the following estimate for the maximum values of the response amplitudes:

$$\frac{\Pi_1^{\rm lin}}{E_0 g_0} : \frac{\Pi_1^{\rm nonlin}}{E_0^2 g_1} \sim e^{-\Omega^2 \tau_p^2/4}.$$
 (12)

Considering that for the pump few-cycle pulse $\Omega_0 \tau_p \sim 1$, one gets from Eq. (12) that the nonlinear field coupling enables a significantly increased excitation response. Hence, usage of the linear optical medium seems ineffective, although its principal applicability appears to be of crucial importance.

Another remarkable fact which follows from Eq. (11) is the weak dependence of the oscillator response on the CEP, which holds when $e^{-\Omega^2 \tau_p^2/2} \ll 1$. As long as this condition is fulfilled, the first expression in Eq. (11) converts to

$$\Pi_1 \approx \frac{1}{2} \sqrt{\frac{\pi}{2}} E_0^2 g_1 \tau_p e^{-\omega_0^2 \tau_p^2/8}.$$
 (13)

According to Eqs. (11) and (13) one gets

$$\max_{\vartheta_{\rm CE}} \Pi_2 / \max_{\vartheta_{\rm CE}} \Pi_1 = \sinh \left(\Omega \omega_0 \tau_p^2 / 2 \right) e^{-\Omega^2 \tau_p^2 / 2} \ll 1;$$

that is, the value of the cosine term amplitude Π_2 is negligibly small compared with the sine term amplitude Π_1 , although it is exactly equal to 0 just when ϑ_{CE} is a multiple of $\frac{\pi}{2}$. Therefore, the response of the oscillator with nonlinear field coupling is extremely close to sine regardless of the CEP value, thus making CEP control insignificant in this case.

For unipolar pulse shaping purposes, we should consider summation of the half-cycle pulses from many single oscillators in a certain manner. Thus we arrive at the excitation of spatially extended arrays composed of specifically arranged optical oscillators. It is interesting to note that, placed into a high-Q cavity, such oscillator arrays serve as the active medium of broad-area passive and active optical systems and can exhibit highly complex spatiotemporal behavior [56–60]. When the oscillator array is excited by few-cycle pulses, which can generally have different wavefront forms, the emitted unipolar pulse shape is determined by the variation of the excitation velocity over the array. In the simplest case of linear string excitation at a constant velocity [33,34,45], flap-top pulses are produced with the amplitude and duration the only varying parameters. Depending on the array and excitation geometry, we can tune the spatiotemporal profile of the emitted unipolar pulse in wide limits. In the next sections we examine the possibilities for controlling the unipolar pulse profile, where we deal with oscillators with nonlinear field coupling arranged into spatially extended arrays and excited by few-cycle pulses having different wavefront forms.

III. PLANAR ARRAY OF OSCILLATORS EXCITED BY CURVED INCIDENT WAVES

Given the results in the previous section, we consider the possible means of unipolar pulse generation and control of their spatiotemporal characteristics in spatially extended arrays of oscillators. The optical properties of different nanoemitters and their arrays have been actively investigated in recent years. Particularly, due to the remarkable progress in the development of metallic nanoparticle fabrication techniques [61,62], their ensembles are finding expanding applications as optical waveguides [63,64], surface enhanced Raman scattering media [65,66], high-quality optical resonators [67–69], and antennas and detectors [70]. Various types of particle arrangements were considered for these purposes, providing the possibility of adjusting the resulting optical characteristics according to the medium geometry.

We start with a planar array excited by two few-cycle optical pulses with $T_0/2$ delay and curved incident wavefronts. As stated above, the oscillator has to exhibit a sine response to a single pump pulse, so the total emission of every oscillator is given by two sine functions with a half-period delay which actually represents a half-cycle pulse. Let us first consider a one-dimensional string of oscillators with nonlinear field coupling under the influence of cylindrical waves (see Fig. 3). Oscillator emission is observed at a point on the string midperpendicular far away from the string at distance $r \gg L$ or, alternatively, in the focal plane of the focusing lens parallel to the string. For the sake of simplicity, we assume that the excitation pulses are linearly polarized, with the polarization direction orthogonal to the plane in Fig. 3. Under this assumption, the summation problem is scalar.

Given Eqs. (4)–(7) and assuming $\gamma \ll \omega_0$, the mathematical expression for the generated pulse shape is

$$E(t) = E_0 \sum_{k=0}^{1} \int_0^L \sin[\omega_0 f_x] \Theta[f_x] dx,$$
 (14)

where $f_x = t - \frac{r}{c} - \frac{\sqrt{H^2 + |x-l|^2} - H}{c} - (k-1)T_p$ describes the emission delay from the oscillator located at the point with coordinate x, Θ is the Heaviside step function, and E_0 is the scaling constant. The results of the numerical calculation of the



FIG. 3. A one-dimensional string composed of oscillators with nonlinear field coupling (blue bar) is excited by two successive ultrashort light pulses of cylindrical wavefronts (orange curve). Oscillator emission is observed at a far-distant point on the string midperpendicular.



FIG. 4. Results of the numerical solution of the integral, Eq. (14), for different values of parameters b, $\frac{H}{L}$, and $\frac{l}{L}$: (a) l = 0; (b) b = 20, $\frac{H}{L} = 0.5$.

integral, Eq. (14), for different values of parameters $b = \frac{\omega_0 L}{c}$, $\frac{H}{L}$, and $\frac{l}{L}$ are plotted in Figs. 4(a) and 4(b). The profile of the generated unipolar pulse turns to be monotonically decreasing from its highest level at the leading edge to the lowest at the trailing edge. In this one-dimensional configuration, this shape results from the fact that the intersection point of the exciting wavefront moves along the string at a superluminal velocity varying along the string due to wavefront curvature. The final shape asymmetry is determined by this excitation velocity decreasing along the string, thus leading to pulse amplitude decay towards the trailing edge.

Figure 4(a) was obtained when the wavefront center was placed right above one of the string ends. In this case excitation wavefronts cross the string at the nearest end at zero angle, resulting in an infinite instantaneous excitation velocity and an abrupt jump in the pulse amplitude near the leading edge. To make the pulse profile more uniform, one can shift the wavefront center off to the side. Figure 4(b) illustrates the unipolar pulses obtained in this way. The pulse shape is now determined by the range of angle values at which the exciting pulses cross the string over its length. When shifting the wavefront center farther apart, this angle range decreases, leading to smoothing of the pulse profile.

To obtain another shape of the output pulse, we can introduce additional degrees of freedom meaning to get varying number of excited oscillator growth rate interfering with the wavefront curvature effects. It will naturally be provided if we turn to two-dimensional geometry and consider a circular disk composed of oscillators and excited by spherical incident wavefronts (see Fig. 5). For the sake of simplicity, we assume that the excitation pulses are linearly polarized, with the polarization direction parallel to the circle plane. Array emission is considered with measurement at a point on the symmetry axis far away from the disk at distance $r \gg R$ or, alternatively, on the focal plane of the focusing lens parallel to the circle plane.

The mathematical expression for the pulse shape in this case as

$$E(t) = E_0 \sum_{k=0}^{1} \int_0^R \sin[\omega_0 f_\rho] \Theta[f_\rho] \cdot 2\pi\rho d\rho, \qquad (15)$$

where
$$f_{\rho} = t - \frac{r}{c} - \frac{\sqrt{H^2 + \rho^2} - H}{c} - (k - 1)T_p$$
.



FIG. 5. Circular disk composed of oscillators (blue region) is excited by two successive few-cycle light pulses with spherical wavefronts (orange region). The medium radiation is observed at a point on the symmetry axis far away from the disk at distance r.

In contrast to the linear array, the observed unipolar pulse, Eq. (15), now has a profile monotonically increasing with time (see Fig. 6; $b = \frac{\omega_0 R}{c}$). Even though the excitation wave moves along the array at a gradually decreasing velocity, the number of excited oscillators increases more rapidly, thus making the pulse shape asymmetric towards the trailing edge.

It should be noted that taking into account the arbitrary polarization direction of the incident pulses can also provide plenty of alternatives for pulse shaping. Therefore, the polarization degrees of freedom seem to have great potential in this context and deserve special consideration, which is outside the scope of the current paper.

IV. CIRCULAR ARRAY OF OSCILLATORS EXCITED BY PLANE INCIDENT WAVES

Another practically relevant way to excite the circular array implies its excitation by pulses with plane-incident wavefronts. We consider once again two few-cycle optical pulses with $T_0/2$ delay possessing plane wavefronts and propagating at an arbitrary angle β to the circle plane. This means that the



FIG. 6. Results of the numerical solution of the integral, Eq. (15), for different values of parameter b; $\frac{H}{R} = 1$.



FIG. 7. A circular string of oscillators (blue circle) is excited by two successive few-cycle light pulses having plane wavefronts and propagating at an angle β with the circle plane. Radiation from the medium is observed at a point on the *z* axis far away from the circle at distance *r*.

excitation fronts move on the circle plane at velocity $V = c/\sin\beta$. The electric field is measured as before at a far distant point at the considerable distance *r* from the circle, $r \gg R$, or on the focal plane of the focusing lens parallel to the circle plane.

As the first step, we take the oscillators arranged along a circular string of radius R (see Fig. 7). The shape of the emitted pulse is now expressed as

$$E(t) = E_0 \sum_{k=0}^{1} \int_0^{2\pi} \sin[\omega_0 f_{\varphi}] \Theta[f_{\varphi}] d\varphi, \qquad (16)$$

where $f_{\varphi} = t - \frac{r}{c} - \frac{R(1 - \cos \varphi)}{V} - (k - 1)T_p$ describes the emission delay from the oscillator located at a point with polar angle φ . The results of the numerical calculation of the integral, Eq. (16), for different values of the dimensionless parameter $b = \frac{\omega_0 R}{V} = \frac{\omega_0 R}{c} \sin \beta$ are plotted in Fig. 8(a). The resulting unipolar pulse has a symmetric but strongly nonuniform profile with a well-pronounced concave shape. This feature is naturally expected to take place since the medium geometry configuration is nonlinear and the intersection point thus moves along the string at varying velocity $V/|\sin \varphi|$. Indeed, the intersection-point velocity has maximum values at $\varphi = 0$ and $\varphi = \pi$, resulting in field jumps near the leading and trailing edges of the pulse. This is why the generation of a rectangular pulse does not occur in this case. With decreasing angle β and thus parameter b, the overall duration of the generated unipolar pulse properly decreases, as it takes less time for the excitation pulses to get through the whole circle; the pulse amplitude at the same time correspondingly increases. In the limiting case where the incident wavefronts are parallel to the circle plane $b \rightarrow 0$, all the particles radiate in phase, thus



FIG. 8. Results of the numerical solution of the integral, Eq. (16), for different values of parameter *b*: (a) with the uniform angular density of the oscillators; (b) with the angular density varying as $N(\varphi) = |\sin \varphi|$.

producing a single half-cycle pulse multiplied by the number of oscillators.

To smoothen the pulse shape, we may take the particle density varying along the circle in a certain manner. With $N(\varphi)$ the angular density of the oscillator distribution, Eq. (16), generalizes to become

$$E(t) = E_0 \sum_{k=0}^{1} \int_0^{2\pi} \sin[\omega_0 f_{\varphi}] \Theta[f_{\varphi}] N(\varphi) d\varphi.$$
(17)

Figure 8(b) shows an important example of such a distribution, (17), for $N(\varphi) = |\sin \varphi|$ with the production of rectangular-shaped unipolar pulses. Its duration increases with the increase in *b*, while the amplitude correspondingly decreases, thus keeping the whole pulse area constant.

Finally, we examine a circular disk-shaped array of oscillators analogous to the geometry in the previous section. The equation for the pulse shape is

$$E(t) = E_0 \sum_{k=0}^{1} \int_0^{2R} \sin[\omega_0 f_x] \Theta[f_x] \cdot \\ \times 2\sqrt{1 - \left|\frac{x}{R} - 1\right|^2} d\left(\frac{x}{R}\right),$$
(18)

where $f_x = t - \frac{r}{c} - \frac{x}{V} - (k-1)T_p$ and the x axis goes along the diameter through the disk center.

Figure 9 shows the results of the numerical calculation of the integral, Eq. (18), for different values of the dimensionless parameter $b = \frac{\omega_0 R}{V} = \frac{\omega_0 R}{c} \sin \beta$. The resulting unipolar pulse has a convex profile in this case. Its duration and amplitude may be inversely varied according to the parameter *b*.

V. CONCLUSION

We have studied theoretically the optical response of a resonant medium with an essentially nonlinear field coupling which is excited by few-cycle pump pulses. The medium has been shown to exhibit a specific response depending on the field coupling nonlinearity, oscillator geometry, and CEP of the



FIG. 9. Results of the numerical solution of the integral, Eq. (18), for different values of parameter *b*.

excitation pulse. Relying on the response specifics, we elaborate the concept of half-cycle pulse generation where the oscillator is influenced by a pair of pulses with the proper time delay, so that the first pulse initiates the half-cycle pulse response and the second one stops it at a certain moment. A general criterion is derived allowing us to determine the applicability of this method to the arbitrary nonlinearity of field coupling.

The proposed method allows us to produce ultrashort unipolar pulses directly from a resonant medium without the use of a complicated technique for coherent waveform synthesis. Since the only restriction we impose on the medium resonant frequency ω_0 is that its corresponding period be much larger than the excitation pulse duration, our approach is applicable for the controllable generation of half-cycle pulses over a wide frequency range. Therefore, the method can be most easily used in the terahertz and midinfrared ranges. However, potential extension of the proposed approach to the femtosecond and subfemtosecond ranges seems possible.

To study the possibility of pulse shaping in our scheme, we have considered emission from spatially extended arrays of different geometry as well as different wavefront forms of the excitation pulses. We have studied in detail several particular cases, namely, when a planar array of oscillators is excited by an incident wave with a curved front and the case of a circular array excited by a plane incident wave. It is shown that modification of the oscillator density and excitation wavefront allows the generation of unipolar pulses with a high variability of pulse shapes: asymmetric ones with the maxima at the leading or trailing edges and with monotonous increase or decrease towards the opposite edge and symmetric ones with either a concave, a convex, or a rectangular shape.

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- K. Ramasesha, S. R. Leone, and D. M. Neumark, Annu. Rev. Phys. Chem. 67, 41 (2016).
- [2] L.-Y. Peng, W.-Y. Jiang, J.-W. Geng, W.-H. Xiong, and Q. Gong, Phys. Rep. 575, 1 (2015).
- [3] M. F. Kling and M. J. J. Vrakking, Annu. Rev. Phys. Chem. 59, 463 (2008).
- [4] T. Pfeifer, M. J. Abel, P. M. Nagel, A. Jullien, Z.-H. Loh, M. Justine Bell, D. M. Neumark, and S. R. Leone, Chem. Phys. Lett. 463, 11 (2008).
- [5] X. Zhang and R. R. Jones, Phys. Rev. A 73, 035401 (2006).
- [6] A. Wetzels, A. Gürtler, L. D. Noordam, F. Robicheaux, C. Dinu, H. G. Muller, M. J. J. Vrakking, and W. J. van der Zande, Phys. Rev. Lett. 89, 273003 (2002).
- [7] C. Raman, C. W. S. Conover, C. I. Sukenik, and P. H. Bucksbaum, Phys. Rev. Lett. 76, 2436 (1996).
- [8] X. Zhang and R. R. Jones, New J. Phys. 11, 105050 (2009).
- [9] R. R. Jones, Phys. Rev. Lett. 76, 3927 (1996).
- [10] T. J. Bensky, M. B. Campbell, and R. R. Jones, Phys. Rev. Lett. 81, 3112 (1998).
- [11] C. O. Reinhold, J. Burgdörfer, M. T. Frey, and F. B. Dunning, Phys. Rev. A 54, R33 (1996).
- [12] B. Rau, T. Tajima, and H. Hojo, Phys. Rev. Lett. 78, 3310 (1997).
- [13] H. Hojo, B. Rau, and T. Tajima, Nucl. Instrum. Methods Phys. Res., Sec. A **410**, 509 (1998).
- [14] G. Orlando, P. P. Corso, E. Fiordilino, and F. Persico, J. Mod. Opt. 56, 1761 (2009).

- [15] Y. Pan, S.-F. Zhao, and X.-X. Zhou, Phys. Rev. A 87, 035805 (2013).
- [16] L. Feng and H. Liu, Opt. Commun. **348**, 1 (2015).
- [17] R. M. Arkhipov, M. V. Arkhipov, I. V. Babushkin, A. Demircan, U. Morgner, and N. N. Rosanov, Opt. Lett. 41, 4983 (2016).
- [18] R. M. Arkhipov, M. V. Arkhipov, I. V. Babushkin, and N. N. Rosanov, Opt. and Spectr. 121, 758 (2016).
- [19] M. Th. Hassan, A. Wirth, I. Grgura, A. Moulet, T. T. Luu, J. Gagnon, V. Pervak, and E. Goulielmakis, Rev. Sci. Instrum. 83, 111301 (2012).
- [20] C. Manzoni, O. D. Mücke, G. Cirmi, S. Fang, J. Moses, S.-W. Huang, K.-H. Hong, G. Cerullo, and F. X. Kartner, Laser Photon. Rev. 9, 129 (2015).
- [21] T. Bartel, P. Gaal, K. Reimann, M. Woerner, and T. Elsaesser, Opt. Lett. **30**, 2805 (2005).
- [22] K. Reimann, Rep. Prog. Phys. 70, 1597 (2007).
- [23] I. Babushkin, S. Skupin, and J. Herrmann, Opt. Express 18, 9658 (2010).
- [24] E. A. Stepanov, A. A. Lanin, A. A. Voronin, A. B. Fedotov, and A. M. Zheltikov, Phys. Rev. Lett. 117, 043901 (2016).
- [25] Y. Gao, T. Drake, Z. Chen, and M. F. DeCamp, Opt. Lett. 33, 2776 (2008).
- [26] H.-C. Wu and J. Meyer-ter-Vehn, Nature Photon. 6, 304 (2012).
- [27] V. V. Kozlov, N. N. Rosanov, C. DeAngelis, and S. Wabnitz, Phys. Rev. A 84, 023818 (2011).
- [28] V. P. Kalosha and J. Herrmann, Phys. Rev. Lett. 83, 544 (1999).

- [29] X. Song, W. Yang, Z. Zeng, R. Li, and Z. Xu, Phys. Rev. A 82, 053821 (2010).
- [30] X. Song, Z. Hao, M. Yan, M. Wu, and W. Yang, Laser Phys. Lett. 12, 105003 (2015).
- [31] E. M. Belenov, A. V. Nazarkin, and I. P. Prokopovich, JETP Lett. 55, 218 (1992).
- [32] E. M. Belenov, P. G. Kryukov, A. V. Nazarkin, and I. P. Prokopovich, JETP 78, 15 (1994).
- [33] R. M. Arkhipov, Opt. and Spectr. 120, 756 (2016).
- [34] R. M. Arkhipov, M. V. Arkhipov, I. Babushkin, P. A. Belov, and Yu. A. Tolmachev, Laser Phys. Lett. 13, 046001 (2016).
- [35] R. M. Arkhipov, A. V. Pakhomov, I. V. Babushkin, M. V. Arkhipov, Yu. A. Tolmachev, and N. N. Rosanov, J. Opt. Soc. Am. B 33, 2518 (2016).
- [36] A. M. Weiner, Opt. Commun. 284, 3669 (2011).
- [37] Y. Silberberg, Annu. Rev. Phys. Chem. 60, 277 (2009).
- [38] D. Goswami, Phys. Rep. 374, 385 (2003).
- [39] G. Tearney, M. Brezinski, B. Bouma, S. Boppart, C. Pitris, J. Southern, and J. Fujimoto, Science 276, 2037 (1997).
- [40] A. Wirth, M. T. Hassan, I. Grguras, J. Gagnon, A. Moulet, T. T. Luu, S. Pabst, R. Santra, Z. A. Alahmed, A. M. Azzeer, V. S. Yakovlev, V. Pervak, F. Krausz, and E. Goulielmakis, Science 334, 195 (2011).
- [41] S.-W. Huang, G. Cirmi, J. Moses, K.-H. Hong, S. Bhardwaj, J. R. Birge, L. J. Chen, E. Li, B. J. Eggleton, G. Cerullo, and F. X. Kärtner, Nature Photon. 5, 475 (2011).
- [42] S.-H. Chia, G. Cirmi, S. Fang, G. M. Rossi, O. D. Mucke, and F. X. Kärtner, Optica 1, 315 (2014).
- [43] J. A. Cox, W. P. Putnam, A. Sell, A. Leitenstorfer, and F. X. Kärtner, Opt. Lett. 37, 3579 (2012).
- [44] G. Krauss, S. Lohss, T. Hanke, A. Sell, S. Eggert, R. Huber, and A. Leitenstorfer, Nature Photon. 4, 33 (2010).
- [45] R. M. Arkhipov, I. Babushkin, M. K. Lebedev, Yu. A. Tolmachev, and M. V. Arkhipov, Phys. Rev. A 89, 043811 (2014).
- [46] A. V. Pakhomov, R. M. Arkhipov, I. V. Babushkin, N. N. Rosanov, and M. V. Arkhipov, Laser Phys. Lett. 13, 126001 (2016).
- [47] P. Ginzburg, A. Krasavin, Y. Sonnefraud, A. Murphy, R. J. Pollard, S. A. Maier, and A. V. Zayats, Phys. Rev. B 86, 085422 (2012).
- [48] N. Berkovitch, P. Ginzburg, and M. Orenstein, J. Phys. Condens. Matter 24, 073202 (2012).

- [49] Y. Qiao, F. Polzer, H. Kirmse, E. Steeg, S. Kühn, S. Friede, S. Kirstein, and J. P. Rabe, ACS Nano 9, 1552 (2015).
- [50] D. Savateeva, D. Melnikau, V. Lesnyak, N. Gaponik, and Y. P. Rakovich, J. Mater. Chem. 22, 10816 (2012).
- [51] Y. Zhang, J. Zhang, S.-X. Wu, and and C.-S. Yu, Int. J. Quantum. Info. 13, 1550053 (2015).
- [52] L. Kouwenhoven, Science 268, 1440 (1995).
- [53] W. G. Van der Wiel, S. De Franceschi, J. M. Elzerman, T. Fujisawa, S. Tarucha, and L. P. Kouwenhoven, Rev. Mod. Phys. 75, 1 (2003).
- [54] S. A. Akhmanov and S. U. Nikitin, *Physical Optics* (Oxford University Press, Oxford, UK, 1997).
- [55] J. W. Nibler and J. J. Yang, Annu. Rev. Phys. Chem. 38, 349 (1987).
- [56] I. V. Babushkin, N. A. Loiko, and T. Ackemann, Phys. Rev. A 67, 013813 (2003).
- [57] N. A. Loiko and I. V. Babushkin, J. Opt. B 3, S234 (2001).
- [58] I. V. Babushkin, N. A. Loiko, and T. Ackemann, Phys. Rev. E 69, 066205 (2004).
- [59] A. V. Pakhomov, N. E. Molevich, A. A. Krents, and D. A. Anchikov, Opt. Commun. 372, 14 (2016).
- [60] I. V. Babushkin, Y. A. Logvin, and N. A. Loiko, J. Exp. Theor. Phys. 90, 133 (2000).
- [61] U. Kreibig and M. Vollmer, Optical Properties of Metal Clusters (Springer-Verlag, Berlin, 1995).
- [62] D. L. Feldheim and C. A. Foss, *Metal Nanoparticles: Synthesis, Characterication, and Applications* (Marcel Dekker, New York, 2001).
- [63] S. A. Maier, P. G. Kik, H. A. Atwater, S. Meltzer, E. Harel, B. E. Koel, and A. A. G. Requicha, Nat. Mater. 2, 229 (2003).
- [64] M. Quinten, A. Leitner, J. R. Krenn, and F. R. Aussenegg, Opt. Lett. 23, 1331 (1998).
- [65] M. Moskovits, Rev. Mod. Phys. 57, 783 (1985).
- [66] G. Bachelier and A. Mlayah, Phys. Rev. B 69, 205408 (2004).
- [67] A. Alu, A. Salandrino, and N. Engheta, Opt. Express 14, 1557 (2006).
- [68] D. S. Citrin, J. Opt. Soc. Am. B 22, 1763 (2005).
- [69] A. L. Burin, Phys. Rev. E 73, 066614 (2006).
- [70] R. W. King, G. J. Fikioris, and R. B. Mack, *Cylindrical Antennas and Arrays* (Cambridge University Press, Cambridge, UK, 2002).