‘Viewing’ objects hidden in highly scattering media by cross-correlating the Fourier transform of the image with the incident field in a second-order non-linear crystal

Alessandra Andreoni a,b, Maria Bondani a,b,* , Marco A.C. Potenza a,b, Fulvia Villani c

a Dipartimento di Scienze Chimiche, Fisiche e Matematiche, Università’ dell’Insubria, Via Lucini 3, 22100 Como, Italy
b Istituto Nazionale di Fisica della Materia, I.N.F.M., Milano-Università’, 20133 Milan, Italy
c I.N.F.M., Napoli-Università’ Federico II, 80131 Naples, Italy

Received 19 August 1999; accepted 18 November 1999

Abstract

The detectability of the image of an object immersed in a scattering medium and transilluminated by a multi-mode laser pulse can be enhanced by cross-correlating the transmitted field with a fraction of the incident one in a β-barium borate crystal. We show that this effect arises from the interplay of the temporal coherence properties of the interacting fields, even in the presence of a modest coherence such as that of a Q-switched Nd:YAG laser pulse. © 2000 Published by Elsevier Science B.V. All rights reserved.

PACS: 42.65. – k; 42.65.Ky; 42.30. – d
Keywords: Frequency conversion; Optical cross-correlation; Optical imaging/mapping; Electromagnetic scattering by random media

1. Introduction

In this paper we show that the coherence of a three-wave interaction such as frequency up-conversion is useful to detect a weak coherent field superimposed to a more intense incoherent one at the same frequency, ω. This case occurs, for instance, when a laser pulse travels through a highly scattering medium: only a small fraction of the incident pulse, often called the ballistic pulse, keeps coherent with the incident one and emerges from the turbid medium after a time given by the ratio of optical path to group velocity in the medium, while most of the light is randomised in propagation direction and optical path length due to multiple scattering. As a result, at the exit of the medium, the largest fraction of the pulse energy in the forward direction is contained in a time-spread incoherent pulse that emerges delayed with respect to the ballistic pulse. Both delay and time-spread are greater the more numerous the average scattering events.

If an object immersed in such a medium is illuminated by a polarised incident pulse, the above effect...
may prevent the transilluminated object from being detected, because all image information is carried by the field components that are unaffected by scattering and constitute the ballistic signal. Different techniques have thus been proposed to enhance the detection of the ballistic signal by selecting it in time (time gating), in space Fourier components (spatial filtering), in polarisation or according to combinations of these features. The early implemented time-gating techniques, based on Kerr gating with picosecond resolution [1], were shown to achieve better performances if coupled to spatial filtering [2].

Time-gating on the femtosecond scale was made possible by the advent of ultrashort pulse Ti:sapphire lasers and by the use of cross-correlation techniques based on non-linear optical interactions, such as second harmonic (SH) generation [3], parametric amplification [4], coherent anti-Stokes Raman scattering [5], and coherent Raman amplification [6,7]. Recently, Fourier spatial filtering in tandem with polarisation selection was demonstrated to allow transillumination imaging through turbid media, as highly scattering as tissues, without need of time gating [8]. Here we demonstrate that SH cross-correlation by itself can select the image-bearing coherent field in critical transillumination experiments, independently of time resolution. This allows us to use nanosecond instead of femtosecond pulses to illuminate the object.

The idea underlying the present work stems from the results of experiments of time-delayed SH cross-correlation of the part of a Nd laser pulse emerging non-deviated from a 1-cm thick turbid medium, with collimated transmittance of values below $10^{-7}$, (pulse 1, wave-vector $k_1$), with a fraction of the incident pulse (pulse 2, wave-vector $k_2$) [9–11]. Pulses 1 and 2 were cross-correlated in a BaB$_2$O$_4$ crystal (BBO I) used in the same non-collinear type I phase-matching geometry as in the present experiments. We observed that, by measuring the frequency-doubled signal at different time-delays, we only obtained detectable signals at zero-delay. The cancellation of the cross-correlated signal at the delays at which the diffuse component is expected [12] was almost as efficient when 1.2 ps, at 1.055 μm, as when 19 ps pulses, at 1.064 μm, (see below) were used as the incident pulses. The tolerance in positioning the optical delay line at zero-delay was congruous with the incident-pulse duration in the two series of experiments. Here we show that SH cross-correlation produces an increase in the detectability of the coherent part of pulse 1 with respect to incoherent one, not only as an obvious result of the angular selection on the wave-vectors imposed by the phase-matching condition, but mainly because of the coherence of the non-linear interaction.

2. Experiments

The experiments presented here, aimed at demonstrating the assessment of Section 1, were all performed by using micelle suspensions of Intralipid (Pharmacia, Italy) at different concentrations in doubly-distilled water as the turbid media. The volume-to-volume percent concentration values quoted in this paper are absolute values, which include the 10% dilution factor of the commercial product [13].

The general layout of the set-up is shown in Fig. 1. In all experiments, the fundamental Nd laser pulses were 50% split into two pulses arriving coincident in time at the BBO I crystal (5 × 5 mm, 2 mm depth, cut angle: $\vartheta_{cut} = 40^\circ$). Different laser sources were used: a chirp-pulse amplified feedback-controlled mode-locked Nd:glass laser (mod. TWINKLE, Light Conversion, Vilnius, Lithuania) generating 1.2 ps pulses (19 cm$^{-1}$ bandwidth, time-bandwidth product twice the transform limit), a feedback-controlled mode-locked Nd:YAG laser (mod. EKSPLA PL2143A, EKSMA, Vilnius, Lithuania) generating 19 ps pulses, and a Q-switched Nd:YAG laser (mod. Quanta-Ray GCR-4, Spectra-Physics, Mountain View, CA) generating 18 ns pulses (< 1 cm$^{-1}$ bandwidth). The output beam of the last one, which was used without amplification, was spatially filtered to achieve a beam quality similar to that of the other sources. In all cases, a telescope was inserted before the beam splitter to reduce the beam diameter to about 0.9 mm full-width at half-maximum (FWHM).

The intensity values at the sample entrance were in the range 10–25 MW/cm$^2$ in all experiments. The samples were constituted by Intralipid suspensions of up 1.25% concentration contained in: (i) a 1 × 1 cm$^2$ quartz cuvette, with four polished windows, at whose centre a vertical, 0.78-mm diameter, stainless steel
needle was inserted; and (ii) a $1 \times 5 \text{ cm}^2$ quartz cuvette (optical path: 1 cm), with four polished windows, partially blanked by a stainless steel blade, with very sharp and straight edges, which was positioned inside the cell, parallel to the larger side window. With reference to Fig. 1, the angle between beam 1 and beam 2, the BBO I orientation in the plane of the figure, that is the tuning angle, as well as the sample-to-BBO I distance, were varied in the different experiments, as specified below. Prior to each experiment, once established the angle between beams 1 and 2, the sample cell was filled with water and the crystal tuning angle adjusted for maximum SH cross-correlation. The focal plane of the far-field lens (nominal $f$: 15 cm, diameter: 2 cm) was made to coincide with the sensor plane of the CCD camera. This, which was alternatively used to detect the up-converted or the fundamental (beam 1) far-field intensity distribution, was a Pulnix (mod. PE2015, Pulnix Europe, Basingstoke, UK) and was connected to a Spiricon (mod. LBA100, Spiricon, Logan, UT) for image analysis. Suitable neutral-density filters with calibrated optical densities were inserted before the CCD camera in each measurement to ensure a linear response. When up-converted signals were to be measured, a band pass filter was added to cut the stray light at the fundamental wavelength.

We measured up-converted far-field diffraction patterns of the needle with beams 1 and 2 crossing at $13.7^\circ$ to each other inside the BBO I crystal (tuning angle: $26.3^\circ$). The sample-to-BBO I distance determined the angle $\psi$, around the forward direction, in which the transmitted pulse 1 was collected by the crystal ($\psi \approx 20.8$ mrad). When the far-field diffraction patterns at the fundamental frequency, $\omega$, were to be detected, care was taken to put the lens at the distance from the sample ensuring light collection within the same angle. By using all three lasers to transilluminate the object with equal incident intensity (25 MW/cm$^2$) and at zero-delay between pulses 1 and 2, we could record diffraction patterns of the needle both at $\omega$ and at $2\omega$ for Intralipid concentrations up to 0.67%. At higher concentration values, the pattern at $\omega$ became insensitive to the presence of the needle on the pathway of pulse 1. On the contrary, the up-converted pattern could be detected up to 1.25% Intralipid concentration with the 1.2 ps
Nd:glass laser at 20 MW/cm² (Fig. 2(a)) and up to 1% with the 18-ns Nd:YAG laser at the same intensity (Fig. 2(b)). As shown by the arrows in the figures, the spacing between the diffraction maxima in Fig. 2(a) is in good agreement with the value expected for the angular spacing of maxima beyond first order (0.68 mrad), whereas the spacing in Fig. 2(b) is very similar to that between the 0th order and the two first-order maxima (0.98 mrad, see Section 3). The fact that diffraction maxima at different orders are displayed in different measurements is due to the dimension of the illuminating beam, which is not much greater than that of the object, and to its radial profile. In fact, to avoid saturation of the detecting system by the central maximum, when necessary, the beam was deliberately not centred with the needle. The comparison of Fig. 2(a) with Fig. 2(b) shows that the huge difference in time duration of the illuminating pulses is rather irrelevant to the detectability of the frequency-doubled far-field diffraction pattern, above a given background of scattered light.

To investigate the reasons for this result, which is apparently in contrast with the recognised role of femtosecond time-gating in non-linear cross-correlation experiments similar to ours, we made the following experiments, in which we used the 1 × 5 cm² quartz cuvette partially blanked by a blade.

We sent the 18-ns Nd:YAG pulse through the cell, filled with 0.67% Intralipid, and recorded the far-field intensity distributions at ω and 2ω, i.e. for transmitted field (field 1 in Fig. 1) and SH cross-correlation to the reference field 2. The measurements were performed with beam 1, of 870 μm FWHM diameter, either passing through the turbid medium, 2.75 mm away from the edge of the blank, or striking the blank, at 4.52 mm distance from the edge (all distances are relative to the beam centre). At frequency ω, we obtained the maps plotted in Fig. 3(a), for the blanked beam, and in Fig. 3(b), when the beam was not stopped. Since the maps were recorded with equal incident intensity (11.5 MW/cm²) and grey filters with different densities were obviously used in front of the CCD camera for the two measurements, the intensity data reported in the figures were corrected for the attenuation of the filters before plotting. For short, we call ‘scattered’ light that mapped in Fig. 3(a) and ‘transmitted’ light that in Fig. 3(b). The former one is much more spread in angle, as compared with the ‘transmitted’ light, and its maximum intensity is smaller by a factor of 2.33 = 10y5. With an incident-pulse power equal to that used for these measurements, we detected the far-field distributions at 2ω. In these SH cross-correlation measurements, beams 1 and 2 crossed at an angle of 5.7° to each other inside the BBO I crystal (tuning angle: 23.4°), while the ψ angle of 20.8 mrad was preserved, as in the measurements of the needle diffraction patterns. When the beam was blanked (‘scattered’ light up-conversion) and no filters were used before the camera, no pixel out of the 120 × 120 digitised recorded even a unit signal (data not shown). The up-converted ‘trans-

![Fig. 2. Up-converted far-field diffraction pattern of a 0.78 mm diameter needle, immersed in: (a) 1.25% Intralipid and transilluminated by a 1.2 ps pulse; and (b) 1% Intralipid and transilluminated by a 18 ns pulse. Both patterns are averaged over 10 shots of 20 MW/cm² incident intensity and are mapped into 8 equally wide intensity zones. The arrows mark the distances between intensity maxima (expected values: 0.68 mrad in (a) and 0.98 mrad in (b), see Section 2).](image-url)
Fig. 3. Far-field intensity distribution, at \( \omega \), of a 870 \( \mu \)m diameter beam (18 ns pulse duration, 11.5 MW/cm\(^2\) incident intensity): (a) striking the immersed blank (see Section 2); or (b) passing through 1 cm of 0.67\% Intralipid suspension. The intensity levels, in number of 10 equally spaced, are expressed in the same arbitrary units.

Mitted’ light consisted of a very confined peak, easily detectable above a vanishing background, as shown in the 3-D plot in Fig. 4(a). This distribution, at \( 2\omega \), was normalised to the peak value for comparing its shape to that at \( \omega \), which is displayed as 3-D plot in Fig. 4(b) (same data as in Fig. 3(b)). A smaller angular spread is actually observable in Fig. 4(a) as compared to Fig. 4(b), namely, 0.35 mrad versus 0.48 mrad (FWHM values). Obviously, the non-linearity of the SH cross-correlation plays in favour of the narrowing of the detected ‘transmitted’ light, but we already demonstrated that, in the low-conversion regime in which we operated, this effect is minimal [10,11]: in fact, the far-field intensity patterns, both at \( \omega \) and \( 2\omega \), of the same beam transmitted through water were very similar to the up-converted pattern through the scatterer (see Fig. 4(a)). Thus the wings around the peak in Fig. 4(b), which are due to scattering, seem to be not converted to SH.

To strengthen this conclusion, we must demonstrate that the zero background in Fig. 4(a), which corresponds to an infinite contrast in the detection at \( 2\omega \) of ballistic versus scattered light, is not an experimental artefact. Actually, some experimental constraints must be taken into account: the power of the incident pulse and the sensitivity and resolution of the camera/digitiser system might be responsible for the lack of counts in the pixels around the peak. To achieve a quantitative evaluation of the real contrast in Fig. 4(a), it was necessary to devise an experimental situation in which also the up-converted scattered light became detectable by our system. We thus enlarged the collection angle, \( \psi \), to a value of \( \approx 80 \) mrad and repeated the far-field mea-

Fig. 4. Up-converted far-field intensity distribution in (a) and distribution at the fundamental frequency (same data as in Fig. 3(b)) in (b) of a 870 \( \mu \)m diameter beam (18 ns pulse duration, 11.5 MW/cm\(^2\) incident intensity) transmitted by 1 cm of 0.67\% Intralipid. The distributions normalised to the peak values. The \( x-y \) axes in (b) coincide with the horizontal and vertical directions in Fig. 3. The FWHM values are: (a) 0.35 mrad; and (b) 0.48 mrad.
measurements at $\omega$ and $2\omega$ for the ‘scattered’ light (blanked beam). We used the same laser (18 ns pulse duration, 870 $\mu$m beam diameter) and repositioned beam splitter and mirrors so as to keep the cross-correlation geometry (beams 1 and 2 crossing at 6.5°) similar to that used for the measurements in Figs. 3 and 4. The results of the far-field measurements at $\omega$ and $2\omega$, both obtained with 11.5 MW/cm² incident intensity, are displayed in Fig. 5, upon correcting the experimental intensity values for the attenuation of the filters before the camera. Note that the intensity scale in these 3-D plots is logarithmic. The up-converted far-field distribution is very flat and about 25 times lower than the corresponding intensity level recorded at $\omega$. Before drawing conclusions from the measured value of this ratio, we must take into account that the two measurements were carried out on an extended source by using different collection optics in the set-up when passing from IR to SH. Thus the results in Fig. 5 have to be corrected for the so-called ‘geometrical factor’, $G$ (in units of mm$^2$ × sterad). In our case, the ratio between the geometrical factors plays in favour of the IR-detected signal, being: $G_{\text{IR}}/G_{\text{SH}} = 3.4 \times 10^{-2}/2.8 \times 10^{-3} \approx 12$. Since the spectral sensitivity of the CCD sensor at $\omega$ is very similar to that at $2\omega$, we can safely assess that scattered light of a given energy produces a detected signal at $\omega$ twice as greater as the one detected upon SH cross-correlation. Even by scaling the 3-D plot at $\omega$ in Fig. 4(b) by this factor, the wings around the peak do not reach a level below the minimum one detectable by the CCD pixels: their disappearance in the far-field intensity map at $2\omega$ (see Fig. 4(a)) must then be explained on a different basis.

3. Discussion

All measurements reported here were performed to understand the experimental result (see Fig. 2(a) and Fig. 2(b)) that the SH cross-correlated diffraction pattern of an object can be detected even in the presence of an incoherent noise, caused by scattered light, prevailing over the image-bearing field to such an extent that the diffraction pattern at the wave-
length of the illuminating beam is not detectable. We could obtain this result by illuminating the object with a long-duration pulse, i.e., without using time-gating techniques.

We first show that the increase in detectability of the coherent part of the pulse emerging from the sample (pulse 1) with respect to the incoherent one is not ascribable to the space filtering, that is to the angular selection on the wave-vectors imposed by the phase-matching condition. A sketch of non-colinear phase-matching I geometry used throughout all measurements is shown in Fig. 6. With reference to the inset in the figure, we note that the angular acceptance, \( \phi = 20.8 \text{ mrad} \), see measurements on the needle in Fig. 2(a) and Fig. 2(b), covered about 7 diffraction lobes on either sides of \( k_{1,0} \). In fact, by calling \( \delta_{L,ext} \) the angular displacement of the \( L \)th diffraction maximum in air at 1.064 \( \mu \text{m} \), we calculate \( \delta_{L,ext} = 8.84 \text{ mrad} \) and \( \delta_{L,ext} = 10.21 \text{ mrad} \) for a needle-diameter of 0.78 mm. Field 1 contributions, ordinarily polarised and travelling along a set of \( k_{1,i} \) wave-vectors, are up-converted in the BBO I crystal by the coupling with the reference field of wave-vector \( k_2 \). Thus, the extraordinarily polarised field generated at \( 2\omega \) is characterised by a set of wave-vectors, \( k_{1,i} + k_2 \). If we suppose that the crystal is tuned for the phase-matched up-conversion of the 0th order, that is

\[
k_{SH,0} = k_{1,0} + k_2
\]

(1)

the wave-vectors, \( k_{1,i} + k_2 \), are symmetrically displaced by the angles \( \delta_i/2 \) relative to \( k_{SH,0} \) (see Fig. 6), being

\[
\delta_i = \alpha_0 - \theta_{cut} - \theta_0 - \sin \left( \frac{1}{n_F} \sin \left[ n_F \sin \left( \alpha_0 - \theta_{cut} - \theta_0 \right) \right] \right).
\]

(2)

In Eq. (2), in which \( n_F \) is the BBO ordinary refractive index at \( \omega \), the tuning angle and the half-angle between the fundamental beams 1 and 2 (\( \alpha_0 \) and \( \theta_0 \), respectively) are linked by the relation

\[
\theta_0 = a_0 \cos \left[ n_{SH}(\alpha_0)/n_F \right]
\]

(3)

in which \( n_{SH}(\alpha_0) \) is the refractive index experienced by the up-converted field of wave-vector \( k_{SH,0} \). The spacing angles between adjacent maxima, which are expected in the patterns at \( 2\omega \), can be calculated by using Eq. (2). They are displayed by arrows in Fig. 2(a) and Fig. 2(b).

![Fig. 6. Scheme of the SH cross-correlation interaction and (inset) wave-vectors of the field diffracted by the needle in air, before entering the BBO I crystal.](image-url)
In the phase-matching condition expressed by Eq. (1), the up-conversion of the maxima with \( l \neq 0 \) is increasingly phase-mismatched for increasing \( l \). It is interesting to evaluate the mismatch \( \Delta k_1 \cos(\delta_l/2) \) in the direction of \( k_{\text{SH},0} \), which turns out to be

\[
\Delta k_1 \cos \frac{\delta_l}{2} = \frac{2 \omega}{c} \left[ n_{\text{SH}} \left( \alpha_0 - \frac{\delta_l}{2} \right) \right. \\
- \left. n_x \cos \left( \theta_0 + \frac{\delta_l}{2} \right) \right] \cos \frac{\delta_l}{2}
\]  

(4)

and to compare it with the interaction bandwidth. Calculations valid for non-collinear frequency up-conversion with photon conversion efficiency low enough to not affect the intensity of field 2 led us to estimate that a mismatch of \( \approx 31 \text{ cm}^{-1} \) in the direction of \( k_{\text{SH},0} \) reduces the efficiency to zero [10,11]. Since Eq. (4) gives 29 cm\(^{-1} \) for \( l = \pm 2 \) and 18 cm\(^{-1} \) for \( l = \pm 1 \) in the geometry used to obtain the results in Fig. 2(a) and Fig. 2(b), a phase-matching bandwidth of \( \approx 31 \text{ cm}^{-1} \) agrees with the number of detected maxima shown in the two figures. 

The main result in these figures is that the detection of the needle diffraction pattern was possible upon SH cross-correlation even when the pattern was unrecognisable in the maps recorded at \( \omega \). In particular, when the 18-ns Nd:YAG laser was used, this happened at a collimated-beam transmittance through the Intralipid suspension below \( T = 1.24 \times 10^{-6} \) (Fig. 2(b)) [13].

To study the effects causing detriment to the detectability at \( \omega \) of the image-bearing coherent field we operated at lower Intralipid concentration \( (T = 1.15 \times 10^{-4}) \). When measured at \( \omega \), a collimated beam generates sizeable signals at all angles (see Fig. 3(a)) while the same beam avoiding the obstacle is detected in an angle covering 0.48 mrad (see Fig. 3(b) and Fig. 4(b)). The up-converted counterpart plot in Fig. 4(a) covers 0.35 mrad only and soon reaches ‘zero’ (see above).

As already observed, a reduction in the FWHM of the detected spot cannot be explained by simply invoking the non-linearity of the interaction (see above the comparison with the beams through water). Furthermore, such a reduction cannot depend on the angular selection: an interaction bandwidth of \( \approx 31 \text{ cm}^{-1} \) (see above) does not operate any effective spatial filtering since it allows to up-convert far-field patterns as spread in angle as that in Fig. 5. In fact, in phase matching, we have \( k_1 \cos \theta_0 \approx 48791 \text{ cm}^{-1} \) and, in order to achieve a mismatch of \( \pm (31/2) \text{ cm}^{-1} \), the \( k_1 \) wave-vector should deviate from \( \theta_0 \) by as much as \( \approx +6.2 \text{ mrad} \) and \( \approx -7.1 \text{ mrad} \) (on opposite sides).

Before explaining our experimental results, we first note that, in general, we made a cross-correlation between the reference field 2 and a field 1 containing both ballistic and scattered contributions. Now we will express the frequency up-converted field as obtained in exact phase matching by using our Q-switched Nd:YAG laser and demonstrate that the coherence of the non-linear interaction, in tandem with the multi-mode structure of the laser line, suffices to explain our experimental results.

At the BBO I entrance, we consider a complex field 1 with wave-vector \( k_1 \) and amplitude

\[
A_1(\omega) = a_1(\omega) + j \sqrt{N} \langle a_{1,\phi}(\omega) \rangle e^{-j\phi(\omega)}
\]  

(5)

in which the ballistic contribution \( a_1(\omega) \) is linked in phase with the complex field 2 amplitude, \( A_2(\omega) = a_2(\omega) \), and \( \Phi(\omega) \) denotes the phase of the overall scattered field 1, as given by the random-walk statistics, being \( N \) the number of speckles and \( \langle a_{1,\phi}(\omega) \rangle \) the amplitude scattered by each speckle [14]. The complex amplitude of the up-converted field at the exit of the crystal of depth \( L \) can be written as

\[
A_3(2\omega) \equiv -j \left[ a_1(\omega) \\
+ j \sqrt{N} \langle a_{1,\phi}(\omega) \rangle e^{-j\phi(\omega)} a_2(\omega) \right] gL
\]  

(6)

where \( g \) is the non-linear coupling coefficient [11].

According to Eq. (5), in the far-field intensity map of field 1, the unscattered contribution will be detected with a contrast to the noise due to multiple scattering given by \( |a_1(\omega)|^2/|\langle a_{1,\phi}(\omega) \rangle|^2 \). If the scattering medium contains an opaque object, its far-field diffraction pattern (Fourier transform of the image) will be detected at \( \omega \) with such a signal-to-noise ratio, \( S/N \). Moreover, if the light shed onto the sample comes from a single-mode laser, Eq. (6) says that the same \( S/N \) ratio will affect the SH cross-correlated intensity pattern.

In the case of a multi-mode laser, oscillating on \( M + 1 \) longitudinal modes, the frequency \( \omega \) on the
right-hand side of Eq. (6) represents any of the frequencies \( \omega_0 + i \Delta \omega, \omega_0 \) being the central frequency, \( \Delta \omega \) the laser-mode spacing and \( i \) the mode index \( (i = -M/2, \ldots, +M/2) \). For a non-linear interaction bandwidth much broader than the laser linewidth, as in our case, the up-converted field at any frequency, for instance at \( 2\omega_0 \), will also originate from \( M/2 \) interactions of \( A_1(\omega_0 + i \Delta \omega) \) with \( A_2(\omega_0 - i \Delta \omega) \) and from \( M/2 \) interactions of \( A_1(\omega_0 - i \Delta \omega) \) with \( A_2(\omega_0 + i \Delta \omega) \). To calculate the overall \( A_3(2\omega_0) \), it is convenient to express the contributions \( A_3(2\omega_0) \) of each couple of interactions involving fundamental fields at frequency oppositely shifted with respect to \( \omega_0 \). In obvious analogy with Eq. (5), we can write

\[
A_3(2\omega_0) = \sum_{j=1}^{M/2} \sum_{\Delta \omega} \left[ 2 a_1(\omega_0) a_2(\omega_0) e^{-j(\varphi_j + \varphi_{-j})} + \sqrt{2N} \langle a_{1,\omega}(\omega_0) \rangle \langle a_{2,\omega}(\omega_0) \rangle e^{-j\Phi} \right] gL.
\]

The term originating from the ballistic contribution,

\[
a_{3,bal}(2\omega_0) = 2 a_1(\omega_0) a_2(\omega_0) e^{-j(\varphi_j + \varphi_{-j})}
\]

and the one originating from scattering

\[
a_{3,scat}(2\omega_0) = \sqrt{2N} \langle a_{1,\omega}(\omega_0) \rangle \langle a_{2,\omega}(\omega_0) \rangle e^{-j\Phi},
\]

are deeply different in that the former one is the coherent sum of two fields, whereas the latter is the sum of \( 2N \) randomly dephased contributions. Note that \( \Phi \) is a random phase. Even when the sum over \( i \) is performed to obtain \( A_3(2\omega_0) \), these two terms behave very differently. The \( M/2 \) terms represented by Eq. (10) give an incoherent sum of \( (2N \times M/2) \) fields, that is:

\[
a_{3,scat}(2\omega_0) = \sqrt{NM} \langle a_{1,\omega}(\omega_0) \rangle a_{2}(\omega_0) e^{-j\Phi}.
\]

The sum of the \( M/2 \) terms represented by Eq. (9) depends on the degree of coherence among the laser modes. It can be demonstrated that, if \( m \) modes out of the \( M+1 \) laser modes are coherent, the overall \( a_{3,bal}(2\omega_0) \) is given by

\[
a_{3,bal}(2\omega_0) = 2 a_1(\omega_0) a_2(\omega_0) \left( \frac{M}{2} \right)^2 \left[ 1 - \left( \frac{m^2}{(M+1)^2} \right) \right] e^{-j\Phi}
\]

The factor \( m^2/(M+1)^2 \) appearing in Eq. (9) represents the probability that \( \varphi_j = \varphi_{-j} = \varphi \) for the \( i \)th couple. Thus \( (M/2)[m^2/(M+1)]^2 \) is the number of coherent couples whereas \( (M/2)[1-m^2/(M+1)^2] \) is the number of couples that sum up incoherently. In conclusion the overall field at \( 2\omega_0 \) is

\[
a_{3}(2\omega_0) = \sum_{j=1}^{M/2} \sum_{\Delta \omega} \left[ 2 a_1(\omega_0) a_2(\omega_0) e^{-j(\varphi_j + \varphi_{-j})} + \sqrt{2N} \langle a_{1,\omega}(\omega_0) \rangle \langle a_{2,\omega}(\omega_0) \rangle e^{-j\Phi} \right] gL.
\]

By comparing the up-converted scattered field term in Eq. (11) with that in Eq. (6) we note that the multi-mode structure of the laser brings about an increase by a factor of \( M \) in the corresponding detected intensity. The result in Fig. 5 provides evidence of this effect. We found that a given scattered infrared intensity produced a SH intensity smaller by only a factor of \( \approx 2 \). According to Eqs. (5) and (11) we can then write

\[
\frac{I_{scat}(2\omega_0)}{I_{scat}(\omega_0)} = \frac{2 \hbar \omega_0 | \sqrt{N} \langle a_{1,\omega}(\omega_0) \rangle \langle a_{2,\omega}(\omega_0) \rangle gL |^2}{\hbar \omega_0 | \sqrt{N} \langle a_{1,\omega}(\omega_0) \rangle |^2} \approx \frac{1}{2}
\]
from which we obtain: $4M|a_e(\omega_2)gL|^2 \approx 1$. Since $|a_e(\omega_2)gL|^2$ is the photon conversion efficiency, which is below 1% in our case, this result is congruous with the number of modes oscillating in our laser ($M \approx 80$).

Since the up-converted ballistic field in Eq. (11) contains two contributions with different phases, one of which is the random phase $F$, the calculation of the corresponding intensity is more difficult. However, if the up-converted far-field intensity map is averaged over a sufficiently great number of laser shots, the S/N ratio becomes

$$
\frac{I_{BAL}(2\omega_0)}{I_{SCAT}(2\omega_0)} = \frac{4\left[\frac{M}{2} \frac{m^2}{(M+1)^2}\right]^2 + \frac{M}{2} \left[1 - \frac{m^2}{(M+1)^2}\right]}{M} 
$$

$$
\times \frac{|a_e(\omega_0)|^2}{N|\langle a_{1,\delta}(\omega_0)\rangle|^2}. \quad (13)
$$

Since $I_{BAL}(\omega_0)/I_{SCAT}(\omega_0) = |a_e(\omega_0)|^2/(N|\langle a_{1,\delta}(\omega_0)\rangle|^2)$, Eq. (13) can be rewritten as

$$
\frac{I_{BAL}(2\omega_0)}{I_{SCAT}(2\omega_0)} = F(m,M) \times \frac{I_{BAL}(\omega_0)}{I_{SCAT}(\omega_0)} \quad (13a)
$$

in which the factor

$$
F(m,M) = \frac{4\left[\frac{M}{2} \frac{m^2}{(M+1)^2}\right]^2 + \frac{M}{2} \left[1 - \frac{m^2}{(M+1)^2}\right]}{M} 
$$

represents the increase in S/N ratio on passing from fundamental to SH. In Fig. 7 we plot $F(m,M)$ as a function of $m$ for $M = 80$; the corresponding curve grows slowly from the lowest value, $F(m = 0, M) = 2$, to the highest one achievable by a fully coherent source, that is $F(m = M = 1, M) = M$. The minimum increase by the factor of 2 in the S/N ratio shows that, even with an incoherent source, it is advantageous to frequency up-convert the intensity pattern. It is likely that we operated in a regime of low coherence with our Nd:YAG pulse. Nevertheless an $F$ value of about 2 was sufficient to bring the needle diffraction pattern to a detectable level at $2\omega$ as shown in Fig. 2(b). This result was obtained at 1% Intralipid concentration, that is in a condition in which the ratio $I_{BAL}(\omega_0)/I_{SCAT}(\omega_0)$ corresponded to a transmittance $T = 1.24 \times 10^{-6}$. When we used the Nd:glass laser, endowed with broader bandwidth and higher coherence, as shown in Fig. 2(a), we could detect the needle immersed in a 1.25% Intralipid suspension, which corresponds to a ratio $I_{BAL}(\omega_0)/I_{SCAT}(\omega_0)$ about 30 times lower. According to Eq. (13a), this means that a $F(m,M)$ value of at least 60 could be achieved: for $M$ in the range 800–1000, this is obtained for $m \approx 500$. This agrees with the fact that the time-bandwidth product for our Nd:glass laser pulses is twice the transform-limit value.

4. Conclusions

We showed that SH cross-correlation, between the incident field and the forward-scattered field is a means to detect the low-intensity residual coherent field immersed in the scattered incoherent one, even when the latter is more intense. The cross-correlation selectivity for the coherent field is a result of the coherence of the non-linear interaction enhanced by the use of broadband laser pulses with high coher-
ence among the modes. It would be challenging to apply our technique by using an ultra-broadband mode-locked laser and check the limit in the increase in S/N ratio that can be achieved on passing from the images detected at the fundamental frequency to those obtained upon SH cross-correlation.

Acknowledgements

This work was supported in part by the Italian National Research Council (C.N.R.) through grant ‘Short-term mobility 1999’, and contracts 96.00264.CT02 and 97.00070.CT02. M.A.C.P. acknowledges E.N.E.A. (Ente per le Nuove tecnologie, l’Energia e l’Ambiente) for his PhD studentship. The authors are deeply indebted to F. Ferri (University of Insubria) for the useful comments and suggestions.

References