Two dimensional nonlinear frequency-mixing photo-acoustic imaging of a crack and observation of crack phantoms

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A two-dimensional imaging of a crack by nonlinear frequency-mixing photo-acoustic method is reported. The imaging contrast is due to nonlinear photo-thermo-acoustic processes taking place in case of simultaneous excitation by lasers of thermo-elastic and acoustic waves in the vicinity of the cracks. The images are obtained by scanning of two co-focused laser beams in region of crack location. The first cw laser beam, modulated in intensity at low frequency \( f_L \), generates a thermo-elastic wave, which is able to strongly periodically modulate the local crack rigidity up to complete closing/opening of the crack. The second cw laser beam, intensity modulated at much higher frequency \( f_H \), generates an acoustic wave incident on the breathing crack. The crack rigidity is also influenced by the stationary non-modulated inhomogeneous thermal stresses caused by the stationary heating of the sample by both lasers. The main contribution to imaging contrast comes from the strong dependence of the reflectivity of acoustic waves on the crack rigidity. The modulation of crack rigidity by thermo-elastic wave leads to the parametric modulation of the reflected acoustic waves and the generation in the spectrum of the acoustic field of the side lobes, which are separated from \( f_H \) by the integer number of \( f_L \). Scan images of a crack with an amplitude dynamics up to 40 dB and a spatial resolution better than 100 \( \mu m \), are obtained through the mapping of nonlinear side lobe amplitudes. The observed dependences of the images on the power of the lasers are discussed and the physical explanation of the appearance of crack phantom images at high level of optical excitation is proposed. For comparison and elucidation of spatial resolution issues related to this imaging method, the one-dimensional images, obtained with one laser beam focus position fixed and the second beam focus position scanning, are also presented and discussed.

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I. INTRODUCTION

The imaging contrast in the nonlinear frequency-mixing photo-acoustic (NFMPA) method is, in its fundamental nature, due to the nonlinear/parametric acoustic phenomena. Modification of the spectrum of the acoustic field is due to its reflection/transmission by the spatially localized inhomogeneity with time dependent elasticity. High sensitivity of not only the frequency-mixing (parametric modulation)¹–³ but also of several other nonlinear acoustic phenomena (such as harmonics generation,⁴–⁶ subharmonics generation,⁷–⁸ demodulation,⁹–¹⁰ self-modulation,¹¹¹² modulation transfer,¹¹¹² and acousto-elasticity¹³) to the presence of cracks or contacting interfaces in the material is well known. These different nonlinear acoustic phenomena are rather frequently applied to the detection of the cracks (see Ref. 3, and references therein). However, the reports on the imaging of the cracks or on the attempts of their imaging are much rarer.¹⁴–²⁶ We believe that this situation is, at least partially, caused by the fact that for the crack detection it is sufficient to detect a nonlinear signal just from one point of the crack with the highest nonlinearity, while for the imaging of the crack, it is necessary to initiate the nonlinear scattering phenomena along complete length of the crack, i.e., even in its parts with lowest nonlinearity. Obviously, this would, in general, require higher amplitudes of the acoustic fields for imaging than for the detection.

Another reason to have high amplitude acoustic fields for imaging of the crack is related to the opportunity to induce crack breathing, i.e., periodic complete opening/closing of the crack.⁶,²⁷–³⁰ This regime of crack motion in the external fields (sometimes called “tapping” when it starts from the initially opened state and “clapping” when its starts from the closed state) is characterized by non-analytic non-classic nonlinearity³ related to abrupt/discontinuous changes in crack rigidity, which is known to be much stronger than the classic analytic power-law-type nonlinearities.³ When the crack is tapping or clapping the nonlinear phenomena on the crack can be distinguished from the nonlinear phenomena in the intact surroundings not only because they are stronger but also because they result in different spectral patterns of the acoustic field. For example, in the case of the frequency-mixing processes, which are of particular interest for the research results presented below, the smooth classic nonlinearities are exciting the side lobes with amplitudes which are diminishing very fast with the increasing order of...
the side lobe because of the cascade character of their excitation. This makes the detection of the side lobes of order 3 and even 2 very problematic in most of the experiments. In contrast, non-analytic tapping/clapping nonlinearity excites, because of the non-cascade multi-frequency nature of the initiated nonlinear interactions, the side-lobes of comparable amplitudes just slowly decreasing with increasing order. It is also worth noting that in the latter case the dependence of the side-lobe amplitudes on their order can be non-monotonous and, depending on the excitation level, the lower order side-lobe can be stronger that the higher-order one or vice versa.

We are aware of several different methods which had been earlier used in order to achieve strongly nonlinear motion of the crack with purposes of its imaging. In the experiments, strongly nonlinear multi-frequency oscillations in the vicinity of the crack/defect had been excited by acting on the complete sample by high power piezoceramic stack oscillators or vibrators. The surface vibrations at harmonics of the excitation frequency, at its subharmonics and ultra-subharmonics, are strongly localized in the vicinity of the defects and can be imaged by laser scanning vibrometer. The localization of the nonlinearly generated frequency components of the acoustic field is controlled in non-one-dimensional geometry not by their acoustic wavelengths but rather by the spatial extension of their nonlinear sources through the effects of the acoustic field geometrical divergence and possible multiple scattering of the acoustic waves in the vicinity of the elastically inhomogeneous and/or interface corrugated defects. The ultimate spatial resolution of this imaging technique in the detection of surface vibrations is limited by the probe laser focusing. However, it should be taken into account that the observed width of the surface breaking crack, i.e., the detected width of the nonlinear vibrations localization, could be controlled not only by the surface width of the crack but also by its depth.

It is worth mentioning here that strongly nonlinear vibrations of the crack/defect caused by the action of powerful vibrators on the sample are accompanied by the preferential heating of the crack vicinity in comparison with the surroundings. The sources of the heating are related to friction between the crack faces and strong dissipation of the acoustic energy in elastically soft parts of the crack, such as crack tips and interface contacts, where the acoustic stresses can be strongly concentrated. The temperature distribution around the crack/defect can be imaged with infrared cameras. The localization of the temperature field in non-one-dimensional geometry is controlled not by the thermal wavelengths but rather by the spatial extension of the heating sources through the effects of the thermal field geometrical divergence. So, potentially, the spatial resolution of these techniques could be controlled by the spatial resolution of the fast infrared cameras. However, in case of rather homogeneous vibration-induced heating of the faces of the deep surface-breaking cracks the one-dimensional propagation of thermal waves normally to the crack faces could take place. In this case, the detected width of the crack image on the surface could be controlled by the thermal wave diffusion length during the time of temperature measurement, i.e., during the response of the infrared camera.

In the experiments, strongly nonlinear acoustic interactions with the crack had been achieved by focusing on the crack, through the use of the time reversal mirror, of the bi-harmonic acoustic field. In the experiments, the crack had been located by applying of a time reversal mirror to the frequency mixed components of the acoustic field. The oscillations of the samples surfaces in the vicinity of the crack at mixed frequencies had been imaged by optical vibrometer. The spatial resolution of these imaging techniques is controlled by the same physical principles as those discussed above in relation to Refs. and. Note, that in Ref. and also in Ref. the acoustic excitation had been insufficiently powerful to ensure the imaging of the complete crack/defect. Only some parts of the crack had produced sufficient nonlinear signal.

In the experimental situations, where the probe acoustic fields, generated by the in-contact actuators at the sample surface, are used to image the breathing or stationary cracks through the parametric modulation or harmonic generation processes, the spatial resolution is classically limited by the wavelength of the probe wave. However, in practice, the spatial resolution is much worse because the finite size of the actuators and the probe beam diffraction. In Ref. the acoustic wave for probing the breathing crack had been generated through the thermo-elastic effect via heating the surface of metallic sample by focused intensity-modulated laser beam. In this experimental configuration, the spatial resolution is controlled by laser focus, because of the three-dimensional (3D) divergence of the probe acoustic wave and does not depend either on the probe wavelength or on the dimensions of the actuators, which had been used to initiate the crack motion and the detection of the nonlinear frequency-mixed components of the acoustic field. However, the crack driving in-contact piezoelectric transducer was not powerful enough to initiate tapping/clapping of the crack and the contrast of the obtained images was rather low because the classic crack nonlinearity was not much stronger than the nonlinearity of the surrounding intact sample.

The first analyses of the possibility to induce strong breathing of the crack/delamination by laser-induced thermoelastic stresses have appeared in the literature in relation to nonlinear thermal wave detection/imaging of the crack. The nonlinear/parametric phenomena in thermal waves are expected because of the modulation of the crack thermal resistance by the crack breathing. These ideas were confirmed experimentally in Ref. For the nonlinear/parametric acoustic diagnosis of the crack, the excitation of the crack breathing by the laser-induced thermoelastic stresses was achieved for the first time in Refs. and with use of high power (~20 W) pulsed-periodic laser light. As mentioned earlier, the spatial resolution in these experiments is limited by the application for the generation and detection of the probe acoustic field of the in-contact transducers. Finally, both the initiation of the crack tapping/clapping by the laser-induced thermoelastic stresses and the photo-acoustic
generation of the probing acoustic field have been recently combined in the NFMPA technique.\textsuperscript{25,26,33} The ultimate spatial resolution of the NFMPA imaging in 3D geometry is only limited by the dimensions of the light absorption region and can be much higher than the one limited by the acoustic and thermal wavelengths. Through the experimental observation of the multiple high-order side lobes, it has been demonstrated\textsuperscript{25} that clapping/tapping of the crack can be initiated by the intensity-modulated cw laser radiation of the power (\textasciitilde 120 mW), which is importantly lower that in Refs. \textsuperscript{21} and \textsuperscript{22}. It has been also demonstrated that the technique can be supplemented by the optical probing of the nonlinear frequency-mixed oscillations, thus providing opportunity for the all-optical probing of the nonlinear acoustics of cracks.\textsuperscript{26} By scanning co-focused pump and probe laser beams across the crack, one-dimensional (“cross sectional”) nonlinear images of the crack have been obtained\textsuperscript{25} demonstrating the contrast \textasciitilde 20 times higher than in linear photoacoustic images.

Below we demonstrate two-dimensional (2D) images of the crack obtained with NFMPA technique at two different levels of laser excitation by co-focused pump and probe laser beams. The images obtained through the detection of the first, second, and the third side-lobes are compared. The issues of the spatial resolution and of the information about the crack, which could be extracted from the visual/qualitative inspection of the nonlinear images, are discussed. The explanation of the crack phantom images providing the split and shifted positions of the crack, which are observed under particular circumstances, is proposed. The origin of the crack phantom images is attributed to the non-monotonous dependence of side-lobes amplitudes on the excitation level in case of strong tapping/clapping type nonlinearity of the crack. The proposed physical interpretations of the 2D images are supported by the presentation of the supplementary 1D images obtained at multiple excitation levels with one laser beam, either pump or probe, centered on the crack and another beam scanning.

II. PRINCIPLE OF THE METHOD

The method presented here makes use of an all-optical excitation with two independent intensity-modulated laser beams at frequencies \( f_L \) and \( f_H \). The photo-generated thermo-elastic wave at frequency \( f_H \) is called the pump wave, and the acoustic wave at \( f_L \), generated by the second intensity-modulated laser beam, is referred as the probe wave. The mixed acoustic frequencies \( f_{H\pm n} = p f_H \pm n f_L \) (with \( p \) and \( n \) as integers) that are detected as the output are necessarily generated by nonlinear modulation processes in the sample and not by a cross-talk between the independent excitation lasers. This is an advantage in comparison to methods relying on harmonics excitation for instance. The signal detection is achieved with an accelerometer placed sufficiently far (several centimeters) from the generation point. It is observed that the nonlinear modulation process is much more efficient when the laser beams are focused on a crack (or in its vicinity) than in a region without crack.\textsuperscript{25,26} Consequently, the modulation side-lobes around the probe wave frequency have a large amplitude when the generation process takes place in a vicinity of a crack but are practically absent out of a crack. We take advantage of this observation to produce 2D images of cracks. In order to improve the efficiency of the nonlinear modulation process at a crack, the photo-generated thermo-elastic wave at low frequency \( f_H \) needs to have the largest possible stress amplitude.\textsuperscript{33} The lower \( f_H \) is, the larger extremal temperatures are achieved and the larger is the generated thermo-elastic stress amplitude.

Being applied to the crack, characterized by an initial width and rigidity, these stresses cause the motion of the crack faces, modification of the crack width, and the modulation of the crack rigidity, which depends on crack width, i.e., is nonlinear.\textsuperscript{35} Consequently, local breathing of the crack occurs during a pump wave period, corresponding to a strong modulation of the crack properties and, as consequence, to the modulation of the probe acoustic field reflected/transmitted by the crack. The frequency \( f_H \) of the probe acoustic wave is chosen coincident with one of the resonance frequencies of the sample to maximize the waves’ amplitudes detected by the in-contact receiver. The nonlinear photoacoustic images of cracks are obtained by monitoring, along a two-dimensional spatial scan over the sample surface of the coincidently focused pump and probe beams, the amplitudes of the different modulation side-lobes \( f_{H\pm n} \) around the probe wave frequency \( f_L \). Several complementary images of a single crack can thus be obtained.

III. DESCRIPTION OF THE SAMPLE AND THE SETUP

A. Sample

The studied sample is a 50 \( \times \) 25 \( \times \) 3 mm\(^3\) plate of light absorbing glass containing a single surface breaking crack (Fig. 1(a)). This crack has been artificially created with a thermal shock in a fast cooling process after the sample has been heated up locally with a flame. The crack has a length of several centimeters and crosses the plate thickness at most of the places. The distance between the crack faces \( h \) can depend on the position along the crack but is of the order of a few hundreds of nanometers.\textsuperscript{40} The crack faces are mainly oriented orthogonally to the plate largest faces but can have oblique orientation at some few locations. An optical image of the scanned area of 1.8 \( \times \) 8.6 mm\(^2\) containing the studied crack is shown in Fig. 2(a). This image of the crack is obtained with a microscope and a transmitted light. The crack has been initiated next to \( x = 1 \) mm, \( y = 0 \) mm. In the top left area, i.e., for \( x < 1 \) mm, \( y \geq 6.5 \) mm (see Fig. 2(a)), the crack appears wider but this is due to oblique in-depth orientation of its faces. Moreover, in this particular area, the crack is only buried and not present on the surface. The sample is attached to a two-dimensional motorized translation stage in order to scan it with the lasers and produce the 2D images presented below.

B. Experimental setup

The experimental set-up is presented schematically in Fig. 1(a). The excitation of the thermo-elastic pump wave is realized by a 532 nm wavelength 2 W power continuous
wave laser (Coherent, Inc., Verdi) intensity modulated at $f_L$. The 100% intensity modulation is achieved by an acousto-optic modulator (AA Opto-Electronics, Inc., Model MQ180) at frequency $f_L = 1$ Hz. The acoustic probe wave is generated by a 800 nm wavelength, 1 W power diode laser, 100% intensity modulated at frequency $f_H$. The frequency $f_H$ is chosen of 25 kHz, so that it coincides with a resonance frequency of the sample. This allows one to maximize the detected wave amplitude and to improve the signal to noise ratio. The resonance curve around $f_H$ shows only a few dB variation for frequencies within a 10 Hz range from $f_H$ (a resonance quality factor of 1000 at 25 kHz provides a $-3$ dB amplitude variation within a 25 Hz range).

Both pump and probe laser beams are focused on the same location on the sample surface. The spot diameters are estimated by measuring the 1/e decrease of the beam intensity with the knife-edge technique. The 1/e diameters are $\sim 100$ and $\sim 300$ μm for the pump and probe beam, respectively. The optical penetration length of these excitation beams, measured by an optical transmission experiment, is equal to $\sim 300$ μm.

The signal, detected by an accelerometer of $\sim 50$ kHz bandwidth, is sent to a spectrum analyzer. The accelerometer is placed far enough from the scanned region to avoid any frequency-mixing process at its contact with the sample because the accelerometer position cannot be reached by the thermoelastic wave. The mixed-frequencies could be also detected optically if necessary.

In the experimental results presented in the following, we particularly analyzed the nonlinear spectral components with the highest amplitudes, associated to $p = 1$ and $n \leq 3$ ($f_H \pm nf_L$). The pump laser modulation frequency and, consequently, the thermo-elastic wave frequency of

![FIG. 1. (a) Schematic representation of the experimental set-up. (b) Detected spectrum when the pump and probe beams are co-focused on intact sample surface few mm from the crack. (c) Detected spectrum when the pump and probe beams are focused on the crack.](image1)

![FIG. 2. (a) Picture of the scanned area with the crack. (b)–(h) Top: Schematic representation of the analyzed spectrum component. Middle: Two dimensional scans of the crack achieved by detection of different side-lobes at $f_H \pm nf_L$. From left to right, the first three nonlinear left side-lobes: $n = -3$ (b), $n = -2$ (c), $n = -1$ (d), the main peak, at $f_H$ (e), and the first three nonlinear right side-lobes for $n = 1, 2, 3$ (f)–(h). All scans are represented with the same amplitude scale. Amplitude difference between two isolines is of 2 dB for $n = 0$, 6 dB for $n = \pm 1$, and 5 dB for $n = \pm 2, \pm 3$. Bottom: Scan section along $x$ for $y = 3$ mm (dotted line in middle figures). All scans are represented with the same amplitude scale.](image2)
$f_L = 1$ Hz, has been found to be a good experimental compromise between the necessity to efficiently drive the crack breathing and the acquisition time.

IV. PRELIMINARY DESCRIPTION OF THE NFMPA IMAGES

Results of the crack imaging are presented in Fig. 2 in the form of 7 images. Each image corresponds to the amplitude mapping of a frequency component of the modulation spectrum: the first three left and right side-lobes ($f_{f1}, n = 1, 2, 3$) and the main peak ($f_{f2}$). The side-lobe associated to the image is schematically shown at the top of each image. For these images, the laser beam diameters are $d_L = 108$ and $d_H = 328$ μm, the powers are $P_L = 76.3$ and $P_H = 46.5$ mW, and the modulation frequencies are $f_L = 1$ Hz and $f_H = 25$ kHz, for the pump and probe beams, respectively. The steps in both $x$ and $y$ directions, i.e., the pixel size of the images is $100$ μm × $100$ μm, which is comparable with the pump beam diameter ($108$ μm). The two-dimensional scans are presented with isolines and the same color scale is used. One dimensional scans along $x$, for $y = 3$ mm, are also presented in Fig. 2 at the bottom of each figure confirming the theoretical symmetry between the left and right side lobes. When compared to the optical image of the crack provided in Fig. 2(a), all the nonlinear photo-acoustic images show some sensitivity to the crack presence.

It is insightful to estimate the magnitudes of elastic strains and displacements that could be produced by the pump and probe beams in our experiments. We evaluated, by combination of analytical and numerical approaches, the 3D temperature distribution caused by laser heating, neglecting the heat resistance of the crack for the estimates. The linear thermal expansion coefficient $\alpha$ of our sample is measured to be $\alpha \approx 5.5 \times 10^{-6}$ K$^{-1}$, and its thermal diffusivity $\chi$ is found to be $\chi \approx 5.5 \times 10^{-7}$ m$^2$.s$^{-1}$. Assuming the pump and probe beams are co-focused and centered on the crack, the maximum stationary heating provoked by these beams is calculated to be $T \approx 50$ K, corresponding to a strain of $\varepsilon = A T = 3 \times 10^{-4}$ and a stress of $\sigma = 6 \varepsilon = 12$ GPa, with $E$ as the sample Young's modulus ($E = 39$ GPa). The relative displacement of the crack faces caused by this constant heating can be estimated by integrating the surface strain along the radial coordinate, i.e., $\Delta h_{\text{cons}} = 2 \times \int_0^\infty T(r)dr = 370$ nm in our experimental conditions, where the factor 2 corresponds to the fact that both faces are moving. The characteristic diameter of the heated zone $D$ can be considered as the ratio of the displacement over the strain, $D = \Delta h_{\text{cons}}/\varepsilon = 1.3$ mm. Concerning the thermal oscillating part, we need to take into account the theoretical attenuation length $\ell_{\text{th}}(f)$ of thermal wave at frequency $f = 1$ Hz in glass at $1/e$ level: $\ell_{\text{th}}(f) = \sqrt{\alpha/|\pi f|} \approx 420$ μm. As $D > \ell_{\text{th}}$, we can estimate the displacement due to the oscillating part of the pump beam power by the following formula: $\Delta h_{\text{osc}} = \Delta h_{\text{cons}} \times (P_L/(P_L + P_H)) \times \ell_{\text{th}}/D = 75$ nm. This leads to a theoretical maximum total relative displacement of the crack faces of 445 nm. Finally, the temperature elevation related to the oscillating part of the probe beam, is of the order of 0.4 K, leading to an acoustic strain of $3 \times 10^{-6}$ and a relative displacement of the crack faces $\Delta h_{\text{ac}} = \varepsilon \times d_H \approx 1$ nm, confirming that the acoustic displacement can be neglected in comparison to the thermoelastic ones.

V. THEORETICAL BACKGROUND

Before presenting and discussing the NFMPA 2D images and 1D images of the crack, it is judicious to present shortly the available theoretical models used in the following to understand the image features. In the case of classic analytic (smooth) nonlinearity of the crack, which could be due to contacting but not tapping/clapping asperities between the crack faces, it is natural to consider the expansion of the stress/strain relationship of the crack in a Taylor series of integer powers. Usually, the quadratic term of these series is the dominant one but higher order terms could be also taken into account if necessary. With the quadratic term, the nonlinear frequency-mixing image of the first side-lobe $A_{\pm 1}$ is proportional to the product of the thermo-elastic stress $\sigma_{\text{ac}}^0$, due to the thermal wave of low frequency $\omega_L$, and of the acoustic stress $\sigma_{\text{ac}}^{\omega_H}$ of high frequency $\omega_H$, both evaluated in the crack position

$$A_{\pm 1}(x) \propto \sigma_{\text{ac}}^{\omega_H}(x)\sigma_{\text{ac}}^0(x).$$

Here, $x$ is the shortest distance between the laser foci, where the generation of the thermo-elastic pump wave and of the acoustic probe wave are centered, and the faces of the crack. The dependence on $x$ in Eq. (1) describes the profile of the $A_{\pm 1}$ image in the case of 1D scan.

Direct three-wave nonlinear processes for quadratic nonlinearity contribute to $A_{\pm 1}$ images in Eq. (1): the frequency-mixing processes $\omega_{\pm 2} = \omega_H \pm \omega_L$, and possible cascade processes with quadratic nonlinearity $\omega_{\pm 2} = \omega_{\pm 1} \pm \omega_L$.

Finally, for the $A_{\pm 3}$ images, also of interest here

$$A_{\pm 3}(x) \propto \sigma_{\text{ac}}^{\omega_H}(x)\sigma_{\text{ac}}^0(x)^3,$$

these are five-wave mixing processes $\omega_{\pm 3} = \omega_H \pm 3\omega_L \pm \omega_{\pm 1}$, which contribute directly, while the other nonlinearities could contribute through different orders of cascades, for example through $\omega_{\pm 3} = \omega_{\pm 2} \pm \omega_L$ and $\omega_{\pm 3} = \omega_{\pm 1} \pm \omega_L$. There are two conclusions that can be derived from the comparison the side-lobes described by Eqs. (1)–(3) and which are useful in the following analysis. First, because of the small amplitude of $\sigma_{\text{ac}}^{\omega_H}$ the amplitude of the images exhibits fast fall with increasing side-lobe order, making the experimental observations of the side-lobes with $n \geq 2$, 3 difficult or even impossible. Second, if $\sigma_{\text{ac}}^{\omega_H}(x)$ falls with increasing distance faster than $\sigma_{\text{ac}}^0(x)$, then with the increasing side-lobe order, the crack images become progressively narrower.

In the case of non-classic non-analytic tapping (or clapping) nonlinearity, the parametric modulation of the
reflected/transmitted acoustic probe field by the breathing crack leads to the following description of the side-lobe images

\[ A_{\pm n}(x) \propto \sigma_{te}^{\pm n}(x)A_{te,n}^{\pm n}(x). \] (4)

Equation (4) describes direct non-cascade modulation of the probe field. For the modulation factors \( A_{te,n}^{\pm n}(x) \), the theory developed in Ref. 33 predicts

\[ A_{te,n}^{\pm n}(x) = \frac{1}{n} \sin \left[ n \arccos \left( \frac{\sigma_{te}}{\sigma_{te}^{\pm n}(x)} \right) \right]. \] (5)

where \( \sigma_{te} \) is the threshold stress, which should be applied to the faces of the crack for the transition of the crack from the initial open state of weak rigidity to the close state of higher rigidity.\(^{33}\) Note that a possible hysteresis in the transitions between the open and close states\(^{33}\) and also the demodulation/rectification of the oscillating thermo-elastic stress at the crack nonlinearity\(^{6,20}\) are neglected in Eq. (5) for simplicity, because these phenomena are not essential for qualitative interpretation of NFMPA images presented below. However, the simplest 1D theory\(^{33}\) has also not taken into account the loading of the crack by the stationary thermo-elastic stresses \( \sigma_{te}^{\pm n}(x) \), which are caused by the time-averaged (stationary) component of the absorbed optical power (both from pump and probe laser beams) and which can never be negligible in 3D geometry. From the physics point of view, the action of stationary thermo-elastic stresses \( \sigma_{te}^{\pm n}(x) \) on the crack surfaces leads to progressive closure of the crack, which can be accounted in Eq. (5) by introducing a decrease of \( \sigma_{te} \) with increasing inhomogeneous stationary heating through the substitution \( \sigma_{te} \rightarrow \sigma_{te} - \sigma_{te}^{\pm n}(x) \)

\[ A_{te,n}^{\pm n}(x) = \frac{1}{n} \sin \left[ n \arccos \left( \frac{\sigma_{te} - \sigma_{te}^{\pm n}(x)}{\sigma_{te}^{\pm n}(x)} \right) \right]. \] (6)

The first conclusion from Eqs. (5) and (6) concerns the amplitudes of the side-lobe images. As it had been already mentioned in the Introduction, the amplitudes of the side-lobes in the case of a pure tapping/clapping nonlinearity diminish with increasing order \( n \) rather slowly. From Eq. (6), this slow trend is \( \propto 1/n \). However, an important role is played in Eq. (6) by the sinus function. On the one hand, it controls the phases of the generated side-lobes. On the other hand, it determines the non-monotonous variation of the side-lobe amplitudes with increasing \( n \), which is an important fingerprint of the tapping/clapping non-classic nonlinearity in comparison with the classic nonlinearity. The relative amplitude ratio of the two subsequent side-lobes of the orders \( n + 1 \) and \( n \) could be larger or smaller than 1 depending on the critical and loading stresses, and thus, could depend on the distance from the crack. To understand the behavior of the \( A_{te,n}^{\pm n}(x) \) qualitatively, at a sufficient level of precision for the interpretation of the NFMPA images later in this paper, we assume that the powers of the pump \( P_L \) and probe \( P_H \) beams increase proportionally (\( P_L \propto P_H \propto P \), where \( P \) is the total power of the two laser beams). Then we evaluate \( A_{te,n}^{\pm n} \) at \( x = 0 \), i.e., in case of centered focusing of both the pump and the probe beams on the crack, when stationary thermo-elastic stress \( \sigma_{te}^{\pm n}(x = 0) \) and oscillating thermo-elastic stress \( \sigma_{te}^{\pm n}(x) \) are proportional \( \sigma_{te}^{\pm n}(0, P) = m \sigma_{te}^{\pm n}(0, P) \), where \( m \) is the proportionality factor. Equation (6) predicts that generation of the side-lobes by a non-analytic nonlinearity, introduced in Ref. 33, starts abruptly at the threshold \( P = P_{cr} \), when the increasing optical power makes the argument of the arcsin function in Eq. (6) equal to 1 (\( \sigma_{te} = \sigma_{te}^{\pm n}(0, P_{cr}) + \sigma_{te}^{\pm n}(0, P_{cr}) = (1 + m) \sigma_{te}^{\pm n}(0, P_{cr}) \)). This corresponds to the beginning of the tapping process, where in average the crack is in the open state, but around the maximum of the oscillating loading by the thermal wave, i.e., by \( \sigma_{te}^{\pm n} \), it enters temporarily in the closed state. The continuous increase in laser power then transfers the crack at \( P = P_{clap}^{\pm n} \), when \( \sigma_{te} = \sigma_{te}^{\pm n}(0, P_{clap}^{\pm n}) = \sigma_{te}^{\pm n}(0, P_{clap}^{\pm n}) \), to the clapping regime, where in average over a period, the crack is in a closed state (more rigid state). Around the minimum of the oscillating loading by the thermal wave, the crack enters temporarily in the open state. When the excitation power increases further above \( P_{clap}^{\pm n} \), the crack motion depends importantly, even qualitatively, on the introduced parameter \( m = \sigma_{te}^{\pm n}(0, P)/\sigma_{te}^{\pm n}(0, P) \). If \( m > 1 \), the stationary loading is stronger than the amplitude of periodic thermal wave loading and, at the critical power \( P = P_{clap}^{\pm n} \), the crack is completely closed. This takes place when the argument of the arcsin function in Eq. (6) becomes equal to \( -1(\sigma_{te} = \sigma_{te}^{\pm n}(0, P_{clap}^{\pm n}) - \sigma_{te}^{\pm n}(0, P_{clap}^{\pm n}) = (m - 1) \sigma_{te}^{\pm n}(0, P_{clap}^{\pm n})) \). In this case, the clapping stops completely, and for \( P > P_{clap}^{\pm n} \) the non-analytic nonlinearity of the breathing crack disappears. There is no more generation of the side-lobes by this non-analytic nonlinearity. The possibility to close the crack completely locally by the stationary heating has been recently proved experimentally.\(^{25,26,39,40}\)

However, if \( m < 1 \) then the clapping could continue with the increasing excitation of the crack. In particular, when \( m < 1 \) and \( \sigma_{te}^{\pm n}(0, P) \gg \sigma_{te} \), the modulation of the side-lobes does not depend anymore on the excitation level

\[ A_{te,n}^{\pm n}(x = 0) = \frac{1}{n} \sin[n \arccos(-m)]. \] (7)

In Fig. 3, we illustrate the dependences of the modulation functions \( A_{te,n}^{\pm n}(0) \) for \( n = 1, 3 \) on the normalized stress \( \tilde{\sigma} = \sigma_{te}^{\pm n}(0, P)/\sigma_{te} \)

\[ A_{te,1}^{\pm n}(0) = \sqrt{1 - \left( \frac{1}{\tilde{\sigma}} - m \right)^2}, \]

\[ A_{te,2}^{\pm n}(0) = A_{te,1}^{\pm n}(0) \left( \frac{1}{\tilde{\sigma}} - m \right), \] (8)

\[ A_{te,3}^{\pm n}(0) = A_{te,1}^{\pm n}(0) \frac{1}{3} \left[ \left( \frac{1}{\tilde{\sigma}} - m \right)^2 - 1 \right], \]

for three different values of the parameter \( m = 0.25, 0.75, \) and 1.05.

The presentation in Fig. 3 takes into account the negative phases of the \( A_{te,n}^{\pm n}(0) \) functions for some values of the excitation.\(^{33}\) This is important if the non-analytic nonlinearity acts in parallel with a smooth analytic classic nonlinearity.
The parts (a) and (b) of Fig. 3 are peculiar amplitude dynamics. However, the moduli of site phase side-lobes which could be canceled out or produce because the processes could lead to the generation of opposite phase side-lobes which could be canceled out or produce a peculiar amplitude dynamics. However, the moduli of these functions $|A_{te,c}^{m}(0)|$, which are important in the case of the negligible classic nonlinearity of the crack, are also presented in Fig. 3. The illustrations in Fig. 3 confirm the earlier statement that the sinus function in Eq. (6) does not modify the general order of magnitude of the ratios $|A_{te,c}^{m}(0)|/|A_{te,c}^{m}(0)|$ predicted as $|A_{te,c}^{m}(0)|/|A_{te,c}^{m}(0)| \propto k/n$. The parts (a) and (b) of Fig. 3 demonstrate that in the case $m < 1$, when the crack cannot be completely closed, the number of zeros in the modulation functions depends on the parameter $m$. For example, the third side-lobe can have a single zero (Fig. 3(a)) or two (Fig. 3(b)). Moreover, depending on this parameter asymptotically at highest levels of the excitation, i.e., for $\sigma \gg 1$, it could be, in accordance with Eq. (7), either $|A_{2}| > |A_{1}|$ (Fig. 3(b)) or $|A_{1}| > |A_{2}|$ (Fig. 3(a)). The part (c) of Fig. 3 illustrates the case $m > 1$ of the complete closure of the crack. In this case, the modulation function of the second side-lobe has always a single zero while that for the third side-lobe has two zeros between the threshold for tapping and the complete closure of the crack.

An additional important prediction, which can be derived from Eqs. (6) and (8) and Fig. 3, is that the side-lobes modulation amplitudes are non-monotonous functions of the laser excitation level. This theoretical prediction leads to expectations of possible spectacular profiles of side-lobes, generated by the non-analytic breathing nonlinearity of the crack. In fact, it is quite straightforward to imagine that when we start to produce a local 1D image described by Eq. (4) in a particular position along the crack, by approaching the crack at $x = 0$ with co-focused pump and probe laser beams starting from $x \to -\infty$, both the stationary and oscillating stresses acting on the crack have the general tendency of increasing with diminishing $x$. Then for sufficiently high excitation levels, the functions $A_{te,c}^{m}(x)$ are non-monotonous with coordinate, because of the non-monotonous behaviors on the excitation level revealed in Eqs. (6) and (8) and Fig. 3. The variation of $A_{te,c}^{m}(x)$ cannot be precisely modeled by Eq. (8) because of the different dependences of $\sigma_{te,c}^{m}(x)$ and $\sigma_{te,c}^{m}(x)$ on $x$, but there are no doubts that this difference does not eliminate the effect of the possible non-monotonous variation of $A_{te,c}^{m}(x)$. This is clear from the analysis of how these functions enter into the argument of the arcsin function in Eq. (6).

Qualitatively speaking, the non-monotonous behavior as a function of the excitation power could be transformed into 1D images which amplitudes are non-monotonous as a function of the distance between the lasers foci and the crack. This is because for a fixed laser power the excitation of the crack increases with diminishing distance $x$. If the non-monotonous behavior of the modulation $A_{te,c}^{m}(x)$ is not wiped away in Eq. (4) by a strong/steep increase of the acoustical stress $\sigma_{ac}^{m}(x)$ with diminishing $x$, then the 1D side-lobe images of the crack contain local maxima at $x \neq 0$, which do not coincide with the position of the crack at $x = 0$. This indicates that crack phantoms could be observed. In the simplest asymptotic situation, when $m > 1$, when the maximum excitation power is high enough for the complete local closure of the crack, and when the laser beams are co-centered, the observation of 2, 4, and 6 phantom cracks could be expected when imaging the evolution of the first, second, and third side-lobes, respectively. These phantoms are equally distributed over both sides of the real crack, while the amplitudes of these side-lobe images exactly at the position of the crack, when the excitation is higher than the threshold for crack closure, will be equal to zero (see Fig. 3(c)). However, in practical situation of imaging, one could expect a smaller number of phantoms. This is because the thermo-elastic excitation is not sufficiently powerful to completely close the crack, because of the smoothing of the phantom maxima by the monotonously increasing acoustic probe field in Eq. (4) or because the contributions to the side-lobe images from the smooth analytic classic nonlinearity monotonously increase with diminishing $x$ (Eqs. (1)–(3)). Another theoretical prediction, which follows from Eq. (6) and is important for the understanding of the possible ultimate spatial
resolution of the NFMPA imaging, is the infinitely fast growth of the side-lobes just above the threshold, \( \sigma_{cr} \equiv \sigma_{cr}^{res}(0, P_{cr}) / \sigma_{cr} = 1/(1 + m) \), for the initiation of the tapping regime. Assuming that the thermo-elastic loading is just above the threshold, i.e., \( \sigma = \sigma_{cr} + \Delta \sigma = 1/(1 + m) + \Delta \sigma \), where \( \Delta \sigma \ll \sigma_{cr} \), we derive from Eq. (8)

\[
A_{\sigma,1-3}(0, \Delta \sigma \ll \sigma_{cr}) \propto \sqrt{\Delta \sigma}.
\]

Equation (9) provides asymptotic description of the behavior of the amplitude modulation functions presented in the insets of Fig. 3 near the threshold.

VI. EXPERIMENTAL OBSERVATIONS: SPATIAL RESOLUTION IN NFMPA IMAGING

Our first experimental observation is relative to the amplitudes of the side-lobe images. We present in Fig. 4 the 1D scans of the three left side-lobes at three consecutive positions/distances parallel to the crack direction (y = 1.2 mm, 1.3 mm, and 1.4 mm). In all these images, there is about a 20 dB difference between the amplitudes of the \( A_{-1} \) image and the comparable amplitudes of \( A_{-2,3} \) images. The same observation can be done at many other positions along the crack, for example at y = 0.2, 0.3, 0.5, 0.6, 0.9, 1.1, 1.5, 1.6, 1.9, 2.1, 2.2, and 2.3 mm. These observations are inconsistent with the hypothesis of a single mechanism of the crack nonlinearity effective in imaging, because in the case of the purely classic analytic nonlinearity, the strong difference in amplitudes could be expected not only between \( A_{-1} \) and \( A_{-2,3} \) but also between \( A_{-2} \) and \( A_{-3} \) could be expected. In contrary, in the case of the purely tapping/clapping nonlinearity, the amplitude ratio for the consecutive side-lobes could be expected to be of the order of 10 dB (see theoretical Sec. V). At the same time, the comparison of the \( A_{-2} \) and \( A_{-3} \) images in Fig. 4 demonstrates that, in general, the amplitudes are not just comparable but one or another can be larger depending on the position along the crack, i.e., depending on the local rigidity of the crack. In fact, we observe that \( A_{-3} > A_{-2} \) at y = 1.2 mm (Fig. 4(a)), \( A_{-3} \cong A_{-2} \) at y = 1.3 mm (Fig. 4(b)), and that \( A_{-3} < A_{-2} \) at y = 1.4 mm (Fig. 4(c)). All three cases are repeated several times along the crack. For example, the first situation is also realized at y = 0.9, 1.1, and 2.3 mm, the second at y = 0.6, 2.1, 2.2 mm, and the third at y = 0.0, 0.5 and 1.9 mm. These observations are characteristic to tapping/clapping non-analytic nonlinearity, which is expected to manifest itself in the excitation level dependent and local crack rigidity dependent ratio of \( A_{-3} \) and \( A_{-2} \) (see theoretical Sec. V and Fig. 3). These observations are inconsistent with the manifestations of the classic nonlinearity, where both due to the direct and cascade multi-phonon processes, it could be expected that \( A_{-3} \ll A_{-2} \). It is worth noting once again that the non-classic non-analytic nonlinearity can lead both to \( A_{-3} > A_{-2} \) and to \( A_{-3} < A_{-2} \) depending on the relative level of the crack-loading thermo-elastic stresses and of the critical stress necessary for the transition of the crack from tapping to clapping, i.e., from the open to the close state. Thus, in view of the earlier discussed theoretical expectations, we conclude that the results presented in Fig. 4 and also those listed in the text above are consistent with the following hypothesis: in some positions along the crack and for the fixed excitation level of the discussed experiments, both smooth classic nonlinearity and tapping/clapping nonlinearity contribute to the generation of the first side-lobes \( A_{-1} \), while the \( A_{-2,3} \) images are dominantly excited by the tapping/clapping nonlinearity.

The second experimental observation concerns the relative widths of the crack images from side-lobes of different orders. In Fig. 5, 1D images at y = 0.4, 0.8, and 1.6 mm are presented. They clearly demonstrate that in these positions along the crack, for the considered laser excitation level, the 1D \( A_{-1} \) images only look broader than the \( A_{-2,3} \) images, because of the wider wings, while formally the widths of all three side-lobes images at −10 dB level are comparable (see Fig. 5(c)). In Fig. 5(a), the width of the first side-lobe image is comparable with the width of the third side-lobe image. In Fig. 5(b), the width of the \( A_{-1} \) image is comparable with that.
light penetration depth in the glass sample. In 2D geometry, the dimension of the pixel, which are both of the same order could be considered for the estimates as being of cylindrical distribution of stresses from the probe acoustic field, which scale in the discussed experiments. First, it is the spatial distribution of the cracks, where the width of all side-lobes at −10 dB level is controlled by the contribution to the NFMPA imaging from the threshold-type non-classic non-analytic tapping/clapping nonlinearity of the crack. Surelly, the balance between the contributions of the two nonlinearities to the $A_{-1}$ images is delicate and depends on the position along the crack, i.e., on local crack rigidity, and on the possible deviation from the classic nonlinearity with increasing stationary thermo-elastic loading. By careful examination of the 2D images presented in Fig. 3, it is possible to identify the particular positions along the crack, where the classic nonlinearity is so strong that it importantly degrades the spatial resolution of the $A_{-1}$ images relative to the spatial resolution attainable in $A_{-2}$ and $A_{-3}$ images. As an example, we present in Fig. 6 the 1D images at $y = 0.3$ mm and $y = 1.0$ mm.

From the images presented in Fig. 6, it can be concluded that in these positions along the crack, the contribution of classic nonlinearity to the $A_{-1}$ images dominates over the contribution of non-classic nonlinearity, and as a result these would be expected only at $x \approx 1.5$ mm. So, this is the faster 3D divergence of the acoustic probe wave, which takes place for at distances $x \geq \ell_{\text{pen}} = 300 \mu m$, that will make the acoustic stress decreasing at a scale comparable to the scale of the $A_{-1}$ image wings. Thus, the role of the acoustic 3D field divergence in the formation of these wings decay cannot be neglected a priori. The stationary temperature distribution at the surface of our glass sample has a spatial half-width of 420 $\mu$m at −10 dB, while the half-width of the distribution of the thermal wave amplitude at 1 Hz frequency is expected to be slightly narrower due to the thermal wave additional exponential attenuation. The latter prediction can be derived differently considering the thermal wavelength $\ell_{\text{th}} \approx 420 \mu m$, i.e., $\approx 480 \mu m$ at the level of −10 dB, and by accounting for the expected additional geometrical divergence due to the non-1D geometry of the heat transport, i.e., to the transition from 2D to 3D thermal conduction process.

These estimates demonstrate that the width of the $A_{-1}$ images in Fig. 5 could be potentially influenced by both thermo-elastic and acoustic field distributions outside the co-focused beams. The monotonous character of the $A_{-1}$ wings in Fig. 5 importantly supports the hypothesis that the $A_{-1}$ wings are excited by nonlinear interaction through analytic classic smooth nonlinearity of the crack (see Eq. (1)). Then the similar widths of the $A_{-1}$, $A_{-2}$ and $A_{-3}$ images at −10 dB level in Fig. 5 could be attributed to dominant contribution of the non-analytic non-classic threshold-type breathing nonlinearity of the crack, which dominates over the classic one when the tapping/clapping of the crack is activated. The results presented in Fig. 5 demonstrate that in these particular positions along the crack, for a fixed excitation level, the tapping/clapping of the crack is initiated only in the vicinity of the common focus of the pump and probe laser beams. However, the abrupt increase of the modulation side-lobe amplitudes just above the threshold (see Eq. (9) and the insets in Fig. 3) guarantees that the non-analytic nonlinearity controls even the width of the $A_{-1}$ images if the classic nonlinearity of the crack is relatively weak. Thus, the structure of the $A_{-1}$ images revealed in Fig. 5 supports the hypothesis that two different mechanisms of nonlinearity contribute to these images, while the contributions of classic nonlinearity in Eqs. (2) and (3) to $A_{-2}$ and $A_{-3}$ images, respectively, is negligible.

Surelly, the balance between the contributions of the two nonlinearities to the $A_{-1}$ images is delicate and depends on the position along the crack, i.e., on local crack rigidity, and on the possible deviation from the classic nonlinearity with increasing stationary thermo-elastic loading. By careful examination of the 2D images presented in Fig. 3, it is possible to identify the particular positions along the crack, where the classic nonlinearity is so strong that it importantly degrades the spatial resolution of the $A_{-1}$ images relative to the spatial resolution attainable in $A_{-2}$ and $A_{-3}$ images. As an example, we present in Fig. 6 the 1D images at $y = 0.3$ mm and $y = 1.0$ mm.

From the images presented in Fig. 6, it can be concluded that in these positions along the crack, the contribution of classic nonlinearity to the $A_{-1}$ images dominates over the contribution of non-classic nonlinearity, and as a result these
images are about 3 times broader than the \( A_{-2} \) and \( A_{-3} \) images, where the situation is just opposite, i.e., the non-classic nonlinearity dominates in imaging over the classic one.

It is worth mentioning, here, that theoretically it could be possible that non-classic non-analytic nonlinearity controls not only the widths of all three side lobes as it takes place in the case depicted in Fig. 5 but also the complete form of the side-lobes including their wings. Experimental confirmation of this was obtained by us in one of the experiments conducted with a crack prepared in the similar conditions but on a different sample. Therefore, the resonant frequency \( f_H \) is different (\( f_H = 70.6 \mathrm{kHz} \)). The other parameters are also different: the powers \( P_H \) and \( P_L \) are 90 and 120 mW and the diameters of the pump and probe beams are 190 and 212 \( \mu \mathrm{m} \), respectively. Besides the pump frequency is equal to 2 Hz and the acoustic wave is now excited by a 532 nm cw laser, 100% intensity modulated. Those different changes however do not modify the physical phenomena and discussion herein. By carefully examining the 1D images, it was possible to localize the point of the crack where the contribution of the classic nonlinearity is negligible even in the wings of the \( A_{-1} \) image. In Fig. 7, all the side-lobe images have similar width and all can be interpreted as being generated due to non-classic non-analytic parametric interaction process. Please note that the amplitudes of all three side-lobes in Fig. 7 are of the comparable amplitudes, which is an essential feature of the non-classic mechanism of their generation.

This experimental part can be concluded by the statement that, in general, the NFMPA imaging from high order side-lobes, \( n = \pm 2, \pm 3, \ldots \), could be advantageous in terms of spatial resolution in the regime of tapping/clapping crack, because the images at the first side-lobes \( A_{-1} \) are the most influenced ones among the others by the smooth classic nonlinearity. In the absence of tapping/clapping of the crack, this classic nonlinearity would provide under particular conditions, importantly broader images. In other words, imaging at higher side-lobes provides better chances to avoid possible degrading influence on the spatial resolution of the classic nonlinear frequency-mixing processes. The imaging at the higher order side-lobes has important advantages in the case where the imaging system operates just near the threshold for the initiation of the clapping/tapping of the crack. Indeed, in this case, the abrupt increase in the side-lobe amplitudes, which is a characteristic feature of the non-analytic nonlinearity (see Eq. (9) and the insets in Fig. 3), can potentially lead to the imaging with a spatial resolution limited by the widths of the pump and probe laser beams only.

VII. EXPERIMENTAL OBSERVATIONS: EVOLUTION OF IMAGES WITH INCREASING EXCITATION LEVEL

In the previous part, we demonstrated the rich physical information that could be obtained on the nonlinearities of the crack even at imaging with fixed pump and probe laser powers, due to the variation of the crack rigidity along the crack length. In fact, the theoretical formulas for the non-analytic tapping/clapping nonlinearity, Eq. (6), predict that the side-lobes images should depend on the initial crack rigidity, which influences the critical stress necessary for the initiation of the tapping/clapping regime, and also on the thermo-elastic loading. In the first part of the experimental results (Sec. VI), we exploited the variation of the crack rigidity along the crack path, while the imaging system was used for fixed powers of the pump and probe beams. In the experiments described below, and compared with the previous ones, the imaging of the same crack is realized at a
A roughly 50% larger power of the pump beam: the pump power was increased from 76 to 106 mW. The other experimental parameters (the probe power, and the pump and probe frequencies and beam diameters) are identical to the previous experiment.

The 2D images obtained under this stronger laser excitation are presented in Fig. 8. The analysis of the images in Fig. 8 and their comparison with lower-power image in Fig. 2 confirm the theoretical expectation on the possible spectacular variation of the ratio $A_{\pm 2}/A_{\pm 3}$ of the higher order side-lobe amplitudes with increasing excitation power and on the imaging of the crack phantoms, which are the most visible in the second side-lobe images at some positions along the crack between $y = 0$ mm and $y = 2.5$ mm. Figure 9 illustrates the variations of the 1D images in the neighbor positions $y = 0.2$ mm and $y = 0.3$ mm with increasing excitation power of the imaging system. The results in Fig. 9 are consistent with the hypothesis that the crack in $y = 0.3$ mm is softer than in the position $y = 0.2$ mm and they indicate that we are in the excitation regime (see Fig. 3) where either the increasing excitation or the diminishing of the crack rigidity leads to the diminishing of the amplitude of $A_{-2}$ image and the increasing of the amplitude of $A_{-3}$ image. In Fig. 9(d), the two phantoms of the crack are visible in the second side-lobe image separated by the local minimum corresponding to the real crack position.

The 1D images in $y = 0.5$ mm and $y = 0.6$ mm presented in Fig. 10, similarly to those in Fig. 9, confirm the theoretically expected tendencies of the phantoms formation either by diminishing rigidity of the crack or by increasing the excitation level. The crack in the position $y = 0.5$ mm appears to be softer than in $y = 0.6$ mm. In Fig. 10(d), there are two phantoms separated by a local minimum in the crack position, similar to Fig. 9(d), while in Fig. 10(c) in addition to two phantoms there is a local maximum in the real position of the crack. The observation of the maximum between two phantoms in Fig. 10(c) indicates that the highest excitation levels in the center of the co-focused pump and probe laser beams are exceeding those necessary to achieve the transition from tapping to clapping. In fact, the theoretical formulas in Eq. (8) (see Fig. 3) indicate that $|A_{-2}(0)|$ first grows after the initiation of the tapping/clapping regime, then, after the maximum, it falls down with increasing excitation just until the transition from tapping to clapping is achieved, while later it grows again. So the local maximum between the two phantoms in the $A_{-2}$ image could be the manifestation of the transition from tapping to clapping regime in crack breathing. However, it could also be caused by the competition between the fall in $|A_{-2}(0)|$ and the increase in the amplitude of the probe acoustic waves with diminishing distance between the laser foci and the crack. The contribution to this peak from the classic nonlinearity, which could be potentially rather narrow for higher-order side-lobes (see Eq. (2) and the discussion in Sec. V), is expected to be extremely weak for the higher order side-lobes, although a priori it cannot be excluded. Two phantoms of the crack are also observed in $A_{-2}$ image for higher excitation level $P_L = P_2$ at $y = 0.8, 0.9, 1.7, 1.8, 2.0, 2.1$, and $2.2$ mm distances along the crack.

FIG. 8. The 2D NFMPA images of the crack taken at the level of pump laser excitation which is about 50% higher than the one used for the images presented in Fig. 2.
The wings of 1D images of the third side-lobes in Fig. 10 are also, in general, non-monotonous, however, with importantly smaller amplitude dynamics. The clearest phantoms in $A/C_0^3$ images, which could be attributed to the manifestation of non-classic non-analytic nonlinearity, are observed for higher excitation level $P_L = P_2$ at $y = 2.7$, 3.2, and 3.3 mm (see Fig. 11).

The wings of 1D images of the third side-lobes in Fig. 10 are also, in general, non-monotonous, however, with importantly smaller amplitude dynamics. The clearest phantoms in $A/C_0^3$ images, which could be attributed to the manifestation of non-classic non-analytic nonlinearity, are observed for higher excitation level $P_L = P_2$ at $y = 2.7$, 3.2, and 3.3 mm (see Fig. 11).

Comparison of the 1D images in Fig. 10 obtained at different excitation levels indicates an important broadening of the higher-order side-lobes images for the higher excitation level $P_L = P_2$ relative to images obtained at lower excitation $P_L = P_1$. This could be explained by the fact that in the positions with weak rigidity of the crack, the amplitude of thermo-elastic loading at $P_L = P_2$ could be sufficient for the initiation of the tapping/clapping regime of the crack breathing. This is true even at a distance from the crack, i.e., not necessarily when the pump and probe laser foci overlap at least partially with the crack, as it had been in the case of $P_L = P_1$ (see the previous experimental Sec. VI). In this case, the threshold character of the non-analytic tapping/clapping nonlinearity, which leads to abrupt increase in the side-lobe generation efficiency just above the threshold, plays a negative role in terms of the spatial resolution of NFMPA method. As far as the threshold is overcome, even by the loading from the distance, the side-lobe amplitudes increase abruptly. Another negative feature of the considered non-analytic nonlinearity for spatial resolution of NFMPA imaging is the saturation in the growth of the modulation amplitudes with further increase of the excitation above the threshold (see Fig. 3). Thus, the side-lobe images are broadening with the increasing excitation level because in the wings of the higher order side-lobe images, where the excitation powers are close to threshold of tapping/clapping regime, the amplitude of the image grows faster than in the centre of the image, where the excitation powers are maximum.

The similar conclusions on the possible broadening of the higher-order side-lobe images with increasing excitation power can be also derived from the 1D images in $y = 0.8$, 0.9, 2.0, and 2.3 mm, for example (see Fig. 12). Figure 12 illustrates the particular situation at $y = 0.9$ mm position along the crack where the higher-order side-lobe widths are importantly broadened with increasing excitation, because of their dominant generation by non-classic nonlinearity, while the width of the first side-lobe practically does not change, because of its dominant generation by the classic nonlinearity.

Before presenting in Sec. VIII some other experimental results on 1D imaging, obtained with tighter focusing of the pump and probe beams, which also confirm the generation of...
crack phantoms in the NFMPA images, it is worth noting that, obviously, the splitting of the crack image in two or more separate peaks could be caused not only by the phantoms. We believe that the existence of the few maxima in our images in some positions with \( y \geq 5 \text{ mm} \) even at low excitation level of \( P_L = P_1 \) is rather due to the complex inclined subsurface structure of the crack, with a few possible spatially separated regions of high nonlinearity, than due to discussed above in the theoretical section (Sec. \( V \)) mechanism of non-monotonous modulation related to non-analytic clapping/tapping nonlinearity. This conclusion follows from the examination of the 1D images both as a function of the distance along the crack, i.e., as a function of local crack rigidity, and as a function of the excitation level. An example of such comparison is presented in Fig. 13.

From the comparison of (a) and (c) in Fig. 13, it is clear that the second maximum in the images grows with increasing excitation from/on the left wing of the image and not by the splitting of the main peak of the image into two parts as it would be expected in the case of the non-monotonous modulation mechanism of phantoms formation. From the comparison of (b) and (d), it is clear that the splitting between the local peaks of the images does not change with increasing excitation level as it would take place in the case of phantoms formation due to the non-monotonous modulation mechanism. We conclude that in the images at \( y \geq 5 \text{ mm} \) the observation of two maxima is due to the real existence of two laterally separated regions with high crack nonlinearity. These are not phantoms. The lateral separation of the two highly nonlinear regions could be due to the inclination of the crack and the existence of a localized narrowing of the distance between the faces of the crack below the sample surface. The indications on the existence of local narrow parts in the depth of the crack have been obtained recently both by the atomic force microscopy imaging of the crack faces and by the pulsed nonlinear photo-acoustic experiments in the same glass, as in the current experiments.\(^\text{40}\) The above comparison also proposes the method for phantoms identification. Phantoms could be identified as the imaged crack positions which are changing with increasing/decreasing excitation level.

VIII. EXPERIMENTAL OBSERVATIONS: ONE-DIMENSIONAL IMAGING WITH SPATIALLY SEPARATED PUMP AND PROBE LASER FOCI

To get additional confirmations for the proposed explanations of the crack phantoms and an additional insight in the factors limiting the spatial resolution of the NFMPA imaging, 1D imaging experiments have been conducted at particular positions along a crack with importantly better focusing of both pump and probe beams: the diameters of both beams are of \( 40 \mu \text{ m} \) at the 1/e level in intensity. This is possible by using the cw 532 nm wavelength laser radiation for both excitation beams, split into pump and probe beams that are independently modulated as previously. The thermoelastic wave frequency is conserved at 1 Hz, but the acoustic wave frequency is tuned to the 91 kHz resonance frequency of the studied sample.

In the first experiment, the probe beam of constant power \( P_H = 35 \text{ mW} \) has been stationary centered on the crack while scanning for imaging has been achieved by the pump beam of varying power \( P_L \). In this configuration, the spatial distribution of the probe acoustic field does not influence the profiles of the \( A_{\text{in},n} \) images, which are...
controlled by the stresses $\sigma_{\tau_1}^{\text{NV}}(x)$ and $\sigma_{\tau_1}^{\text{NN}}(x)$ created by the pump laser beam. In Fig. 14, we present the measured dependence on the $P_L$ of the magnitudes of the side-lobes in the position $x = 0$, when pump and probe beams are both focused in the close vicinity of the crack. The results presented in Fig. 14, on the one hand, confirm the theoretical predictions in Eq. (8) and in Fig. 3 on the non-monotonous dependence of the modulation caused by non-analytic nonlinearity on the excitation power. On the other hand, through the comparison of Figs. 14 and 3, they indicate that in this particular position of the crack there is a very important contribution of the classic smooth nonlinearity to the frequency-mixing process for all side-lobes. We have evaluated that the contributions to the frequency-mixing process from the non-analytic nonlinearity just above the threshold for the initiation of tapping/clapping of the crack are in phase with the contributions from the classic nonlinearity. So, qualitatively speaking, the difference between the dependences presented in Figs. 14 and 3 could be attributed to the important contribution of classic nonlinearity, which is increasing with increasing pump power.

In fact, nearly the best spatial resolution in all three side-lobe images has been observed around $P_L = 40$ mW. These 1D images are presented in Fig. 15. Both the amplitudes of the images and the widths are approximately in proportion predicted by Eqs. (1)–(3) for the classic frequency-mixing processes. For example, the widths of the $A_{-1,-2,-3}$ images at −10 dB levels are in proportion 435 µm/235 µm/140 µm ∝ 3/2/1. The higher is the order of the side-lobe the better is the spatial resolution of the imaging. Although the contribution from the tapping/clapping nonlinearity to these images cannot be completely excluded, the comparison of Figs. 14 and 3 rather indicates that contributions from the non-classic nonlinearity start to grow faster than those from the classic nonlinearity only at $P_L \geq 40 - 45$ mW. Only the third side-lobe images are narrowing up to $P_L \leq 70 - 80$ mW in accordance with the expectations that just above the threshold for the tapping/clapping initiation the contribution of the non-analytic nonlinearity to the side-lobe diminishes with the increasing order of the side-lobe. Otherwise the experiments confirm the conclusions from Sec. VII that sufficiently above the threshold, the contributions from the non-classic nonlinearity cause the degradation in the spatial resolution.

Non-monotonous modulations presented in Fig. 14 theoretically (Sec. V) could cause the phantoms in the 1D side-lobe images. The experiments clearly demonstrated the formation and the expected spatial dynamics of the phantoms with increasing pump power in the second side-lobe images (Fig. 16). Moreover, the formation of the phantoms in the third side-lobe images has been observed at lower pump powers that in the second side-lobe (Fig. 17) as it could be expected from the theoretical predictions (Fig. 3).

At $P_L \geq 60$ mW, the phantoms in $A_{-3}$ images disappear from a rather limited observation field of this experiment (Fig. 17). However, the next stage of the phantoms formation in $A_{-3}$ images, expected theoretically because of the few maxima in the dependences $A_{-3}^{\text{Lm}}$ modulation function on the excitation level (see Eq. (8) and Fig. 3) starts at $P_L \geq 100$ mW (Fig. 18) through the appearance of a local minimum on the images in the real position of the crack.

At the same time, it is worth noting, that the formation of the phantoms in the first side-lobe NFMPA image has not
been observed in these experiments. This could be expected because of the corresponding thermo-elastic strain modulation function in Fig. 14, even at the maximum power of the pump, does not exhibit a sufficiently fast decrease after the maximum, i.e., the maximum in Fig. 14 is not enough pronounced. This could be formulated also differently. The absence of phantoms in the $A_{-1}$ images in these experiments is due to a relatively strong contribution to these images of the classic nonlinearity, which does not generate phantoms, as it has been already mentioned earlier in the theoretical section.

The second series of the 1D imaging experiments with improved focusing of the pump and probe beams has been performed by centering the pump of fixed power $P_L = 50\,\text{mW}$ on the crack and by scanning the probe beam with increasing power $P_H$ by discrete steps from one scan to the next one. In this configuration, the spatial distribution of the thermo-elastic stresses generated by the pump laser does not influence the spatial profiles of the NFMPA images. The results for the first right side-lobe image $A_1$ are presented in Fig. 19. From Fig. 19, it is immediately clear that the magnitudes of the first side-lobe in the position $x = 0$, i.e., when pump and probe beams are both focused in the close vicinity of the crack, is a strongly non-monotonous function of probe power $P_H$. And, in fact, as it could be expected from the theory, the phantoms were clearly observed experimentally for the first time in the $A_1$ images (Fig. 20). This is a manifestation of a relatively weak increase of the smooth classic nonlinearity when the increasing probe power induces important non-monotonous variation of the contributions to frequency-mixing from the tapping/clapping nonlinearity of the crack. The formation of the phantoms has been also observed in the higher-order side-lobe images.

In this experimental configuration, the width of the images could be controlled by the spatial divergence of the probe acoustic field $\sigma_{\text{ac}}(x)$ and the influence of the stationary thermo-elastic stresses induced by the probe laser on the closure of the crack. It has been observed that the half-widths of all three side-lobe images are approximately the same and are equal to $\sim 125\,\mu\text{m}$ at $-10\,\text{dB}$ level and only weakly depend on the probe power for $P_H \leq 60\,\text{mW}$. These observations are consistent with the hypothesis that the pump power of $P_L \leq 50\,\text{mW}$ is sufficient to initiate the clapping/tapping of the crack in the examined crack position (this is indicated by the comparable amplitudes of the $A_{2,3}$ images). Also they are consistent with the fact that the influence of the heating caused by the probe laser on the crack closure is relatively weak at these probe powers, and that the

FIG. 17. Formation of the phantoms and their dynamic separation with increasing pump power in the third right side-lobe image. Our images indicate that the crack is situated between the measurement positions at 0 and $-50\,\mu\text{m}$.

FIG. 18. Formation of the second pair of phantoms and their dynamic separation with increasing pump power in the third right side-lobe image. Our images indicate that the crack is situated between the measurement positions at 0 and $-50\,\mu\text{m}$.

FIG. 19. Color encoded presentation of the 1D NFMPA images at the first fundamental right side-lobe obtained by focusing the pump laser beam on the crack and by scanning with the probe laser beam of increasing power. The complete closure of the crack at $P_H \geq 90-100\,\text{mW}$ is observed. Our images indicate that the crack is situated between the measurement positions at 0 and $-50\,\mu\text{m}$.

FIG. 20. Formation of the phantoms and their dynamic separation with increasing pump power in the first right side-lobe NFMPA image. Our images indicate that the crack is situated between the measurement positions at 0 and $-50\,\mu\text{m}$.
profiles of the images are dominantly controlled by the divergence of the high-frequency acoustic field. In 2D geometry, at distances $x \gg (d_H/2) \approx 20 \mu m$, the acoustic stress scales as $\sigma_{ac}(x) \propto \sqrt{d_H/(2x)}$ decreases, and the $-10 \text{dB}$ decrease in the stress amplitude would be expected at $x \approx 170 \mu m$. However, the transition to faster 3D divergence of the acoustic probe wave, which should be accounted for because of the finite penetration depth of the probe laser beam, will make the acoustic stress decrease at the scale comparable to the scale of the $A_1$ image wings. It is worth noting that the theoretically evaluated stationary temperature field on the sample surface, which is induced by the probe, exhibits a $-10 \text{dB}$ decay at $x \approx 200 \mu m$, close to the acoustic stress scale. However, the stationary thermo-elastic stresses induced by the probe beam have in this regime of tapping/clapping frequency-mixing, Eq. (4), much weaker indirect influence, through the function $A_{Ac}^{th}(x)$, on the image profiles, than the influence of the acoustic stress distribution $\sigma_{ac}(x)$, to which the images are directly proportional. In this regime, the contribution to crack loading from the stationary heating produced by the probe excitation is importantly smaller than the contribution from the pump excitation. The control of the image profiles by the acoustic field divergence is also confirmed by the weak dependence of the image widths on probe power in this regime.

However, the increase in the probe power above $P_H \propto 60-70 \text{mW}$ leads to the progressively increasing role of the stationary stresses induced by the probe beam. Through the modification of the threshold for the tapping/clapping of the crack (see Eqs. (6) and (8)) and by realizing this, with increasing probe power, from progressively larger distances these stationary stresses induce the complete spectrum of the theoretically expected phenomena. With increasing probe power, the images are broadening and the formation of the phantoms is observed. As expected theoretically, the formation of the phantoms is first observed in the highest (third) order side lobes. Formation of the phantoms on lower order side-lobes requires additional increase in probe power. Finally, at probe powers $P_H \geq 90-100 \text{mW}$, the complete closure of the crack takes place (see Fig. 19).

Similar estimations as in Sec. IV can be achieved with the present experimental parameters. An evaluation of the relative displacement of crack faces induced by the constant heating of the pump and probe beams, with their respective powers $P_L = 50 \text{ mW}$ and $P_H = 100 \text{ mW}$, provides $\Delta h_{\text{const}} \approx 470 \text{nm}$. The characteristic diameter of the heated zone $D$ is here equal to $800 \mu m$, still larger than the thermal wavelength $\ell_{th}$. Thus, the oscillating part of the pump beam power is evaluated as previously, $\Delta h_{\text{osc}} = 80 \text{nm}$. Complete closure of the crack means that the oscillating thermoelastic stress cannot open the crack. Thus, the crack width can be considered as $\Delta h_{\text{const}} - \Delta h_{\text{osc}} = 390 \text{nm}$, which agrees with the typical width of such cracks, measured by AFM technique. 

**IX. CONCLUSIONS**

The ensemble of our experimental observations demonstrated various manifestations of nonlinear acoustic phenomena in two-dimensional frequency-mixing photo-acoustic imaging of cracks. The theoretical interpretation of the observed images and of their evolution with increasing excitation power is proposed. The documented phenomenon of the phantoms formation in the side-lobe images is attributed to the manifestation of the strong non-analytic non-classic crack nonlinearity, which is essential for the tapping/clapping regime of crack breathing. In general, the formation of the phantoms leads to overall broadening of the crack image and could lead to mistakes in the determination of the local crack position. However, in our opinion, it would be erroneous to claim that the formation of the phantoms is completely non-desirable in NFMPA imaging. As it follows from our experiments and their theoretical interpretation, the phantoms directly indicate the positions along the crack, which are characterized by the weakest rigidity. So the imaging of the phantoms could be potentially useful for fast qualitative inspection of the cracks and determining of their weakest parts.

The progress in understanding of the physics of the NFMPA images formation provides insight on the possible improvements of the spatial resolution of this experimental technique. In general, the spatial resolution of the photoacoustic imaging is controlled by our abilities to generate thermo-elastic and acoustic waves at the shortest spatial scales. In the above considered experiments, the spatial resolution is mostly influenced by the focusing of the pump and probe laser beams on the sample surface. However, the NFMPA technique is essentially nonlinear and we observed the influence of the nonlinear acoustic phenomena on the image formation. Our current observations and theoretical analysis demonstrate that the influence of the classic and non-classic non-analytic tapping/clapping nonlinearities of the crack on the spatial resolution of the NFMPA imaging could be very different. The classic nonlinearity could provide the side-lobe images, which are progressively narrowing with the increasing order of the side-lobe used for the imaging (see, for example, Fig. 15). Potentially the images could be narrower than the focusing of the laser excitation beams. The role of the smooth classic analytic nonlinearity here is similar to the role played by optical nonlinearity in the process of the higher harmonics excitation, where the time duration of the pulses at higher optical harmonics become progressively shorter and shorter than the duration of the laser pulse at the fundamental optical frequency. However, the amplitudes of the images at higher order side-lobes, obtained due to classic acoustic nonlinearity, are strongly and progressively diminishing with the increasing order of the side-lobe used for imaging. The non-classic non-analytic clapping/tapping nonlinearity of the crack could also potentially provide a spatial resolution in NFMPA imaging which is not limited by the focusing of the laser excitation. It is important that, because of the threshold character of the clapping/tapping nonlinearity, the images of comparable widths and of comparable amplitudes could be expected at all side-lobes. In the case of the tapping/clapping nonlinearity, the general tendency of the diminishing of the side-lobes amplitudes with increasing order is much slower than in the case of the classic nonlinearity. The images, narrower than the lasers foci, have not been observed in our experiments yet. However, we have documented the
situations where the spatial resolution for all the detected side-lobes is controlled by the non-classic nonlinearity and the amplitude of all side-lobe images are comparable (see, for example, Figs. 5 and 7). Our experiments also confirmed the theoretical expectations that tapping/clapping nonlinearity can improve spatial resolution of the NFMPA imaging only in the vicinity of the threshold for tapping initiation. At higher laser excitation levels, the amplitudes of the images in the crack position saturate, while the clapping threshold can be overcome by the laser excitation, which is importantly spatially separated from the crack. This leads to image broadening and the phantoms formation.

We see the perspectives of the above reported research in conducting crack imaging with diffraction-limited focusing of the pump and probe laser beams. This should potentially provide images of the crack softness/rigidity distribution along the crack length with a sub-micrometer spatial resolution. These experiments could also lead to the imaging of the distributions of different types of acoustic nonlinearities along the crack length with much better spatial resolution than in the experiments reported above. From the results of the above reported experiments, we can qualitatively judge, with the spatial precision of about 100 μm in positioning along the crack, where the nonlinearity of the crack manifests itself as a classic one, as a non-classic one, and where the crack nonlinearity manifests itself as a mixture of the classic and of the non-classic nonlinearities. However, the experiments with better laser focusing and better spatial resolution are necessary to discriminate the contributions to manifested classic nonlinearity from real classic nonlinearity (of a closed contact between the crack faces, for example) and from the non-classic nonlinearity (from the clapping/tapping contacts, with different thresholds for clapping, distributed along the crack faces in the tested part of the crack). We mean that, in the experiments reported above, the tapping/clapping contacts distributed along the tested part of the crack and exhibiting non-classic type tapping/clapping nonlinearity individually but with the different thresholds, i.e., with distribution of thresholds, could provide collective contribution to manifested smooth classic nonlinearity when their individual contributions are averaged over the distribution of their clapping thresholds. So for the imaging of the distribution of closed and tapping/clapping contacts along the crack length, experiments with improved spatial resolution are highly desirable. The advanced future experiments should be accompanied by the development of the three-dimensional theory of the nonlinear frequency-mixing photo-acoustic processes.

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