Universal wave phenomena in multiple scattering media

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Measuring intensity correlations using diffuse second-order interference

Methods based on nonlinear optical processes have shown to be very sensitive indicators of the microscopic structure of materials, specially at the interfaces [114]. Despite the challenges of detecting low signals, these nonlinear measurements provide valuable information about about interfaces or molecular structures that are completely inaccessible with linear microscopy [50]. The applications of measuring higher harmonics are not limited to optics and can for example be influential in studying the interior of earth by using seismic waves. Second harmonic ultrasound measurements have also been used to get a higher contrast in blood perfusion measurements inside human body.

In section 2.3, we derived the relation between the efficiency of a nonlinear conversion process with the intensity-intensity correlations inside random media. This proportionality can be potentially used to extract new information about the internal structure of opaque medium. Furthermore, some important phenomena are specific to intensity measurements and cannot be detected in amplitude-level measurements. Anderson localization is one example. The multifractal behavior of eigenfunctions that was introduced and discussed in the past three chapters can only be characterized based on the intensity distribution and its correlations [32]. The relation between fluctuation of second harmonic efficiency and the mode structure in a disordered system have also been theoretically investigated for planar composites [122].

From the theoretical results of chapter 2, we saw that the intensity-intensity correlations add an additional contribution to the total second-harmonic generation. Determination of this surplus will require an absolute measurements, which is often a challenge in realistic situations due to the unknown instrumental response functions. In this respect, a relative measurement is much easier to perform. In this chapter, we introduce such a relative measurement, and present experimental results on turbid nonlinear samples, like porous gallium phosphide and laser-crystal powder. Our results show clearly that intensity correlations of type $C_0$ are non-universal and their dependence on the microscopic structure is much more pronounced that typical mesoscopic parameters like scattering mean free path. These experiments also show that a scalar wave treatment is not suitable for describing
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the nonlinear efficiency and underscores the prominent role of light polarization and the tensorial form of the nonlinear susceptibility.

7.1 Two classes of nonlinear random media

Before describing our experimental method, we briefly review the two common types of samples that are relevant for nonlinear random media. In the first category, scattering is introduced in a slab of nonlinear dielectric by a small density of point-like scatterers. This case is theoretically analyzed by Kravtsov et al. [73]. The density of the intrusions is considered to be so small that the nonlinear susceptibility is not affected, thus the microscopic nonlinear coefficient is supposed to be uniform across the sample. The background medium can however be optically dispersive, \( \Delta n \equiv n_{2\omega} - n_{1\omega} \neq 0 \). This is often the case in nonlinear crystal. It is also the main limitation factor behind the extraction of the converted light due to the presence of phase mismatch and destructive interference between fundamental and second-harmonic light. The conventional way of overcoming this issue is to use birefringence crystals, but that rules out the use of several materials with high nonlinear coefficients. That is why, introducing small scatterers may enhance the second harmonic yield because it prevents the waves from destructive interference.

By applying the diffusion approximation to the nonlinear Maxwell equations, Kravtsov et al. derived the second-harmonic yield in a semi-infinite medium. Their results show that when \( \Delta n \leq 0 \), the bulk contribution to the diffusely-reflected second-harmonic light \( R_{2\omega} \) is independent of the scattering parameter \( k_{1\omega} \ell_{1\omega} \). When \( \Delta n \geq 0 \) then \( R_{2\omega} \) depends on \( k_{1\omega} \ell_{1\omega} \) like

\[
R_{2\omega} \propto \cot^{-1}[Ak_{1\omega} \ell_{1\omega}],
\]

where \( A \) is a number that is dependent on the optical dispersion of the medium and the ratio between mean free paths at second-harmonic and fundamental frequencies. For this results to hold, the scattering mean free path must be much larger than the wavelength of light.

In the second case, a suspension of small spherical particles have been considered by Makeev and Skipetrov [83]. In their model, the conversion centers are the same as the scatterers and the background medium is linear. In their derivation the second-harmonic yield per scatterers shows no explicit dependence on the linear scattering properties of the suspension, i.e. there is no mesoscopic effect in the lowest order of density.

Unlike the first system, nonlinear conversion in the second model is local the second-harmonic intensity generated at each point is proportional to the square of the local intensity. This model holds all the assumptions we took when deriving the relation between conversion efficiency and mesoscopic \( C_0 \)-correlations in chapter 2. The additional term on the order of \( (k_{1\omega} \ell_{1\omega})^{-1} \) is half the product of \( C_0 \) and the zeroth-order term:

\[
R_{2\omega}^{(1)} = \frac{1}{2} C_0 R_{2\omega}^{(0)}. \tag{7.2}
\]

Based on this result, one should be able to measure the \( C_0 \)-correlations by looking at the change in second-harmonic yield as a function of scattering strength. In practice, performing such an absolute measurement with a precision that is high enough for extracting the \( C_0 \) contribution is experimentally challenging. Furthermore, the nonlinear susceptibility of the colloidal particles may be influenced by changing their density or adding extra scatterers.
We have found a better solution and that is to use a two-beam setup. This experiment is introduced in the following section.

A second solution is to focus on a single nonlinear conversion center embedded inside the turbid medium and look at the fluctuations of the conversion efficiency when this scatterer probes different realization of disorder. All the following arguments holds also for incoherent two-photon processes such as two-photon fluorescence. One advantage of using fluorescent sources is that the life-time is also a probe of local density of states (LDOS) [20] and one can experimentally look into the relation between LDOS-fluctuations and mesoscopic intensity correlations.

7.2 Two-beam experiment

In this section, we introduce the setup for measuring intensity-intensity correlations inside random media by using nonlinear combination processes. In this scheme, two weakly focused Guassian beams with the same frequency are incident on the same spot of a multiple-scattering slab. The second-harmonic light is generated inside the sample. A fraction of the scattered second-harmonic intensity is measured in reflection or transmission. Ito and Tomita have used a similar scheme to measure the spatial and temporal extent of the speckle inside multiple scattering medium [65]. They have compared their measurement results and found a very good agreement with a theoretical model based on the scalar wave approximation. In this section, we first show why using a scalar wave approximation is fundamentally wrong for such an experiment. In the following section, we show how the specific choice of the analyzed material could have influences their observation and brought the experimental results close to the prediction of a scalar treatment.

7.2.1 Underlying theory

Following the formalism introduced in section 2.3, we first look at the the scalar model. The amplitude of the wave-functions corresponding to each of the impinging beams are denoted by complex variables $\psi_{1\omega,a}(x)$ and $\psi_{1\omega,b}(x)$ at position $x$ inside the sample. For simplicity, we omit the position dependence in the following short calculation, noting again that the medium is statistically homogeneous, and averaging over position is the same as averaging over realizations of disorder.

Following Eq. (2.43), for the second-harmonic polarization density generated at a ny position inside the sample we have

$$p_{2\omega,a+b} = \tilde{\psi}_{1\omega,a} + \psi_{1\omega,b})^2.$$(7.3)

with the subscript $a+b$ denoting the total yield in presence of both impinging beams. For the moment, we have assumed that the nonlinear susceptibility tensor can be replaced by a scalar. The second-harmonic source intensity at each point reads

$$J_{2\omega,a+b} \equiv \bar{p}_{2\omega,a+b} p_{2\omega,a+b}$$

$$= |\tilde{\psi}_{1\omega,a} + \psi_{1\omega,b})^2(\psi_{1\omega,a} + \psi_{1\omega,b})|^2$$

$$= |\tilde{\psi}_{1\omega,a} + \psi_{1\omega,b})|^2[I_{1\omega,a} + I_{1\omega,b} + 4I_{1\omega,a}I_{1\omega,b}$$

$$+ (I_{1\omega,a} + I_{1\omega,b})(\psi_{1\omega,a}\psi_{1\omega,b} + \psi_{1\omega,b}\psi_{1\omega,a}) + \psi_{1\omega,a}^2 \psi_{1\omega,b}^2 + \psi_{1\omega,b}^2 \psi_{1\omega,a}^2].$$ (7.4)

Note that the source intensity is not yet averaged over realizations of disorder or position. The terms in the last line of equation (7.4) do not survive the ensemble averaging (or
integrating over an area much larger than the speckle size), except for the trivial case that the impinging beams are exactly overlapping. Using the definition of mesoscopic correlation functions $C^{(1)}$ and $C_0$, we can write down, up to the first order in inverse scattering strength, the following identities:

$$\langle I^2_{\omega,a} \rangle = (2 + C_0) \langle I_{\omega,a} \rangle^2,$$

$$\langle I^2_{\omega,b} \rangle = (2 + C_0) \langle I_{\omega,b} \rangle^2,$$

$$\langle I_{\omega,a} I_{\omega,b} \rangle = (1 + C^{(1)}_{ab} + C_0) \langle I_{\omega,a} \rangle \langle I_{\omega,b} \rangle. \quad (7.5)$$

We have used the facts that $C^{(1)}_{aa} \equiv 1$ for all $a$ and $C_0$ has an infinite range. In the experiment, the second-harmonic signals $V_a$ and $V_b$ are measured when beam $b$ and $a$ are blocked respectively. These signals (voltages) are directly proportional to the second-harmonic source intensity if we consider the second-harmonic light is not depleted. This is a valid assumption since the medium is not absorbing and the second-harmonic intensity is much smaller than the fundamental intensity. Since the propagation of second-harmonic light from the point of creation to the detector is completely linear, the proportionality factor is independent of the impinging beam and is solely determined by the instrument response function. The collective signal $V_{a+b}$ is measured when both beams are incident on the sample and illuminating the same volume in space. Considering the ensemble average of Eq. (7.4) and using identities (7.5) we get

$$V_{a+b} = V_a + V_b + \frac{4(1 + C^{(1)}_{ab} + C_0)}{2 + C_0} \sqrt{V_a V_b}. \quad (7.6)$$

From this experiment, $C_0$ can be measured when $C^{(1)}_{ab} \to 0$.

7.2.2 Beyond the scalar approximation

Light waves are not scalar. Yet, in most of the experiments regarding random media in the linear regime, the polarization is scrambled and most of the intensity measurements can be close to perfectly modeled by a scalar treatment. The situation is different for nonlinear random media, since the polarization shows up directly in the conversion process and hence manipulates the intensity of the higher harmonics. For the case of second-harmonic generation, the converted light is determined by the tensorial nonlinear susceptibility at each point:

$$\mathbf{p}_{2\omega} \cdot \mathbf{\hat{u}}_i = \sum_{j,k} \chi^{(2)}_{ijkl}(\mathbf{\Psi}_{1\omega} \cdot \mathbf{\hat{u}}_j)(\mathbf{\Psi}_{1\omega} \cdot \mathbf{\hat{u}}_k), \quad (7.7)$$

with $\mathbf{\hat{u}}_i$ the unit vector along each Cartesian coordinate. The susceptibility tensor depends both on the microscopic material properties and the geometry of the scatterer. Its determination, already a challenge for single crystals, becomes very complicated when the surface to volume ratio increases as in porous or powder structures. Therefore, one has always to resort to very simplified approximations. One such approximation is to consider a nonlinear point scatterer with isotropic internal polarizability.

Interestingly, even with such a simplification, the result of vectorial treatment for the two-beam experiment introduced above will be different from the scalar case. For a vectorial electric field, the electric field from each source should be added vectorially before taking its square value. This summation incorporates the angle between their polarizations $\theta$, which
7.3 Experimental settings

The outline of the experimental setup is schematically drawn in Fig. 7.1. Light from a 5-ns pulsed tunable Nd:YAG laser (Opolette, Opotek) is split in two paths and overlapped again by a second beam-splitter. One of the mirrors is put on a translation state. A lens after the second beam-splitter focuses light on the sample at its focal plane. With this configuration, the displacement of the mirror will result in changing the incident angle for one of the beams on the sample. The beam polarization in each path can be varied independent of the other one. The samples are few tens of microns thick and opaque, but

is well-defined at each point of space and moment of time, but varies rapidly both in time and space. Instead of Eq. (7.3), we will get

\[ p_{2\omega,a+b} = \tilde{I}(\psi_{1\omega,a}^2 + \psi_{1\omega,b}^2 + 2|\Psi_{1\omega,a}|^2\Psi_{1\omega,b}| \cos \theta). \]  (7.8)

For the second-harmonic source intensity, after the ensemble averaging and omitting the oscillatory terms, we have

\[ \langle J_{2\omega,a+b} \rangle = \langle I_{1\omega,a}^2 \rangle + \langle I_{1\omega,b}^2 \rangle + 4\langle I_{1\omega,a}I_{1\omega,b}\rangle\langle \cos^2 \theta \rangle. \]  (7.9)

The averaging for \( \langle \cos^2 \theta \rangle \) must be performed over the unit sphere with a proper weighting. Assuming a homogeneous distribution for \( \theta \), this average equals \( \frac{1}{3} \). For the more general case of an arbitrary nonlinear susceptibility tensor, this averaging must be weighted, incorporating the components of the tensor. Due to the quadratic averaging, the average can never be less than the value from the isotropic case (due to the Cauchy Schwarz inequality). In the most general case, we can summarize the results from the two-beam experiment on any sample with the following expression

\[ V_{a+b} = V_a + V_b + C_X \sqrt{V_aV_b}. \]  (7.10)

with \( C_X \) a non-universal and bounded quantity,

\[ \frac{2}{3} \leq C_X \leq 2, \]  (7.11)

which includes both the material structure and mesoscopic effect. Separation of these two effects in \( C_X \) is in general not possible except for the very simplified cases like isotropic point scatterers where we have considered in Eq. (7.4) or a highly uni-axial susceptibility tensor, which we shall discuss further in section 7.4.3. Note that for this result, only diffuse interference is considered. The interference of coherently propagating beams can enhance the signal by a factor 16, because all the terms in Eq. (7.4) will then survive averaging. For the isotropic model in the diffuse regime the results is

\[ C_X = \frac{4(1 + C_{ab}^{(1)} + C_0)}{3(2 + C_0)}. \]  (7.12)

Given a sample which follows this approximation, it should be possible to directly measure mesoscopic \( C_0 \)-correlations with our two-beam method. For other types of samples, this experiment can be a great tool for characterizing the internal structure of the sample by measuring the structure-dependent \( C_X \) quantity.
the substrate is transparent. The second-harmonic light generated inside the sample is detected in transmission. The transmitted light at the fundamental frequency is filtered by a cold-glass filter. The second-harmonic signal is collected by an objective and focused on a sensitive photo-diode. Saturation effects are avoided and the voltage of the diode is linearly proportional to the total second-harmonic intensity generated in the sample.

To extract the $C_X$ parameter introduced in the previous chapter, a series of measurements are performed for each configuration of direction and polarization of the two incident beams. This series includes varying the intensity of one of the beams and recording three values of $V_a$, $V_b$, and $V_{a+b}$ for each intensity, after correcting for the dark signal. The $C_X$ parameter is equal to the slope of the line fitted to the measurement points $(V_{a+b} - V_a - V_b)$ plotted versus $\sqrt{V_a V_b}$.

Various samples have been investigated with this setup. Most of the results presented in this chapter are measured on porous GaP samples with different scattering strengths. The samples are fabricated for this experiment using the method of photo-electrochemical etching [102]. The thickness and scattering strength of all the samples are characterized by using SEM images and coherent backscattering measurements. A summary of the samples characterization is presented in table 7.1.

### 7.4 Results and discussion

The first control experiment was to check the polarization dependence that was discussed in this chapter. In this experiment, the $C_X$ parameter is measured as a function of the angle between the two incident beams for parallel and crossed polarizations. The results are shown in Fig. 7.2. For the crossed polarization, $C^{(1)}$ is zero for any angle and thus the $C_X$ is independent of the angle between the two beams. If $C_0$ could have been extracted from $C_X$, this observation would have been the strongest evidence ever seen for the infinite range of $C_0$-correlations. For the parallel polarization, at large angles, the effect of $C^{(1)}$ vanishes again and the enhancement falls on the value observed for the cross-polarization case. At small angles however, the enhancement due to $C^{(1)}$ can clearly be seen as an increase in the enhancement. At even smaller angles, close to zero, the interference of coherent beams
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<th>$\kappa \ell (1200)$</th>
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Table 7.1: Summary of specifications for the GaP samples that are analyzed by the two-beam method. The thickness of etched porous-GaP layer is derived from scanning electron micrographs. The mean free paths are estimated from total transmission measurements. The resulted values are in agreement with measurements using white-light EBS setup [99] within fifty percent accuracy.

wins over the diffuse interference effects and a rapidly fluctuating interference pattern is observed. This interference pattern is not fully resolved due to its sensitivity to the incident beam jitters, but in theory the observed enhancement (or suppression) should be fluctuating between -2 and 14, when the angle is approaching zero.

In the second experiment, the polarization of collinear incident beams are set to crossed and $C_X$ is measured on a single GaP sample as a function of wavelength. The results are presented in Fig. 7.3. Although, it is not easy to model this result, this observation clearly shows the non-universal nature of $C_X$. Performing the same experiment on the laser powder (LiNbO$_3$) results in even a higher value of $C_X$, even though this sample has a longer scattering mean free path than porous GaP. Once again this observation is a sign of material dependence of $C_X$. In section 7.4.3, we will discuss our understanding of the underlying physics that explains the difference between the GaP and LiNbO$_3$ that can partly explain the measured enhancements.

### 7.4.1 Dependence on scattering strength

If the tensorial form of nonlinear susceptibility was spectrally uniform, the results shown in Fig. 7.3 could have been used to extract the dependence of $C_X$ on scattering strength, since we know that the mean free path of GaP is highly dispersive due to its porous structure. But the same geometrical effects of the pore shapes that are responsible for the dispersive mean free path can also affect the effective nonlinear susceptibility. To better understand this dependence, we have performed the diffuse nonlinear interference measurement on several GaP samples with various scattering strength (see table 7.1) at the same wavelength. The results are presented in Fig. 7.4. Although the resulted points are largely dispersed in the graph, the overall trend shows reduction of $C_X$ for smaller scattering strength. We should
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Figure 7.2: Dependence of the two-beam nonlinear enhancement signal on the angle between the incident beams for parallel (circles) and crossed (squares) polarizations for a typical sample. The large fluctuations in the for PP case are due to direct interference between the incident beams. In principle, the signal should oscillate and can be maximum 14 and minimum -2. These fringes are not completely resolved in this measurements due to the focusing of the incident beams. The flat signal for CP case indicated to the infinite range of $C_X$. The enhancement is normalized to the signal of a single beam and the angle is measured in degrees.

note that if mesoscopic $C_0$-correlations were to be observable in these measurements, their contribution should have increased at smaller values of $k\ell$. Therefore, we conclude that the measured dependence of $C_X$ on scattering strength is dominated by the structural dependence of the susceptibility tensor on the variations of pore geometry and cannot be exclusively representing a mesoscopic effect.

7.4.2 $C_X$ measurement in a single shot

The $C_X$ parameter is directly connected to the internal structure of nonlinear scattering centers. Meanwhile, due to its determination in a relative measurement and natural sampling of many positions and directions in the turbid sample, its determination can act a robust indicator for the changes in the microscopic composition of the scatterers. This property can be used to monitor the microscopic crystalline structure of for examples drugs, which have a vital role in the effectiveness of a medical treatment. Therefore, it may be favorable to introduce a method that measures $C_X$ in a single shot, instead of the three successive measurements that were introduced above. This single shot technique can also be used to monitor evolving samples like suspensions. Due to the short duration of the incident pulses, the two-beam experiment can function as a stroboscopic technique to monitor the dynamics of physical or chemical reactions in the suspension.

The main idea of this one-shot setup in schematically drawn in Fig. 7.5. In this setup, the incident beams are extended along one of the beam axes using cylindrical lenses. The elipticity axes of the two beams make an angle with each other such that the central part of the ellipses overlap while the ends do not. This way, the signal at the non-overlapping regions can be used as a reference for $V_a$ and $V_b$ (after spending adequate effort on profile
7.4. Results and discussion

Figure 7.3: Dependence of $C_X$ on wavelength for porous GaP sample S6 (squares) and LiNbO$_3$ powder (circles). The behavior is attributed to a mixture of both structural dependence of nonlinear susceptibility and multiple-scattering effects.

corrections), while the signal at the center is a measure for $V_{a+b}$. Figure 7.5 shows an schematic drawing of such a measurement and Fig. 7.6 shows one typical measurement aside with the relevant cross-cuts.

Comparing the results from this one-shot experiment with those from the Mach-Zender-type one shows a quantitative good agreement. When combined with more advanced microscopy techniques, this experiment can provide structural information that are not accessible in the conventional second-harmonic microscopy experiments. In some sense, the $C_X$ measurement is the nonlinear extension of interferometric phase-contrast imaging in linear microscopy.

7.4.3 Applicability of the scalar model

Finally, it is worth discussing the use of a scalar model for non-linear diffuse interference. In the introduction of this chapter, we mentioned that Ito and Tomita [65] have used a similar two-beam experiment in the past to measure the $C^{(1)}$ correlations inside a sample of laser powder and in fact used a scalar model to fit their data. Contrary to our statement that a scalar model is wrong for the interpretation of this experiment, a relatively good agreement has been found between their measurements and the scalar model that was used. The question arises: how is that possible?

We think the explanation for this apparent confusion is in the very choice of sample that was used in that article. They have used LiNbO$_3$, which has the following bulk nonlinear susceptibility tensor:

$$d^{LiNbO_3} = \begin{pmatrix} 0 & 0 & 0 & 0 & 14 & -7.4 \\ -7.4 & 7.4 & 0 & 14 & 0 & 0 \\ 14 & 14 & -98 & 0 & 0 & 0 \end{pmatrix}. \quad (7.13)$$

The element $d_{33} = 2\chi^{(1)}_{333} = -98$ is much larger than the other elements which means the second harmonic conversion efficiency is much higher when the polarization of the two fun-
damental photons are aligned with the 3rd lattice vector. This means that the crystal, effectively, acts as a local polarizer that filters those photons responsible for (efficiently) generating the second harmonic photon. This picture, rehabilitates the scalar model for describing their experimental data. We have also measured on the some LiNbO$_3$ powders. A comparison with porous GaP is made in Fig. 7.3. We have also found a higher enhancement than the prediction of an isotropic vector calculation, inline with the literature. The difference between our final result and two, the value that comes out the scalar model, may be related to the small size of powder grains or the emergence of the vectorial nature of conversion process due to the other non-zero elements of the nonlinear susceptibility tensor.

Having looked at actual material properties of LiNbO$_3$, it is worth discussing the results for GaP. For the bulk of this substance, the nonlinear susceptibility tensor has a completely different structure:

$$d^{\text{GaP}} = \begin{pmatrix} 0 & 0 & 0 & 16 & 0 & 0 \\ 0 & 0 & 0 & 0 & 16 & 0 \\ 0 & 0 & 0 & 0 & 0 & 16 \end{pmatrix}.$$ \hspace{1cm} (7.14)

In fact, the second harmonic conversion in bulk GaP has a rotationally symmetric nature. All the conversion processes can be described by combining two photons with perpendicular polarization into the the third second-harmonic photon with polarization again perpendicular to both fundamental photons. The cyclic permutation of polarization directions does not influence the efficiency; $\chi_{123} = \chi_{231} = \chi_{312}$. This property, makes GaP a very suitable candidate for using an isotropic vector model, which shows the minimum $C_X = \frac{2}{3}$.

Although, from our data, the GaP results are closer to the theoretical prediction from isotropic vector model than for other substances, it is short of quantitative agreement. This observation may have several reasons. The most probable explanation is the porosity of our samples, which introduces a large surface to bulk ratio and causes deviation from the bulk nonlinear susceptibility. If these structural factors could have been measure independently, one could extract multiple-scattering effects from these results. Therefore, other two-beam experiments on single nano-crystals may be needed to make a firm conclusion about this
7.5 Final remarks

In this chapter, we have presented a method based on second harmonic generation to study mesoscopic intensity correlations inside multiple scattering media. We have performed several measurements which are consistent with the theoretical prediction but stayed short of disentangling the mesoscopic part from the intrinsic material properties that influence the output of our method. However, these results are enough for proving the shortcomings of a scalar model. By carefully looking into the nonlinear susceptibility tensor, we have shown that the scalar model holds only for very specific cases.

The next steps for this research involve the investigation of colloidal suspensions, for which the scattering strength can be varied without changing the structure of individual scatterers. This experiment would be the only way in our opinion that can really split the mesoscopic effects from the structural ones. A helpful trick would be to use the sum-frequency generation of two input beams with different frequencies, instead of second harmonic generation. This way, the overlap signal can be spectrally split from the signal of individual beams and therfore perform the actual measurement “background free”. Using the two-photon fluorescence instead of the coherent second harmonic signal will also have the benefit of comparing the correlation measurements with the lifetime variations of the dye molecules and by that further exploring the relation between LDOS fluctuations and mesoscopic $C_0$-correlations.
Figure 7.6: Left: A typical “single-shot” measurement image from a 100-µm thick slab of porous GaP. Right: The horizontal cross-cut of the measured data with the total SHG signal shown with dashed gray line and SHG signal of each individual beam shown by light and dark gray dots. The black line shows the difference between the total signal and the sum of SHG signal from two beams when measured individually.