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We demonstrate a successful combination of several optical methods of remote sensing (coherent fringe projection illumination (CFP), holographic in-line interferometry (HILI), laser induced fluorescence,) for detection, identification, and characterization of crude oil. These methods enable the three-dimensional characterization of oil spills that is important for practical applications. Combined methods of CFP and HILI are described in the frame of coherent superposition of partial interference patterns. We show that in addition to detection/identification of oil, laser illumination in the green-blue region can also degrade oil slicks. We tested these methods on different surfaces contaminated by oil, which include: oil on water, oil on flat solid surfaces, and oil on curved surfaces of. We use coherent fiber bundles for the detection and monitoring of the laser-induced oil degradation in pipes.. Both continuous wave (CW) and pulsed lasers are tested using pump-probe schemes. This finding allows us to suggest that properly structured laser clean-up can be an alternative environmental-friendly method of decontamination and cleaning, which can be an alternative to chemical methods, which are dangerous to environment Application of holographic amplifier with phase conjugation will allow to increase sensitivity, reduce aberrations from atmospheric distortions and to focus back-reflected amplified beam on the contaminated area thus accelerating laser cleaning.

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# Crude oil remote sensing, characterization and cleaning with ContinuousWave and pulsed lasers.

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## Abstract

We demonstrate a successful combination of several optical methods of remote sensing (coherent fringe projection illumination (CFP), holographic in-line interferometry (HILI), laser induced fluorescence,) for detection, identification, and characterization of crude oil. These methods enable the three-dimensional characterization of oil spills that is important for practical applications. Combined methods of CFP and HILI are described in the frame of coherent superposition of partial interference patterns. We show that in addition to detection/identification of oil, laser illumination in the green-blue region can also degrade oil slicks. We tested these methods on different surfaces contaminated by oil, which include: oil on water, oil on flat solid surfaces, and oil on curved surfaces of. We use coherent fiber bundles for the detection and monitoring of the laser-induced oil degradation in pipes.. Both continuous-wave (CW) and pulsed lasers are tested using pump-probe schemes. This finding allows us to suggest that properly structured laser clean-up can be an alternative environmental-friendly method of decontamination and cleaning, which can be an alternative to chemical methods, which are dangerous to environment. Application of holographic amplifier with phase conjugation will allow to increase sensitivity, reduce aberrations from atmospheric distortions and to focus back-reflected amplified beam on the contaminated area thus accelerating laser cleaning.

## 1. Introduction

In spite of the great need for effective methods of remote detection/identification of crude oil in water and/or on solid surfaces, we still lack of efficient and reliable methods [1]. Spectroscopic methods enable characterization of oil composition [2] while interferometry allows determination of the film thickness [3]. Usually these two approaches need different experimental implementation. We suggest combining a spectroscopic method of laser-induced fluorescence with holographic interferometry in one set-up for reliable detection/identification of oil.

We propose a new measuring/cleaning method for closed surfaces (pipes) applicable to removing contaminants (oil films, fungus, rust) from contaminated instruments and spacecraft parts. This technique relies on a combination of Holographic in-line interferometry (HILI) and Coherent fringe projection (CFP) techniques in one set-up with modification needed to work inside closed surfaces of tubes and pipes. Additionally, we propose a unified approach for modeling of both HILI and CFP that is suitable for remote characterization of contaminants inside these structures. This modeling approach is based on the concept of the coherent superposition of partial interference patterns. We will show that the combination of laser-induced fluorescence and laser induced optical trapping and evaporation will provide more accurate detection and a new method of laser-induced cleaning of contamination. Based on our previous experience with oil contamination, we will use a modified version of fringe projection interferometry to monitor contamination in the pipes, which will include a flexible coherent optical fiber bundle to allow inspection of pipe's inner space. For cleaning of closed surfaces (pipes, tanks) we will use flexible optical fibers. Laser evaporated volatile fractions will be removed by air pumps and the condensate may be used for further analyses. Coherent fringe projection (CFP) techniques proved to be efficient in our previous efforts involving non-contact metrology of microstructured objects.

## 2. Experiments for remote characterization of oil droplets and films using fringe projection interferometry.

The methodology and experimental setup method for determining the size of the droplets of oil on the water surface is based on illumination of the oil/water interface by an interference pattern with known period  $d$ . Figure 1 shows the experimental setup, which uses a DPSS laser with wavelength  $\lambda = 532$  nm. The plane-wave laser beam (diameter 24 mm; (1), Figure 1) illuminates glass plate (wedge angle 1, 2'') with a semi-transparent back plane. As a result, the interference pattern of reflected signals from the front and back surfaces of the plates with period  $d = 3$  mm illuminates the analyzed object (3). Since the plane-wave beam illuminates the glass plate, the reflected beams are also plane waves, and the period of interference pattern does not change with distance.

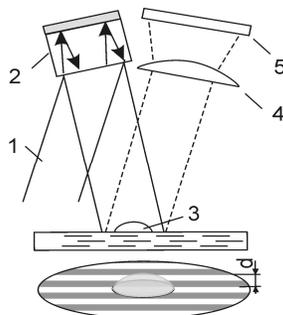


Figure 1. Depicts the experimental set up: 1) Plane-parallel laser beam, 2) Wedged glass plate 3) Object, 4) Lens, 5) Screen

When the droplet of oil spreads on the water surface, there is interference over the entire surface of the object (Figure 2). Interference rings are the result of interference between the reflected beam from the front surface of the oil droplet and a plane wave reflected from the water surface.

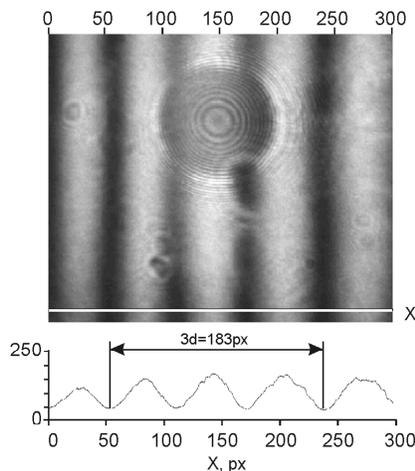


Figure 2. Interference pattern of the drop of motor oil

A graphical editor to determine the pixel-by-pixel distribution of the grey scale along the frame processed the image. A one-pixel-thick line (white line X, Figure 2) was chosen and scanned perpendicularly to the interference pattern along the entire image (total of 300 pixels). Fig. 2 shows the pixels brightness distribution (from 0 to 255 of the grey scale) where the alternation of maxima and minima corresponds to the a priori known fringe pattern period  $d = 3$  mm. By dividing the value of the  $d = 3$  mm period by the number of pixels between the maxima (or minima), we found that 1

mm of the real object dimension corresponds to ~20 pixels of the image. This allows determination of the dimensions of motor oil drop at pixel-by-pixel scanning of the object in graphical editor.

### 3. Pump-probe experiments

The fringe projected interferometry gives us the opportunity to distinguish oil and various organic components on the water surface using different wavelength of laser beam. In addition, using two-color method we can estimate minimum laser energy for oil film degradation. Illumination of the oil-in film by a spot (diameter 1.5 cm) of islands were seen as dark spots with interference fringes. When we applied unexpanded CW green beam ( $P = 15\text{mW}-250\text{ mW}$ ) to the red illuminated spot on or near the oil film spot, then we saw fast rearrangement of the oil island shape. Also, the reflected green beam (observed at a distance of ~ 80 cm) showed fast changing “blooming” interference patterns. Both beams (red probe and pump green) illuminate the samples on a near-normal angle of incidence. The interaction of the interference pattern, generated by the red laser ( $\lambda=633\text{nm}$ ) and the green laser beam ( $\lambda=514\text{ nm}$ ) with oil film spread on water is shown on Fig.3-5 for different laser power.

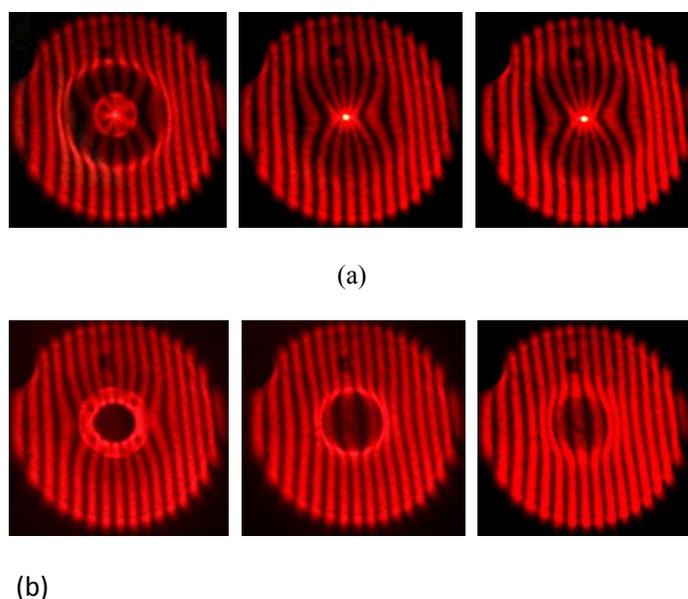
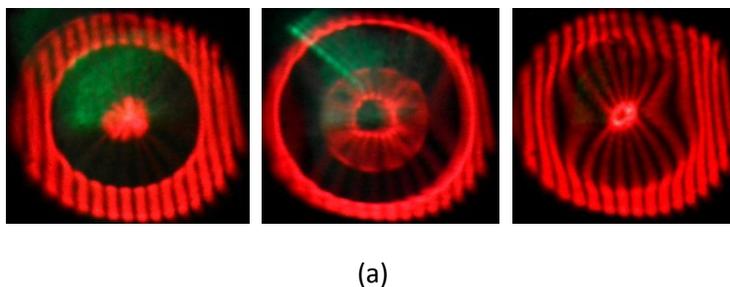
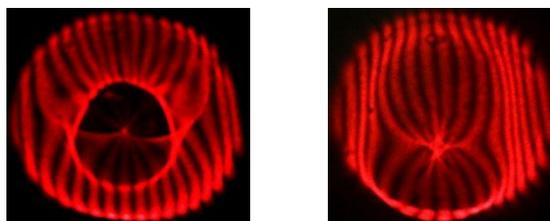


Fig.3. Kinetic of reflection interferometric pattern changes after 5second illumination by green laser -15mW (sequence between images 5 sec.) a ) laser beam on, (b)- laser beam off





(b)

Fig.4. Kinetics of reflection interferometric pattern changes after 5 second illumination by green laser -250mW

(a) laser beam on, (b)- laser beam off (5sec. interval)

During the 12 sec of laser illumination (260 mW) 1ml of oil spot with diameter 80 mm oil spot was destroyed (Fig.5)



Fig.5. Degradation of oil film

Similar experiments, when oil-in-water sample was replaced by the organic solution, shows quite different behavior: illumination by the pump green laser did not produce noticeable changes in the probe red beam reflective pattern. Also, there were not any blooming interference fringes in the reflected pump (green) beam (Fig.6).

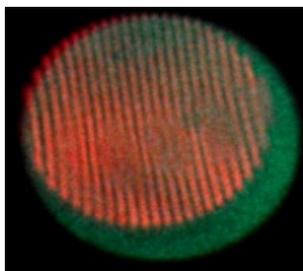


Fig.6. Fringe projection by red-green laser (P=15mW-250mW) on water surface with various organic components

#### 4. Theory of the stripe and circular fringes formation.

We will consider the interference pattern on the screen, formed by the two near-parallel waves reflected from the oil-water surface. Complex amplitude of these two fields can be written as:

$$S_l = (R_l + R_{ld})S_{l0} \exp(-ik_l x) \quad (1)$$

Here, the reflection coefficients from water ( $R_l$ ) and oil spot ( $R_{ld}$ ) and input amplitudes ( $S_{l0}$ ) can be presented in the form

$$R_l = r_l \exp(i\varphi_l), R_{ld} = r_{ld} \exp(i\varphi_{ld}), S_{l0} = \sqrt{I_{l0}} \exp(i\varphi_{l0})$$

Where, subscript  $l=1,2$ ,  $k_l$  is the x-component of the wave vector,  $x$  is a coordinate perpendicular to the lines of the fringe pattern. For near collinear propagation of two beams, we assume that the reflection coefficients for two beams are equal ( $r_1 = r_2 = r$ ,  $r_{1d} = r_{2d} = r_d$ ), so the pattern on the screen may be presented as

$$I = |S_1 + S_2|^2 = I_0(r + r_d)(1 + m \cos(\varphi) + M \cos(\phi) + 0.5mM(\cos(\phi + \varphi) + \cos(\phi - \varphi))) \quad (2)$$

Here,  $I_0$  is input intensity, and phases are

$$\phi = \phi_0 + 2\pi x / d, \varphi = \varphi_0 + (\varphi_l - \varphi_{ld})$$

zero indices mean, that possible constant phase shift may be introduced during reflection from the surface. From Eq.(3) it can be seen that  $\phi$  is the phase difference between the plane (reflected from the unperturbed water) waves that forms partial interference fringe pattern with contrast  $M$ , while  $\varphi$  is the phase difference in the partial interference pattern (with contrast  $m$ ) formed by the plane wave and the wave diffracted from the oil spot. We will model the oil film convex-type shape on the water surface as a product of two shapes: 1) segment of sphere with curvature radius  $R$  that looks from above as a plane-convex lens with radius  $P$  and height  $h$  (sagitta), and 2) step-like function  $F$ . This model of oil thickness changes  $Z$  (as measured from the water level) can be written as:

$$Z(x, y) = \left(\frac{1}{2R}\right)(P^2 - x^2 - y^2)(1 - \exp\left[\frac{b}{R^2}\{P^2 - x^2 - y^2\}\right])^{-1} \quad (3)$$

This shape of the oil film was dictated by an effort to simulate the experimental profile of the oil film. In this simulation parameter  $b$  is chosen as a big number (in the order of  $10^5 - 10^6$ ) in an effort to best fit the experimental shape of the boundary of the oil film on the water. The relation of phase modulation  $\varphi(x, y, z)$  introduced by the oil film can be found from the general expression, known in interferometry of reflective surfaces

$$\varphi(x, y, z) = \left(\frac{2\pi}{\lambda}\right)(\vec{n}\vec{S}) = \frac{4\pi}{\lambda} Z(x, y) \cos(\theta) \quad (4)$$

Here,  $\vec{n}$  is the unit vector of the surface deformation,  $\vec{S}$  is the so-called sensitivity vector, equal to the difference between the reflected and incident wave vectors,  $\theta$  is the angle of incidence (measured from the z-axis).

The experimental interference pattern of the oil-on water spot on the screen (Fig. 2) is compared with the theoretical one. Choosing a set of parameters  $P=0.369$  cm,  $L=0.7$  cm,  $\lambda=532$  nm,  $R=170$  cm,  $M=m=0.5$ ,  $b=8.5 \cdot 10^5$ ,

$$\varphi_0 = \phi_0 = 0$$

we will get the density plot of intensity on the screen and plot the modeled shape of the oil film (cross section in  $xz$  plane) (Fig.3 a,b).

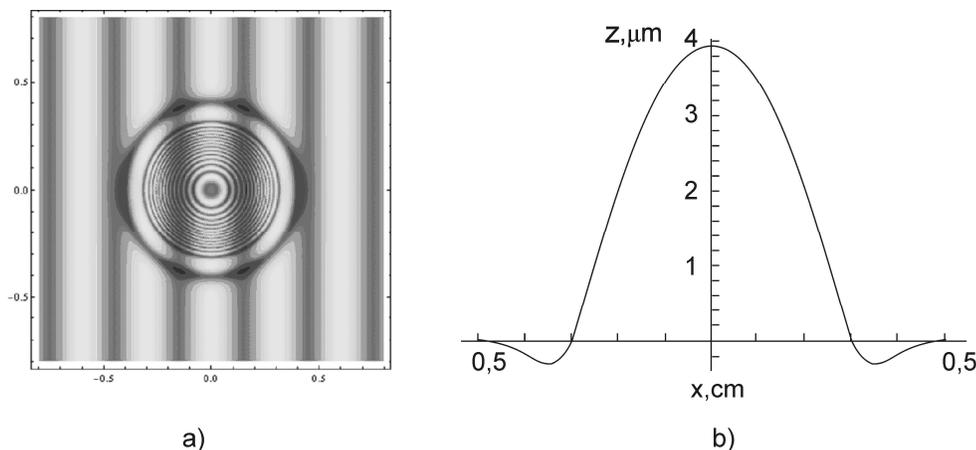


Figure 7. a) Calculated density plot of the combined intensity pattern on the screen; b)  $XZ$  cross-section of the modeled surface profile of the oil spot. For parameters best fitted experimental density plot ( $m = M = 0.5$ ,  $P = 0.3369$  cm,  $L = 0.7$  cm,  $R = 170$  cm,  $b = 8.5 \cdot 10^5$  and assuming no constant phase shift during reflection).

## 5. Comparison of experimental results with theoretical modeling.

In the presented approach for deciphering of the interference pattern from the oil spots, we used a method of probed shapes by modeling oil spot surface profile with functions, according to the symmetry of spot (in our case by convolution of part of sphere with a step-function with several adjusted parameters). Some of parameters, like stripe fringe period  $d$ , contrasts with the partial interferograms  $m$  and  $M$ , circular symmetry is estimated and deduced from the experimental results. By varying the two remaining adjusted parameters (curvature radius  $R$  and parameter  $b$  of step-function), we achieved a best-fitting comparison between experimental and theoretical density plots. This fitting allows us to find maximal height (sagitta) of the modeled shape of the oil circular spot.

Our approach of choosing a probed function for the oil spot surface with parameters, defined by a best-fitting comparison with the experiment may work well for the shapes allowing modeling with analytical functions (like circular, elliptical and similar shapes).

In our case of cylindrical symmetry of the oil film shape is defined by two parameters: radius of spot  $P$  and sagitta (height  $h$ ). From the fringe projection pattern, radius  $a$  was determined as  $P = .3$  cm. Oil film maximal thickness  $h$  (sagitta) was estimated as  $h = 0.375$  microns from fitting density plots (see Fig.7 a,b.).

## 6. Fringe projection with coherent imaging fiber bundle

Fig.4 shows a scheme for inspection of oil films inside the tubes, using coherent fiber bundle (for imaging inside the tubes). In this experiment we tested coherent fringe projection technique, using for illumination a HeNe red laser.

Vertical fringes were formed with the help of a glass plate with a small wedge. Reflection of the collimated laser beam from the wedged glass plate created an interference pattern (fringes) with period of about 1 mm. It is important to note, that in this method of fringe formation, the period of the projected interference pattern (fringes) does not depend on the distance to the illuminated surface. At the same time, for a given fringe period, there is a range of measurable distances

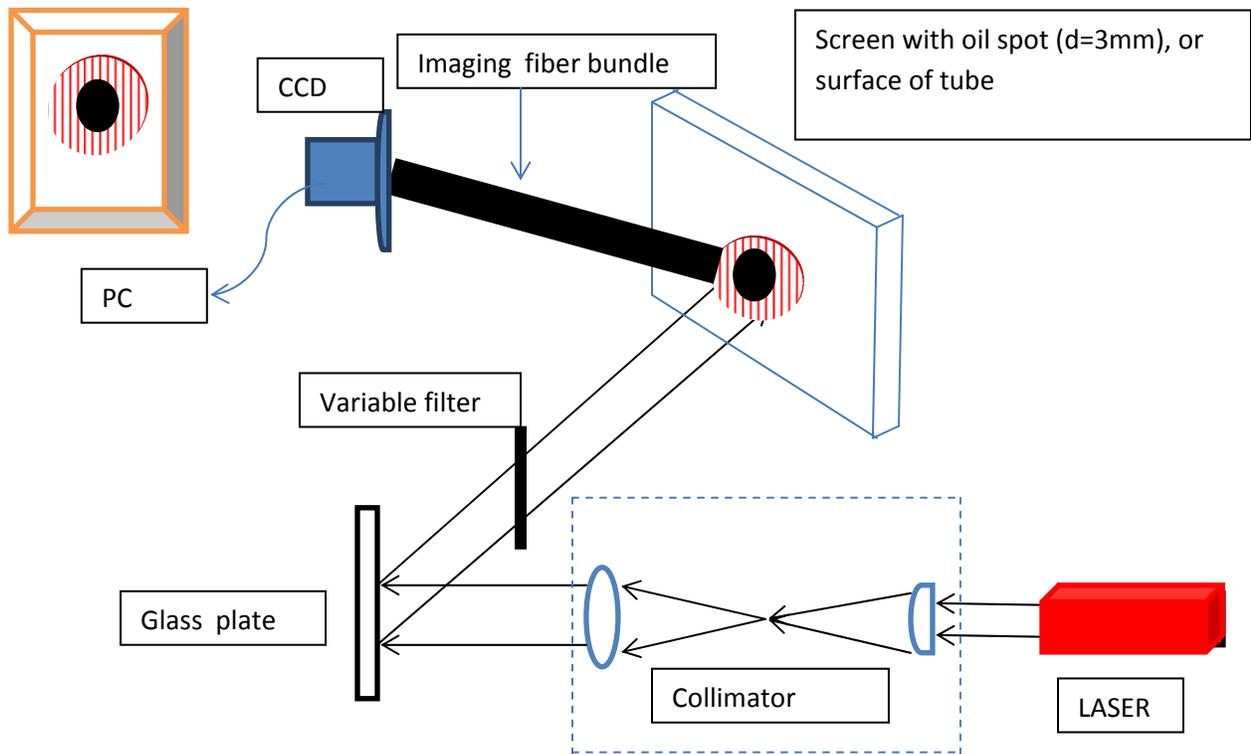


Fig.8, Scheme of inspects of oil spots on the surface of tube or on flat surface with fringe projection for metrology and imaging coherent fiber bundle that project image on the CCD camera with later digital processing on the computer (PC).

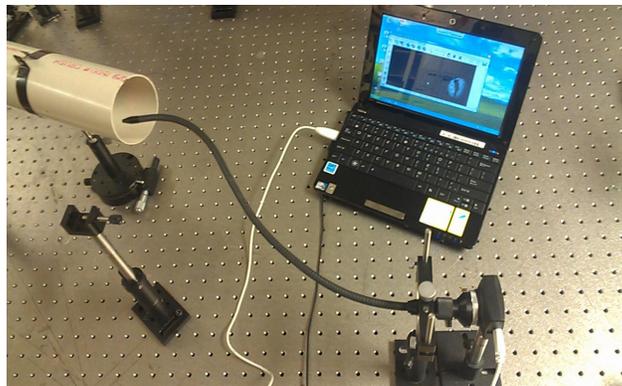


Fig.9 Photo of experimental setup for imaging of crude oil spots inside tubes with coherent fringe projection (by red HeNe laser) and imaging fiber bundle.

Image of the oil spot inside the tube, illuminated by coherent fringe pattern, picked-up and transmitted by the imaging coherent fiber bundle to the CCD camera connected to the PC computer ( as it is shown on the Fig.10).

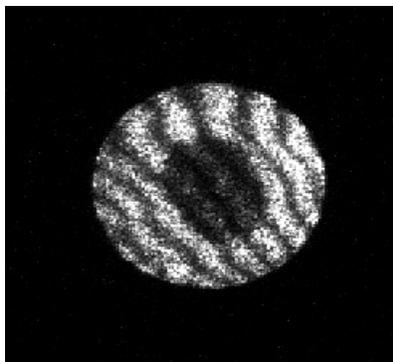


Fig.10 Image of oil spot with coherent fringe projection. Knowing the distance between interference fringes we can determine oil spot size.

From Fig. 10 we can find the size of the oil spot using the number of fringes and fringe period, which is  $\sim 1$  mm, we can conclude that the oil spot size is about 3 mm. The technique with coherent fringe projection also allows determination of the oil spot thickness, as was demonstrated previously in our laboratory for the crude oil spots on the water surface [1,2]. However, to be able to measure the diameter and thickness of the oil spot in the tube, we need to use imaging coherent fiber bundle with higher spatial resolution.

#### 6. Practical simple single-beam scheme is tested with blue laser.

Due to the higher absorption coefficient of the blue light in oil, the laser spot locally heats the oil surface and creates thermal lensing effect and spatially modulate reflectance from the oil-film interface. As a result, several diverging waves emerge from the surface, creating dynamic interference pattern on the screen or on the CCD camera (Fig.11). The kinetics of emerging of this dynamic pattern and structure of interferometric fringes encode thermal and rheological parameters of oil (such as heat conductivity, specific heat, dynamic viscosity, and surface tension).



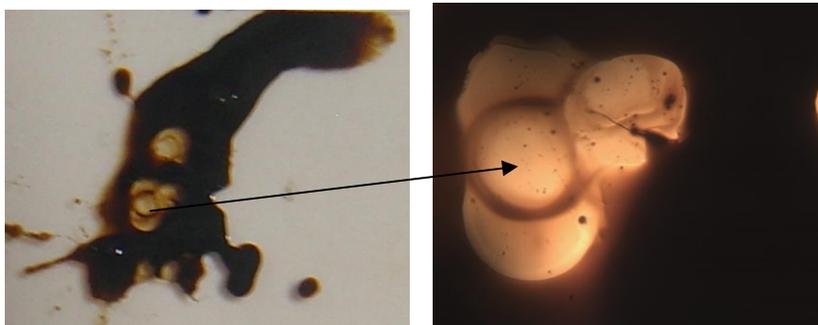
Fig.11. Nonlinear reflection of the blue laser light from the border of the oil film-water interface. The bright spot in the center is the reflection from the water, interference fringes are from the oil film, and the green light is due to fluorescence.

At the same time, absorption of the blue laser (Fig. 11) will generate green fluorescence that will also indicate presence of oil.

## 7. Oil spill degradation/containment by laser light.

Some of the currently used cleaning methods of oil spills cause damage to the environment and pose a risk to human health,.. Oil removal/containment by Lasers offers advantages with respect to these techniques such as the absence of additional residues and a minimal damage to the underlying substrate material.

We have observed rapid degradation of oil film during CW blue laser illumination (wavelength 470 nm, power about 100 mW). Fig.12 shows typical results of such laser degradation with white spots produced to about 2 sec laser exposure. Oil droplet clouds are clearly seen during laser-induced oil film degradation. It is reasonably to expect that properly structured interference pattern contains oil film due to the gradient forces known in the optical trapping technology [ 4,5 ].



M= X40

Fig.12. Degradation of the oil spot by blue laser (470 nm) illumination, right picture show amplified (X40) image of the selected part of the spot.

In addition, we have tested laser cleaning using pulsed Nd YAG laser with parameters: (Fig.13)

( $\lambda=532\text{nm}$ ,  $1064\text{nm}$ ,  $P= 4.9 \text{ W}$ ) cleaning of plastic tube (d=1cm, surface  $7\text{cm}^2$  30 sec



Fig.13 Cleaning of crude oil from the surface of plastic tube using pulsed laser.

## 8. Discussions and future work

We suggest combining several optical methods (coherent fringe projection illumination (CFP), holographic in-line interferometry (HILI), laser induced fluorescence,) for the detection and characterization of crude oil. Combined methods of CFP and HILI are described by coherent superposition of partial interference patterns. We show that in addition to the detection/identification of oil, laser illumination in the green-blue region also degrades oil slicks. We use coherent fiber bundles for the detection /monitoring of the laser-induced oil degradation in pipes.. Both

continuous-wave (CW) and pulsed lasers are tested using pump-probe schemes. We show that properly structured laser clean up can be alternative methods of decontamination and confinement of the oil spots.

Our future work will be focused on using holographic amplification of weak reflected signals using photorefractive materials with simultaneous phase conjugation for reduction of atmospheric aberrations. It is known, that phase-conjugated waves will restore aberrations when double passing the aberration source [9-11]. Phase conjugation can be done fast with simultaneous amplification. Reference [11] reports fast phase conjugation with pulsed lasers using photorefractive  $\text{Sn}_2\text{P}_2\text{S}_6$  crystals. For 7.2-ps pulses at  $1.06 \mu\text{m}$ , it was achieved phase-conjugate reflectivities of up to 45% with very fast build-up times, about 15 ms at an intensity of  $23 \text{ W/cm}^2$  using Te-doped  $\text{Sn}_2\text{P}_2\text{S}_6$ .

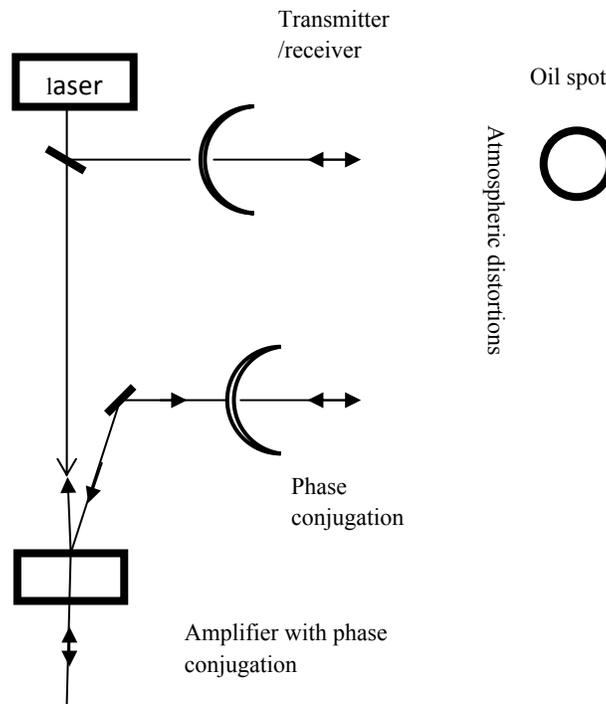


Fig.14 The double-round-trip optical phase conjugate LADAR (Laser Detection and Ranging) system.

Fig.14 shows a double-round-trip optical phase conjugate LADAR (Laser Detection And Ranging) system. The Laser beam illuminates the object (i.e., oil spot on the water) through a transmitter receiver telescope. The back-reflected wave will be very weak and modulated by atmospheric distortions. This returned signal will be amplified by the system using dynamic holographic grating with phase conjugation and directed back to the oil spot by the second phase-conjugate telescope. During two-round-trip passages of the phase conjugate signal, phase aberrations will be reduced and the oil-spot target will be illuminated by the amplified and focused laser beam. Amplification of the signal wave in the holographic amplifier can be done by reading the dynamic holographic grating with a CW or pulsed laser. Using holographic amplification with phase conjugation has several advantages:

- allows increasing sensitivity of detection by amplifying the signal
- allows mitigating atmospheric phase distortions
- focuses amplified phase conjugated signal back to the target (oil spot) which may also be used for removal of oil spots.

Interesting modification of experiments with the high-pressure (50 – 100 atm) oil and water jets (with cavitations in narrow tubes) revealed a new potential for a more efficient cleaning of pipes and tanks. It was shown in recent publications [12-13] that supersonic spindle oil and water jets can produce intense laser-like optical, ultraviolet and X-ray radiation. Experiments were conducted using commercial KMT water jet systems. Optical, UV- and X- ray radiation experiments were explained by strong pressure spikes during cavitation in liquid jets. These experiments were not directly tested for the pipe cleaning, but their results are very encouraging, as they show a new possibility to deliver laser radiation inside the tubes. Intensity of X-ray flux was about  $2.5 \cdot 10^9$  quanta /s = 0.1 Ci, that may be high enough for the efficient crude oil cleaning.

## 9. Conclusion.

We have tested a method of remote detection and characterization of oil spills on water, based on a combination of CFP and HILI. Theoretical modeling of CFP and HILI is based on the introduction of partial interferograms (stripe/circle diagrams), that in paraxial approximations give adequate description of oil spots with near-spherical or elliptical shapes. The suggested method allows determination of the drop size and slicks of petroleum products accurately from a remote distance by illumination of an object using its plane-parallel interference pattern. Determination of oil thickness films on the sea requires pulse laser application to overcome the effect of wind and wind stress on the water. The measurements can be carried out remotely (ship and aircraft) using a parallel laser beam and semi-transparent plane-parallel plates. Structured light illumination, in addition to metrology, can be used for oil spill containment and as alternative means of decontamination. In addition, the fluorescence phenomenon can be used for oil detection. Application of holographic amplifier with phase conjugation will allow to increase sensitivity, reduce aberrations from atmospheric distortions and to focus back-reflected amplified beam on the contaminated area during laser cleaning.

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