Laser-pulse-induced frequency chirp in surface second-harmonic generation

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The concept of the self-phase-modulation and related frequency chirp is introduced in the connection of the surface second-harmonic generation, which is induced by a short-duration laser pulse.

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I. INTRODUCTION

The theoretical model of the second-harmonic generation (SHG) in reflection from a surface of nonlinear medium is explained in Bloembergen and Pershan's seminal paper [1], and the experimental technique for the SHG detection is explained, e.g., by Sonnenberg and Hefner [2]. At present there is a diversity of papers on surface SHG. Most of them describe the SHG from a metal surface. Here we refer only to two review articles written by Sipe and Stegeman [3] and Coutaz [4].

In this paper we consider the self-phase-modulation and related frequency chirp in second-harmonic wave, which is generated from a smooth surface of nonlinear medium. The self-phase-modulation is believed to occur when an intense, short-duration, laser pulse is incident the surface.

The self-phase-modulation is familiar from fiber optics [5]. The idea of self-phase-modulation has been successfully applied in the construction of a soliton laser [6] and furthermore in the realization of a high-bit-rate long-distance optical-communication system [7].

II. SELF-PHASE-MODULATION

If we consider the SHG in reflection, as presented in Fig. 1, the power of the second-harmonic wave $P(2\omega)$, which is induced by an intense laser pulse, can be expressed in the formula [8]

$$P(2\omega)=R_{2\omega}P^2(2\omega) \tag{1}$$

in the frequency domain. Such a power is recorded in the direction of the specular reflection provided that the surface roughness is relatively low. The SHG power depends on the square of the incident power $P(\omega)$, where ω is the angular frequency of a monochromatic laser radiation. The nonlinear reflectivity $R_{2\omega}$ depends on the angle of incidence of the laser beam and the complex permittivity and surface nonlinear (second-order) susceptibility of the medium [8].

The description of SHG in time domain is somewhat problematic since the second-order nonlinear polarization of electrons can be nonlocal in time and space [9]. We make the usual assumption that there exists a local relationship between the second-order polarization and the electric field strength. If we record SHG using a phototube and a memory oscilliscope, the SHG signal usually is a bell-shaped function of time for a short-duration laser pulse. Although the detection devices have their own responses on SHG signal it is reasonable to expect that the time dependence of the envelope of incident electric field has an effect on the time evolution of the second-order polarization at the short period when the laser pulse interacts with the electrons. The second-order polarization is expected to be proportional to the square of the incident electric field. Now the strength and the phase of the electric field, radiated with frequency 2ω, should be dependent on the time evolution of the envelope of the incident electric field, $E_i(t)$ where t denotes time.

We may define a nonlinear reflection coefficient,

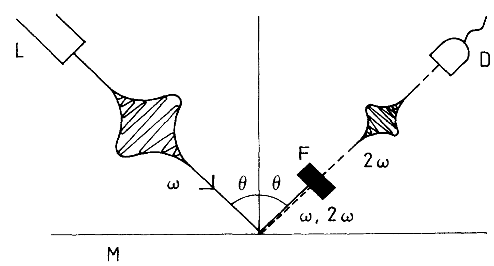

FIG. 1. A schematic diagram for second-harmonic generation. *L*, laser; *M*, nonlinear medium; *F*, filter to block the fundamental frequency; and *D*, detector.

$r_{2\omega}(E_i(t))e^{i\varphi_{2\omega}(E_i(t))}$, which describes the strength $r_{2\omega}$ and phase $\varphi_{2\omega}$, of the second-harmonic electric field. Here we have proposed that $\varphi_{2\omega}$ is changing as a function of $E_i(t)$. The dependence of $\varphi_{2\omega}$ on $E_i(t)$ is nonlinear in the general case. The calculation of $\varphi_{2\omega}$ may be problematic. In order to get some idea about $\varphi_{2\omega}$ we assume the reflection coefficient to be of the form $a(t)E_i(t)e^{i\varphi_{2\omega}(t)}$. In other words $\varphi_{2\omega}$ is assumed to depend linearly in $E_i(t)$. Now we may exploit the concept of analytic signal [10] and make use of Hilbert transform [11] (see also Refs. [12–14]) to obtain

$$\varphi_{2\omega}(t')=-\frac{1}{\pi}\mathrm{P}\int_{-\infty}^{\infty}\frac{\ln[a(t)E_i(t)]}{t-t'}dt , \tag{2}$$

where P denotes the Cauchy principal value. The related frequency chirp is obtained by forming the derivative $d\varphi_{2\omega}/dt'$. In Fig. 2 the chirp of frequency is illustrated [calculated using Eq. (2)] in a case where $E_i(t)$ is assumed to have a Lorentzian shape and $a(t)=\text{const}$. The chirp is strongest when E_i has the maximum value.

Now one may ask when the proposed chirp of frequency could be detected. The answer may not be straightforward since the surface roughness has an effect on the strength of SHG. The microscopical variations in surface affect how the incident field is enhanced by grooves etc. [15]. If we forget the surface quality an estimate is obtained from Heisenberg's principle. If the laser pulse duration is about in the femtosecond range, then a chirp in second-harmonic frequency could be detected in the spectral region of visible light.

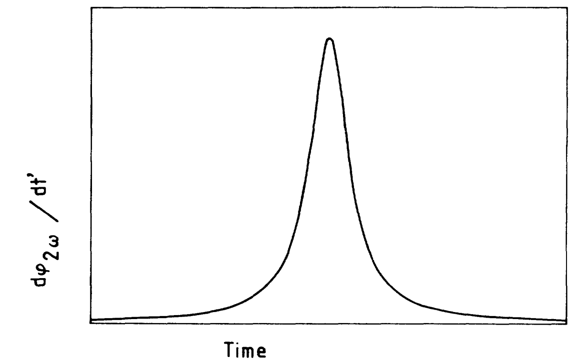

FIG. 2. Frequency chirp of second-harmonic field in a case of Lorentzian envelope of incident field.

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