Extension of holography to multifrequency fields

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Holographic processes are generalized to two cases: (a) the recording of the interference pattern of mutually coherent fields of several different frequencies and (b) recording in the form of perturbations of higher-order optical susceptibilities. The relationship with second-harmonic generation in optical waveguides is discussed.

Österberg and Margulis[1] have reported coupling the output from a picosecond-range, periodic-pulse neodymium laser into a single-mode optical waveguide. After some 2 or 3 h of this process, the incident light ($\lambda = 1.06\ \mu$m) began to be converted into the second harmonic ($\lambda = 0.53\ \mu$m) with an efficiency $\sim 10\%$, nearly independent of the length of the waveguide (from 0.5 to 10 m).

One of our purposes in the present letter is to offer a mechanism for this process. The hypothesis is that a second harmonic $E_{2\omega}(\mathbf{R})$ which arises at the entrance as the result of some fluctuation records a hologram of the quadratic polarizability, $\delta\chi^{(2)}(\mathbf{R}) \propto E_{2\omega}(\mathbf{R}) E^*_\omega(\mathbf{R}) E^*_\omega(\mathbf{R})$ (more on this below). The incident field E_ω itself then reads this hologram in a two-photon manner, becoming converted into the amplified second harmonic: $\delta E_{2\omega}(\mathbf{R}) \propto \delta\chi^{(2)} E^2_\omega(\mathbf{R}) \propto |E_\omega(\mathbf{R})|^4 E_{2\omega}(\mathbf{R})$. Remarkably, the matching condition is satisfied automatically here.

In the classical arrangements for holography[2-4] with ordinary (single-photon) absorption of light, information on the intensity interference pattern $A(\mathbf{R})B^*(\mathbf{R})$ of two waves $A(\mathbf{R})\exp[-i\omega_A t + i\varphi_A(t)]$ and $B(\mathbf{R})\exp[-i\omega_B t + i\varphi_B(t)]$ is recorded. It is usually necessary to use equal frequencies ($\omega_A = \omega_B$) and mutually coherent waves $\varphi_A(t) = \varphi_B(t)$ in order to avoid "painting over" the pattern. In this letter we discuss the possibilities of extending holography to multifrequency fields.

1. We first consider the use of several frequencies in readout. We assume that the interference pattern of two plane waves, $AB^*\exp[i(\mathbf{k}_A - \mathbf{k}_B)\mathbf{R}]$, which is recorded by means of ordinary single-photon absorption, excites in the medium not only a modulation $\delta\epsilon(\mathbf{R})$ but also a modulation of the quadratic optical polarizability of the medium $\delta\chi^{(2)}(\mathbf{R}) \propto A(\mathbf{R})B^*(\mathbf{R}) + \text{c.c.}$ We can cite some physical mechanisms which might be responsible for $\delta\chi^{(2)}(\mathbf{R})$. During the absorption of light, the local values of the temperature T, the chemical composition c, and the population (N_i) of a particular electronic or vibrational term may vary. If the condition $\chi^{(2)} \neq 0$ holds in the initial medium (e.g., a crystal without an inversion center), then the dependence $\chi^{(2)}(T,c,N_i)$ will give rise to $\delta\chi^{(2)} \propto AB^* + \text{c.c.}$

Furthermore, when light is absorbed in an insulator with impurity centers, the latter may undergo an ionization followed by a spatial separation and buildup of charge (Refs. 5–7, for example). If the rather strong ($\sim 10^4$ V/cm) static electric fields $E_{st}(\mathbf{R}) \propto A(\mathbf{R})B^*(\mathbf{R})$ + c.c. that arise are to be manifested as an "ordinary" modulation $\delta\epsilon_{ik} \propto rE_{st}$—i.e., as a photorefractive effect—the electrooptic coefficient r must be nonzero. However, the appearance of a $\delta\chi^{(2)} \propto \gamma E_{st}$ is permissible in any medium, e.g., an amorphous glass or quartz. Ferroelectric crystals just above the Curie point, where γ is anomalously high, while the opalescence of the medium is only poorly expressed, may be particularly attractive in this connection.

The readout process itself may consist of (for example) the generation of the second optical harmonic of the reconstructing wave, $D\exp(-i\omega_D t + i\mathbf{k}_D\mathbf{R})$, which induces in the medium a dipole moment per unit volume

$$P_i(\mathbf{R}) = \delta\chi^{(2)}_{ijm}(\mathbf{R})D_jD_m \exp(-2i\omega_D t + 2i\mathbf{k}_D\mathbf{R}). \tag{1}$$

For bulk media, the matching condition

$$(2\omega_D/c)^2\epsilon(2\omega_D) = (2\mathbf{k}_D + \mathbf{k}_A - \mathbf{k}_B)^2 \tag{2}$$

must be satisfied for effective excitation of the second harmonic. It turns out to be possible to satisfy condition (2) even if there is a frequency dispersion $\epsilon(2\omega_D) > \epsilon(\omega_D)$, by virtue of the increment $\mathbf{k}_A - \mathbf{k}_B$. During the generation, the sum frequency $\{P \propto \delta\chi^{(2)}(\mathbf{R})CD\exp[i(\mathbf{k}_D + \mathbf{k}_C)\mathbf{R} - i(\omega_C + \omega_D)\mathbf{t}\}$ or the difference frequency $\{P \propto \delta\chi^{(2)}(\mathbf{R})C^*D\exp[i(\mathbf{k}_D - \mathbf{k}_C)\mathbf{R} - i(\omega_D - \omega_C)t]\}$ can be read. A particular case might be a diffraction of the readout wave $D\exp(-i\omega_D t + i\mathbf{k}_D\mathbf{R})$ controlled by a quasistatic electric field $C(t)$; this would be a diffraction by a grating $\delta\chi^{(2)}\mathbf{R} \propto AB^*\exp[i(\mathbf{k}_A - \mathbf{k}_B)\mathbf{R}]$. Yet another example of the use of a spatially periodic distribution of $\chi^{(2)}$ is in phase conjugation in three-wave Bragg mixing (see Ref. 8 and §8.6.2 in Ref. 9).

2. We turn now to the recording of holograms in the case with multiphoton absorption as a medium is illuminated by fields of three frequencies:

$$D(\mathbf{R})\exp(-i\omega_D t), \quad A(\mathbf{R})\exp(-i\omega_A t) \text{ and } B(\mathbf{R})\exp(-i\omega_B t).$$

If the medium (or a given microscopic part of it, say, a molecule) does not have an inversion center, either a single-photon absorption of the field D or a two-photon absorption of the pair of fields A,B can occur on the same $1\rightarrow 2$ transition with $\omega_{21} = \omega_D = \omega_A + \omega_B$. Furthermore, during the simultaneous application of all three fields, the local transition probability is proportional to the quantity

$$|d_{12}D(\mathbf{R})\exp[i\varphi_D(t) - i\omega_D t] + m_{12}A(\mathbf{R})B(\mathbf{R})\exp[i\varphi_A(t) + i\varphi_B(t) - i(\omega_A + \omega_B)t]|^2,$$

where d_{12} and m_{12} are the matrix elements for the single-photon transition and the composite matrix element of the two-photon transition, respectively. In other words, when the condition for multifrequency coherence is satisfied, $(\omega_D - \omega_A - \omega_B)t + \varphi_A(t) + \varphi_B(t) - \varphi_D(t) = \text{const}$, it is possible to record the interference pattern of three fields of different frequencies. This condition is satisfied automatically if the field D is produced in a separate device which generates the sum frequency of the fields A and B or if A and B are obtained from D in a parametric light source. Another possibility is absorption involving two photons ω_A and ω_B under the condition $\omega_{21} = \omega_A - \omega_B$ (of the nature of Raman scattering). For media or molecules lacking an inversion center, there can again be an interference with a single-photon transition $\omega_{21} = \omega_D$. The mechanisms by which the medium then responds to the absorbed energy are approximately the same as in single-photon recording processes.

3. We turn now to the case of the recording of a hologram $\delta\chi^{(2)}(\mathbf{R}) \propto A^*(\mathbf{R})B^*(\mathbf{R})D(\mathbf{R})$ during the application of three mutually coherent pulsed fields A, B, D with $\omega_D = \omega_A + \omega_B$. We assume that the initial medium has an inversion center (e.g., glass) but consists of very small ($\ll \lambda$) clusters or of molecules without an inversion center. The excitation of a given molecule by the interference of the fields is proportional to $d_{12}m_{12}A^*B^*D$. The sign of the product $(d_{12}m_{12})$ is opposite for molecules with oppositely directed polar axes. In a medium which has an inversion center, the distribution of polar axes of the molecules is random, so that the energy evolution averaged over the molecules does not contain an interference term, and we have $\delta\epsilon = 0$. If, however, the excitation leaves a mark on the given molecule and on its surroundings for a substantial time, then the intrinsic quadratic polarizability of the molecule, $\delta\beta^{(2)}$, will change. Since there is no inversion center, this change will again take opposite signs for molecules with oppositely oriented polar axes. Specifically, we find

$$\delta\chi^{(2)} \propto \langle \delta\beta^{(2)} d_{12} m_{12} \rangle \propto A^*(\mathbf{R})B^*(\mathbf{R})D(\mathbf{R}).$$

Remarkably, if the resulting nonlinearity hologram is read out by the same reference waves, $A(\mathbf{R})\exp(-i\omega_A t)$ and $B(\mathbf{R})\exp(-i\omega_B t)$, the polarization at the frequency $\omega_D = \omega_A + \omega_B$ will have a structure $P_D \propto |AB|^2 D(\mathbf{R})$. This structure automatically satisfies the matching condition for the coherent excitation of wave D from the entire volume, regardless of the frequency dispersion and regardless of any possible optical inhomogeneities in the medium. Furthermore, a suitable phase shift could cause the process of recording and readout of a dynamic hologram $\omega + \omega \to \omega$ to occur in a regime of an exponential growth of a weak seed signal at the frequency 2ω, in a manner similar to stimulated hyperscattering of light.[10] We do not rule out the possibility that this is the mechanism for the recently observed generation of the second optical harmonic in optical fibers.[1]

A case that deserves special mention is $\omega_A + \omega_B = \omega_D$, in which the frequency of one of the fields (ω_A) is in the rf range. In this case, the field D is obtained through a modulation of the field B by the field of the rf frequency, ω_A, and a demodulation occurs in the hologram.

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