Fourier Synthesization of Optical Pulses and “Polar” Light

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It is shown that the direct Fourier synthesization of light beams allows one to create polarity-asymmetric waves, which are able, in the process of nonlinear interaction with a medium, to break its inversion symmetry. As a result, these "polar" waves may show the effect of optical rectification in nonlinear centrosymmetric media by generating light-induced dc electric polarization. At the same time, the waves of this type, due to their unusual symmetry properties, can be used for detecting the direction and sign of a dc electric field applied to the medium. The prospects of application of polar waves to data recording and processing are discussed.

I. INTRODUCTION

The problem of generation of ultrashort light pulses has drawn considerable attention in recent years. This attention is a result of unprecedented (subfemtosecond) temporal resolution and giant intensities achieved with their use, a potential possibility to significantly increase the data transmission and processing rates, and the unexpected successes in application of comb spectra of ultrashort light pulses in metrology of optical frequencies (see, e.g., the review [1]). Since almost all existing methods for producing ultrashort pulses involve generation of equidistant pulse trains, the generated emission is also described by a discrete set of equidistant harmonics in the spectral representation. In practice, this is achieved either due to proper spectral characteristics of the emitting system, e.g., an equidistant cavity-mode structure in laser mode locking [2], or due to a natural temporal periodicity of the physical process, as, e.g., in the well-known experiments on generation of comb spectra under high-power photoexcitation of noble gas atoms [3-4].
To address the problem of shortening the light pulse duration, Hansch proposed in 1990 an idea of a direct synthesization of a wave with the necessary field profile from a corresponding set of Fourier harmonics [5]. Indeed, a train of ultrashort pulses may be obtained by superposing several harmonics ($\omega_0$,$2\omega_0$,$3\omega_0$,...$n\omega_0$), with the highest of them ($n\omega_0$) controlling the pulse duration and the lowest ($\omega_0$) controlling the repetition rate. The “light” pulses synthesized in this way are close in shape to a half-cycle of the highest harmonic. The phases of the light beams being combined are supposed to be sufficiently well defined, with the phases of different harmonics synchronized due to their common origin from the same master oscillator or due to phase-locking of different oscillators (as in [3]).

Figure 1 shows as an illustration the field profile of two hypothetical waves synthesized from 30 harmonics of the CO$_2$ laser (wave b only from odd harmonics). The highest harmonic corresponds to the wavelength $\sim$ 0.35 $\mu$m. The pulses thus obtained are shorter than 1 fs and are spaced in time by $\sim$ 35 fs. Implementation of such waves is an interesting and difficult (as far as we know, so far unsolved) technical problem. In this paper, however, we want to call attention to simple and unusual properties of the synthesized waves which may be easily revealed even with a minimal number of harmonics and may be interesting both from the cognitive and practical points of view.

II. POLAR LIGHT WAVES AND THEIR PROPERTIES

The two waves shown in Fig. 1 differ qualitatively from each other. In one of them (Fig. 1a) all the pulses are unipolar, while in the other (Fig. 1b), they are bipolar. The latter wave, in spite of the unusual shape of the oscillations, still holds the main symmetry of a conventional monochromatic wave with respect to sign reversal of the field in each half-cycle. In the first (“unipolar”) wave, the polar symmetry of the oscillations is broken.

It is evident that in the processes of linear interaction between the light and a medium for which the superposition principle is obeyed the ”unipolarity” of the wave cannot be revealed because this property is distinguished only by a certain phase matching of the constituent
harmonics. However, in the processes of a nonlinear interaction, the wave of this type may show unique properties which are absent in its constituent harmonics.

First of all, to describe adequately the physical asymmetry of the unipolar wave field, the polarization vector of the wave should become polar. It means that to describe polarization state of a linearly-polarized wave one would have to specify not only the azimuth of the polarization plane but also the direction of its “polarity” (say, “up” or “down”). Two beams with “parallel” and “anti-parallel” polarizations are two physically distinguishable situations. In particular, such beams will interfere differently with each other. Figure 2 shows, as an example, a possible shape of the polar wave synthesized from six harmonics (the wave analyzed in [5]) and the profile of the interference pattern for the two waves of this kind with parallel and anti-parallel polarizations. As is seen from the figure, these two profiles are essentially different and, in particular, completely destructive interference is possible only for the interfering beams with antiparallel polarizations. The interference pattern of this kind makes it possible to examine visually the peculiar profile of the synthesized wave. Note, however, that such an observation should be implemented with an appropriate broad-band detector (e.g., an IR viewer), capable of detecting all spectral harmonics of the interfering waves.

The most important property of the polar waves, in our opinion, is their ability to induce, in a centrosymmetric nonlinear medium, static electric polarization or dc electric current, i.e., to induce a perturbation with the symmetry of polar vector (see, e.g., [6]). It is easy to show that a result of combining the nonlinear polarizability of a centrosymmetric medium \( P = \alpha E + \beta E^3 + \ldots \) with the polar asymmetry of the wave \( E \) is that the integral of the polarization \( P \) over a period becomes nonzero. For noncentrosymmetric media (with quadratic nonlinearity), the effect of this kind is known as optical rectification [7]. In the case under consideration, the inversion symmetry of the problem is lifted by the light wave itself. For a polar wave, the direction of the induced dipole vector will depend on the azimuth of the plane of polarization of the wave and will be evidently inverted upon rotation of the polarization plane through \( 180^\circ \). Such a linearly polarized wave is capable of bringing into
the medium an electric dipole moment exactly like a circularly polarized wave is capable of bringing into the medium a magnetic moment. Unlike other methods of light-induced charge separation in centrosymmetric media (as, e.g., in the photorefractive effect \[8\], where the polar vector that lifts the inversion symmetry is the gradient of the optical excitation density), the use of polar waves allows one, in essence, to locally create, in the illuminated spot, an effective electric field with the magnitude and direction controlled by the intensity and polarization of the light beam and thus to polarize the medium in this area.

It seems evident that the polarization induced with polar light can be detected (read out) using the same kind of light. In other words, properties of a polarized medium (again, in the nonlinear regime of interaction) will be different for the beams with opposite directions of the polarization vectors. Therefore, the polar wave that probes the polarized medium is capable of detecting the sign of its polarization or the sign of the electric field applied to the medium. It may be easily shown also that a usual monochromatic light wave after passing through a polarized medium (e.g., placed in an external electric field) acquires a "polarity", with the sign of this polarity (i.e., the type of phase matching of the harmonics) being determined by the sign of polarization of the medium, which can thus be detected. We will consider these issues in a more rigorous way elsewhere.

It should be emphasized that to obtain such polar light there is no need to combine a great number of harmonics and to design exotic waves like those presented in Fig. 1. Figure 3 shows profiles of the polar waves synthesized from two and three harmonics. As is seen, even for two harmonics, the field amplitudes of opposite polarities may differ by a factor of two. Therefore, it may be expected that even the simplest combinations of two harmonics will be fairly efficient for observation of the above effects. Moreover, one precedent of observation of the effect of this kind, when the polar wave was formed and revealed in the same medium, is known. This is optical frequency doubling in a glass optical fiber \[9\], where the action of two coherent harmonics of laser light polarized the medium within the regions of the order of the phase-matching length and, thus, allowed the normally forbidden second harmonic generation.
III. NUMERICAL ESTIMATES

Synthesized polar light waves provide fundamentally new possibilities for perturbing and probing nonlinear media. The possibility to locally polarize the medium with a light beam and to subsequently read-out this polarization is very attractive from the viewpoint of practical application of polar light, e.g., for data recording and processing. This approach, however, has a disadvantage: the medium can distinguish polarity of the wave only when the process of interaction is nonlinear. Note that this relates not only to data recording (which is inevitably nonlinear), but also to read-out. For this reason, it is evident that application of polar light for light-induced polarization of the medium or for diagnostics of such a polarization may become efficient, either in the case of high cubic nonlinearity of the medium, or in the case of sufficiently high sensitivity of the measurements. Let us consider this point in more detail.

As for media with high cubic nonlinearity, one may rely on further progress in the area of synthesis of new nonlinear materials based on polymers with conjugated bonds where significant successes have been achieved in recent years (see, e.g., [10,11]). As can be easily shown, the strength of the electric field $E_0$ produced in the medium with cubic susceptibility $\chi^{(3)}$ by a coherent superposition of two harmonics with approximately equal amplitudes $E$ is given, to within an order of magnitude, by $E_0 \sim \chi^{(3)}E^3$. Using the scale of cubic nonlinearities achieved nowadays in polymers with conjugated bonds, $\chi(3) \sim 10^{-10}$ CGSE (see, e.g., [12]), one can estimate that the amplitudes of the light wave harmonics needed to create in the medium an effective dc field of about 100 V/cm will lie in the range of $10^3$ CGSE. This corresponds to the light power density of about $10^9$ W/cm² or $\sim 10$ W through an area of 1 $\mu$m². It should be borne in mind, however, that we made this estimate for the most unfavorable case of superposition of two harmonics, when the achievable polarity of the wave (which may be specified by the ratio of amplitudes of the field vector in two opposite directions) is the smallest. With increasing number of harmonics and increasing polar asymmetry of the wave, the above estimate will become much more favorable. It is
possible also that considerable improvement may be obtained by using semiconductors and quantum-confined heterostructures with giant nonlinear susceptibilities (see, e.g., [13]). For these cases, however, the relevant estimates require a more serious analysis of the mechanism of polarizability of the medium in a bichromatic field.

As applied to the problem of detecting the electric polarization of the medium with the polar-light waves, the situation appears much more attractive. The point is that the spectral composition of polar light which has passed through a polarized (by an external electric field) nonlinear mediumproves to be different for the cases of parallel and antiparallel orientations of the light and medium polarization vectors. For simplicity, we will again consider superposition of only two harmonics. From the viewpoint of the measurement technique, the most interesting point is that the amplitudes of these two harmonics appear to be odd functions of the beam polarity. In other, more exact, words, when the sign of polarity of each of the beams is inverted, the amplitude increment of each harmonic also changes its sign. Beam polarity, in turn, may be easily modulated by transmitting the beam through a medium with a modulated (e.g., by an applied field) refractive index. Due to dispersion of the medium, the difference between its refractive indices for the two frequencies spaced by one octave ($\omega$ and $2\omega$) will also be modulated and, for a properly chosen modulation amplitude and thickness of the medium, the beam polarity will appear to be modulated as well. As a result, as noted above, the amplitudes of the constituent harmonics of the light beam that has passed through the polarized medium under study will be modulated.

It is important that in an experiment of this kind, when the beam is modulated in such a delicate fashion, the sensitivity of measurements may easily reach its shot-noise limit. As can be shown for this case, relative modulation of the fundamental harmonic, will be given by the factor $6\chi^{(3)}E_0E/\chi^{(1)}$, where $\chi^{(1)}$ and $\chi^{(3)}$ are the linear and cubic susceptibilities of the medium, respectively, $E_0$ is the dc field applied to the medium, and $E$ is the field of the light wave. Now, to estimate the limiting sensitivity of such a technique, we have to compare this value with photocurrent fluctuations of the detector, which registers the same light with the amplitude $E$. As a result, the equation that connects the strength of the
applied dc electric field with the field amplitude of the polar wave capable of detecting the corresponding polarization of the medium will have the form.

$$6 \chi^{(3)} E_0 E / \chi^{(1)} \sim 2 \cdot 10^{-11} / (E \cdot \sqrt{S})$$

Here $S$ is the beam cross section in the medium. If we again take cubic susceptibility $\chi^{(3)} \sim 10^{-10}$ CGSE and the dc field applied to the medium equal to 30 CGSE ($\sim 10 \text{ kV/cm}$), it appears that this polarization of the medium can be detected, e.g., in a 3-mW light beam with a cross section of 1 mm$^2$ (the power density 0.3 W/cm$^2$) or in a 3-$\mu$W light beam with a cross section of 1 $\mu$m$^2$ (300 W/cm$^2$). As before, sensitivity may be further increased by increasing the number of harmonics that form the polar light beam. It is noteworthy that in the above experiment with detection of polarization of the medium, modulation of the light beam polarity can be replaced by modulation of the sign of the field applied to the medium. Of course, the result will be the same. However, the possibility of such an observation indicates that with polar Light, a centrosymmetric medium could exhibit a linear electrooptic effect.

**IV. CONCLUSIONS**

In this paper, we call attention to the fact that polar waves which may be easily synthesized from a few Fourier harmonics of a fundamental frequency may produce, in a nonlinear medium, a perturbation with the symmetry of a polar vector (as, e.g., the electric field or current). The action of such light beams upon a nonlinear medium is equivalent to local application of an electric field to the illuminated spot. In our opinion, this effect is primarily interesting from the point of view of symmetry, as a way to control and determine the sign of the polar vector using a probe light beam. The estimates made above show that the method of diagnostics of polarization of the medium using polar light may be fairly efficient even for a medium with a small cubic nonlinearity. The authors gratefully acknowledge useful discussions with S.G. Przhibel’skii.
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FIG. 1. The field profiles of two hypothetical waves synthesized from 30 harmonics of the $CO_2$ laser radiation ($\lambda = 10.6 \mu m$); a - unipolar wave and b - bipolar wave. The wave b is formed only from odd harmonics of the fundamental frequency.
FIG. 2. The field profile of the wave synthesized from six Fourier harmonics (a) and the pattern of interference of two such waves with parallel (b) and antiparallel (c) polarizations.

FIG. 3. The field profile of the polar waves synthesized from two (a) and three (b) harmonics.