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Spectrally resolved femtosecond Maker fringes technique

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We present a femtosecond third-harmonic generation Maker fringes technique capable of simultaneously providing the magnitude of the cubic nonlinearity and the refractive index dispersion of optical materials. This technique takes advantage of the high intensity and broad spectral band of femtosecond pulses, but requires the use of a spectrometer to deconvolute the information contained in Maker fringes produced by the broadband light. © 2008 American Institute of Physics.

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The third-harmonic (TH) Maker fringes method has been extensively used in investigations of purely cubic electronic nonlinearities of optical materials. The characterization of such nonlinearities is important to determine if a given material has the prospect for applications in optical devices such as fast switches and modulators. The Maker fringes technique is based on the observation of an oscillatory pattern in the TH generated as a function of the incidence angle on the sample. Usually, when picosecond or longer pulses are employed, the harmonic measurement is carried out with a single detector, such as a photomultiplier. However, femtosecond lasers are readily available nowadays and they are likely to be employed for optical nonlinear characterization due to their high peak intensities. In this case, owing to the broad spectral band of such ultrashort pulses, the oscillatory pattern can be destroyed because of the sample’s dispersion and the superposition of different frequency oscillation patterns produced by different wavelengths within the large bandwidth.

The present work shows how to circumvent this problem by the use of a spectrometer to deconvolute the Maker fringes. As a byproduct of using a broadband spectrum, we show that it is possible to obtain the index of refraction curve, not achievable with longer pulses. As we know, the refractive index is one of the most important properties of an optical material and techniques based directly on the beam deviation and Snell’s law, such as in the Abbe and Pulfrich refractometers, can be employed for its determination. Such methods require precise angle measurement and several monochromatic light sources to characterize the index dispersion. Ellipsometry and interferometry are methods based on different optical phenomena that can also be used for that purpose. Here, we demonstrate that in addition to the cubic nonlinearity, broadband TH generation also provides a way of measuring the index of refraction curve of optical materials.

The experimental setup is the same as the one employed in the traditional Maker fringes technique, with the difference that instead of a single detector, we used a 0.5 nm resolution portable spectrometer to detect the TH signal. The TH generation was achieved by using a wavelength tunable pulse, generated by an optical parametric amplifier pumped by a 1 KHz Ti:sapphire chirped pulse amplifier laser system. This setup produced ~120 fs duration pulses in the infrared range (~10 nm bandwidth), with a typical average power of about 15 mW. The beam was focused onto the sample with a 20 cm focal-length lens and s-polarization was used to generate the TH. The sample was placed in a computer-controlled high-resolution rotational stage. The fundamental and the TH beams were separated by a prism to avoid damage of the spectrometer due to the intense fundamental beam. The TH spectrum was measured for each angular step with the use of the high-resolution spectrometer. Due to the material’s dispersion, the measured TH intensity depicted a spectral modulation. In addition, similarly to the traditional technique, a modulation proportional to the coherence length was observed as the angle was scanned. A TH spectrogram containing wavelengths at the y axis and angles at the x axis was obtained with this procedure. Because the measured TH intensity is convoluted with the spectrometer instrumental response, the fringes visibility depends on the spectrometer resolution. This resolution has to be accounted for in the fitting procedure to deconvolute the signal intensity from the instrumental response.

There are two possible ways of analyzing TH Maker fringes results, either in the time or frequency domains. While the later is traditionally used for long pulses with negligible bandwidth, the time domain analysis is more convenient for femtosecond pulses because it easily enables the introduction of the group velocity dispersion. Later on, a Fourier transform can be applied to provide spectral results. In our case, however, although ultrashort laser pulses are used, the introduction of the spectrometer permits the direct observation of the TH spectral distribution and each Fourier component presents a very small bandwidth. Therefore, the analysis in the frequency domain can be employed provided that we take into account that the TH generation can be non-degenerate within the bandwidth of the fundamental wavelength. Supposing that the index of refraction and the nonlinear susceptibility are nearly constant within the 10 nm bandwidth of the fundamental beam, one can follow the approach of Ref. 2 and write

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where \( l \) is the sample thickness, indices 1 and 2 are, respectively, related to the air and the sample, \( \Delta \varepsilon \) is the dispersion of the dielectric constant and \( f(\theta_i) \) is a correction factor depending on the experimental geometry of light detection. The function \( G(\omega) \) accounts for all frequency combinations and is given as

\[
G(\omega) = \int \int E(3\omega - \omega_2 - \omega_3)E(\omega_2)E(\omega_3)d\omega_2d\omega_3,
\]

where \( \omega_i \) \((i=1,2,3)\) are spectral components of the fundamental beam and \( \omega \) is their average value such that \( 3\omega = \omega_1 + \omega_2 + \omega_3 \). This integral depends on the magnitude and relative phases of the spectral components, but it is intrinsic to the light beam and not to the sample. It can be experimentally determined with a calibration procedure in a well-known sample. The transmittance coefficient \( t_{12}^\omega \) follows directly from Fresnel’s equation and is given as

\[
t_{12}^\omega = \frac{2n_1 \cos \theta_1}{n_1 \cos \theta_1 + n_2^3 \cos \theta_2^\omega},
\]

while the factor \( A \) is obtained from the boundary conditions,

\[
A = \frac{n_2^3 \cos \theta_2^\omega + n_1 \cos \theta_1^\omega}{n_2^3 \cos \theta_2^\omega + \frac{\cos \theta_2^\omega}{n_1^3 \cos \theta_1^\omega}}.
\]

where the angle \( \theta_i \) is given by Snell’s law and corresponds to the angle inside the sample.

The coherence length can be deduced from the spacing between the minima (maxima) of the Maker fringes while the nonlinear coefficient is related to the peak amplitudes of the oscillations. To fit the measured data we have to consider the broad spectrum of both pump and TH beams, convoluting Eq. (1) with the spectrometer instrumental response. Thus, we obtain the coefficients of the Sellmeier equation for a given material, or in other words, the complete dispersion curve. The Sellmeier equation is expressed as

\[
n^2(\lambda) = 1 + \frac{a\lambda^2}{(\lambda^2 - b)} + \frac{c\lambda^2}{(\lambda^2 - d)} + \frac{e\lambda^2}{(\lambda^2 - f)},
\]

where \( a, b, c, d, e, \) and \( f \) are coefficients to be adjusted.

To demonstrate the feasibility of the femtosecond Maker fringes technique we used a 0.6 mm thick fused silica and a 1.0 mm thick BK7 samples. Both samples present well-characterized indices of refraction dispersion, being ideal to demonstrate the possibility of obtaining Sellmeier’s coefficients with our measurements. In addition, fused silica presents a well-known nonlinear coefficient.

Figure 1 shows experimental and theoretical results obtained for fused silica and BK7 samples pumped at 1200 nm (TH at \(~400\) nm). The magnitude of the BK7 signal leads to the conclusion that its nonlinear coefficient is twice that of fused silica, which is known to be \( \chi^{(3)} = 2.79 \times 10^{-14} \text{ esu} \). We also performed measurements at \( 1300, 1400, 1550, 1700, 1800, \) and \( 1900 \text{ nm} \). Owing to the large number of parameters to be adjusted, we decided to use a fitting routine based on an evolutionary genetic algorithm (GA) to find one set of Sellmeier’s coefficients for each pump wavelength used. Initially, a certain number of coefficients was randomly selected to give an initial population of refractive index. This population was tested, classified, reproduced, and mutated by the GA routine. The best solutions are more likely to be selected. Each set of Sellmeier’s coefficients gives one theoretical TH spectrogram. The test of fitness was made by comparing experimental and theoretical spectrograms by means of the minimum square root deviation. In order to obtain the best set of Sellmeier’s coefficients in a short period of time, we have to specify the order of magnitude of such constants and set some restriction on their ranges. As the starting condition,
we set the minimum and maximum values for the Sellmeier’s parameters in a wider range but around the expected values for the sample used. After several tries, these parameters start to converge and we narrow the search range to improve the convergence speed. With the purpose of achieving a more reliable solution, we average Sellmeier’s parameters obtained for different pump wavelengths. Such set of coefficient gives a dispersion curve that is valid in a broad range.

As seen in the TH spectrograms, the oscillatory pattern present at each wavelength contains information about the coherence length, which is related to the fundamental and the TH refractive indices difference. The curvature of a particular maximum (minimum) of a TH signal band contains information about the refractive index derivative. In other words, the TH Maker fringes process connects the values of the refractive index and its dispersion over two remote wavelengths in a wide wavelength band. Table I shows Sellmeier’s coefficients, obtained for both samples, that are very close to those found in the literature. The main factor that limits the accuracy of this technique is the GA used because it provides a good solution, nor necessarily the optimum solution. Other techniques devoted to measure the index of refraction are more accurate because they provide a direct measurement of this quantity.

In summary, we developed a powerful method based on the Maker fringes technique capable of providing reliable nonlinear coefficients and refractive index dispersion given by Sellmeier’s coefficients, using a broadband femtosecond laser pulse together with a spectrometer. We believe that this new single beam and simple technique can be a very powerful tool to characterize several new nonlinear samples.

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<table>
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<tr>
<th>TABLE I. Sellmeier’s coefficients obtained with the TH Maker fringes data and their tabulated values.</th>
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<td>Coeff.</td>
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8CVI Laser Optics and Coating Catalog, Corning Glass Catalog (CVI Laser Corporation, Albuquerque, New Mexico, 1999).