Experimental modelling of the physical process of laser tattoo removal

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Abstract. Two model objects are proposed to study the physical mechanism of tattoo removal using nano-, pico-, and femtosecond laser pulses. Experimental results on laser decolourisation of cotton fabric coloured with black tattoo ink and a pig skin sample with tattoo ink deposited on its rear side, in dependence of the duration, energy, and number of laser pulses and the laser spot size, are presented and discussed. It is established that decolourisation of cotton fabric and pig skin without their destruction occurs under irradiation with ultrashort laser pulses and is absent in the case of nanosecond pulses. An analysis of the results obtained has made it possible to draw a number of conclusions, useful for gaining a better insight into the physical mechanism of laser tattoo removal.

Keywords: nano-, pico-, and femtosecond laser pulses; laser tattoo removal technology; cotton fabric; pig skin.

1. Introduction

Despite the rising popularity of tattoos in modern society, demand for their removal has also increased. A particular attention is paid to the quality of removal [1]. The development of laser technologies gives grounds to believe that tattoos can be removed without skin damage, in contrast to such known methods as surgical excision, dermabrasion, or chemical destruction, which lead to the formation of scars [2, 3].

The biological nature of tattoo application/removal is fairly complex. A dye mass introduced (using a needle) into skin stratum decomposes into particles of micrometer and submicrometer sizes. Since the dye material is foreign for human organism, a universal protective reaction occurs in the latter. First, fibrin flakes arise around dye particles, and then fibroblasts begin to synthesise efficiently a connecting tissue, thus forming an insulating capsule [3–6].

The procedure of laser tattoo removal, despite seeming simplicity, is most difficult to implement in laser cosmetology and must be performed by a highly experienced cosmetologist. The mechanism of laser tattoo removal is based on the destruction of the insulating capsule and fragmentation of dye particles (reducing their size). Released dye fragments are absorbed by phagocytes and extracted with lymph flow. A specific laser treatment mode is chosen in dependence of the colour and type of dye and its location depth. Currently, in view of the large variety of dyes used in tattooing, lasers with

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Received 27 September 2018 *Kvantovaya Elektronika* **49** (1) 52–58 (2019) Translated by Yu.P. Sin'kov different wavelengths are applied for efficient fragmentation of dye particles. One generally uses nanosecond pulsed lasers operating in the red and IR regions, because radiation in this spectral range can penetrate deep into the skin and be absorbed efficiently by black, green, and blue dyes, which yield the most widespread tattoo colours [7]. A dye can be efficiently destroyed using different pulsed laser sources: ruby laser with a wavelength $\lambda = 694$ nm and a pulse width $\tau = 28-40$ ns [7–12], alexandrite laser (λ = 755 nm, τ = 50–100 ns, pulse repetition frequency f = 10 Hz) [10–13], and Nd: YAG laser $(\lambda = 1064 \text{ and } 532 \text{ nm}, \tau = 5-10 \text{ ns}, f = 10 \text{ Hz})$ [9, 10, 12, 13]. The laser fluence is chosen in the range from 2 to 12 J cm⁻² [7, 14] in order to destroy the tattoo pigment without visible damage of cutaneous covering (e.g., occurrence bleeding or blisters) in the treated area. The irradiation spot is large in size (1.5-8 mm) [7, 14]. However, it often turns out that two tattoos, being indistinguishable in colour, are removed by laser irradiation at different wavelengths. Therefore, the main question – possibility of implementing a universal laser mode for dye fragmentation without skin destruction - remains open and calls for detailed study.

The development of ultrashort pulse lasers has significantly expanded the potential of laser technologies. To date, lasers with $\lambda = 532$, 755, and 1064 nm; $\tau = 350-750$ ps; and f = 1-10 Hz are used. The laser fluence is varied from 0.2 to 12.5 J cm⁻² [14–21]. It was found that, using picosecond lasers, one can significantly increase the tattoo removal efficiency due to the dye fragmentation into smaller particles. However, since there are no generally accepted algorithms for choosing optimal irradiation conditions, further comparative studies should be carried out in this field.

In view of the complex biological nature of tattoo deposition/removal, we simplified this problem and investigated the effect of irradiation with nano-, pico-, and femtosecond laser pulses on two model objects: a cotton fabric coloured with black tattoo ink and a pig skin sample (extracted skin layer about 2 mm thick) with the same ink deposited on its rear surface. Colouring of cotton fabric is accompanied by dye penetration into the fabric bulk. In contrast, when colouring the pig skin, the dye remains on the rear sample surface.

In this paper, we report and discuss the experimental results on laser decolourisation of model objects in dependence of the laser pulse duration, laser energy, spot size, and number of pulses.

2. Laser destruction of the first model object (coloured cotton fabric)

The first model object was a piece of PEPPY fabric (single grey-beige colour, 100% cotton) with a density of 140 ± 5 g m⁻²,

a fibre thickness of \sim 420 μ m, and a gap between fibres ranging from 130 to 360 μ m.

The fabric was impregnated with True Black tattoo ink (World Famous Tattoo Ink, United States) of black colour. The ink contains acrylic resin, black pigment (Pigment Black 6-PB6, C.I.77266, carbon soot), glycerol, water, isopropyl alcohol, extract of Hamamelis virginiana, and antimicrobial component (DMDM Hydantoin). The black pigment consists of iron oxide (II, III) (Fe₃O₄), iron oxide (II) (FeO), carbon, and extract of campeachy wood (Central America and West Indies). The black pigment consists of magnetite and wüstite crystals, animal (bone) coal, amorphous carbon (soot), and extract of campeachy wood core. The black pigment absorbs radiation in a wide wavelength range, and its reflectance is sufficiently low; however, the highest absorptivity is observed in the range of 600-800 nm [22]. According to the data of [23], the black ink absorption coefficient decreases from 11770 to 5253 cm⁻¹ with an increase in wavelength from 470 to 1064 nm.

2.1. Irradiation with nanosecond laser pulses

Model objects were irradiated on a Minimarker-2 system (OJSC Laser Center, Russia) using a pulsed ytterbium fibre laser ($\lambda = 1.064 \ \mu m$, $\tau = 50 \ ns$, average power $P = 1-20 \ W$, $f = 3-500 \ kHz$, laser focal spot diameter 60 μm). The radiation power and energy were measured with a Gentec-EO SOLO2 microprocessor-based meter (Gentec Electro-Optics, Inc., Canada).

In view of the inhomogeneity of fabric, which is a set of periodically alternating fibres and gaps between them, laser beam was focused onto a fibre, and the light spot was discretely displaced along it. A typical pattern of a cotton fabric coloured with black tattoo ink after irradiation with nanosecond pulses is presented in Fig. 1. The laser fluence Q was varied by chang-



Figure 1. Optical reflected-light images (fivefold magnification) of cotton fabric samples coloured with black tattoo ink, after their irradiation with nanosecond laser pulses having the following parameters: $\tau = 50$ ns; f = 20 kHz; P = 2 W; number of pulses N = 1000; and Q = (a) 0.4, (b) 0.9, (c) 1.9, and (d) 3.3 J cm⁻².

ing the distance between the focusing objective and sample surface.

According to the results of irradiation of cotton fabric by nanosecond laser pulses, we revealed three irradiation modes, characterised by different Q values. The first mode, in which no irradiation traces were observed, occurred at low fluences $(Q < 0.3 \text{ J cm}^{-2})$. The second mode, in which a dark spot was observed in the irradiated region, corresponded to $Q = 0.3-3.3 \text{ J cm}^{-2}$. The third mode, accompanied by the formation of an ablation crater in the fibre, was implemented at $Q > 3.3 \text{ J cm}^{-2}$. The dark area in the irradiated zone is likely to result from carbonisation of the upper fibre layer. We failed to implement an irradiation mode in which the fabric could be decolourised without the formation of a dark spot.

2.2. Irradiation with picosecond laser pulses

This experiment was performed using a pulsed picosecond laser PL2143 (EKSPLA, Lithuania) with $\lambda = 1.064 \,\mu\text{m}$, $\tau = 30 \,\text{ps}$, $f = 10 \,\text{Hz}$, and pulse energy $E_{\text{p}} = 0.9 - 2.3 \,\text{mJ}$. The laser beam was focused into a spot with a diameter $D = 300 \,\mu\text{m}$. The number of pulses was varied (N = 100, 200, 500) at a constant frequency f.

Figure 2 presents optical images of cotton fabric coloured with black tattoo ink, after its irradiation with picosecond laser pulses. The images were obtained by the dark-field method in reflected light, with a fivefold magnification.



Figure 2. Optical images of coloured cotton fabric irradiated with picosecond laser pulses at Q = (1) 1.1, (2) 1.7, (3) 2.0, and (4) 2.8 J cm⁻² and different *N* values.

We revealed decolourisation of fabric (in an area ~ 300 μ m in size) without the formation of a crater at Q = 1.1-2.8 J cm⁻² and N = 100. An increase in the number of laser pulses to 200 caused fabric destruction (occurrence of carbonisation and microdefects), and a crater was formed after exposure to 500 pulses. Most likely, the heat accumulation effect manifests itself under multipulse irradiation [24].

The degree of fabric decolourisation in the laser-irradiated area was determined by the method for estimating the sample colour based on the analysis of digital photographs within the CIE Lab colour model [25], in which the component L determines the CIE lightness scale (psychometric lightness) and varies from minimum (black, 0) to maximum (white, 100), the component a changes from green to red, and the component b changes from blue to yellow. According to the data of [26], when carrying out laser purification, one can interpret L as a degree of fabric decolourisation in the laserirradiated area. Images were analysed and processed using the Adobe Photoshop CS5.1 program, which makes it possible to determine the averaged values of model parameters (L, a, b). Note that the numerical values obtained within the program are converted into generally accepted CIE units by recalculating according to the following expressions: L_{CIE} = $L \times (100/255)$, $a_{\text{CIE}} = a - 128$, and $b_{\text{CIE}} = b - 128$, where L, a, and b are the estimated values of colour components, yielded by the Adobe Photoshop CS5.1 program; and L_{CIE} , a_{CIE} , and b_{CIE} are the values of colour components recalculated into CIE units.

Figure 3 shows dependences of the degree of decolourisation (L_{CIE}) in the irradiated area of cotton fabric coloured with black tattoo ink on the picosecond laser fluence for N =100 and 500. The initial fabric lightness L_{CIE} was 63, and the lightness of the coloured fabric prior to laser irradiation amounted to 7. It can be seen that the coloured fabric exposed to picosecond laser pulses is not only decolourised but its L_{CIE} value exceeds that of the initial (grey-beige) fabric; the degree of decolourisation changes only slightly with a change in the laser fluence in the range from 1 to 2.8 J cm⁻². The degree of decolourisation increases with an increase in the number of pulses. In all measurements the parameter b is positive and does not exceed 3.1, which corresponds to yellow hue, close to the natural (beige) colour of fabric.



Figure 3. Dependences of the degree of decolourisation of cotton fabric coloured with black tattoo ink (in an area irradiated by a picosecond laser) on Q for different numbers of pulses.

2.3. Irradiation with femtosecond laser pulses

This experiment was performed on a laser system including a femtosecond Ti:sapphire laser TiF-100-F4 (Avesta-Project, Russia) and a regenerative amplifier RAP1500 (Avesta-Project, Russia), with the following radiation parameters: cen-



Figure 4. Optical images of cotton fabric coloured with black tattoo ink, after irradiation by a femtosecond laser with Q = 0.3 J cm⁻² and different numbers of pulses: N = (a) 100, (b) 200, (c) 500, and (d) 1000.



Figure 5. Optical images of cotton fabric samples irradiated with femtosecond laser pulses ($E_p = 0.8 \text{ mJ}$, N = 200): (a) coloured fabric, Q =(1) 0.2, (2) 0.4, (3) 0.5, (4) 0.8, and (5) 1.4 J cm⁻² and (b) uncoloured fabric, Q = (1) 0.7, (2) 0.8, and (3) 0.9 J cm⁻².

tre wavelength 800 nm, pulse FWHM ≈ 100 fs, $E_p^{\text{max}} = 1.2 \text{ mJ}$ (TEM₀₀ mode), and f = 10 Hz. The laser beam was directed to the target by an objective with a focal length of 4 cm.

The cotton fabric coloured with black tattoo ink was irradiated by 100, 200, 500, or 1000 femtosecond pulses at E_p = 0.9 mJ; the laser spot diameter in the irradiated area was ~ 600 µm, a value corresponding to $Q \approx 0.3$ J cm⁻² (Fig. 4). As a result, we observed decolourisation of fabric without its destruction; the colour of decolourised area had a white hue. The decolourised area was surrounded by a dark ring. With a decrease in E_p by half, the decolourised area acquired a yellow hue.

To establish the dependence of the degree of fabric decolourisation on the laser fluence, coloured and uncoloured fabric samples were exposed to 200 laser pulses at $E_p = 0.8$ mJ at different values of focused spot size (fluence Q); the latter was changed by reducing the distance between the objective and sample surface with a step of 1 mm in each measurement (Fig. 5).

Decolourisation of coloured fabric without fibre destruction was observed at $Q = 0.2-0.5 \text{ J cm}^{-2}$. The decolourised area had a white hue and was surrounded by a dark ring. At $Q > 0.8 \text{ J cm}^{-2}$ ablation of the fabric fibre occurred. First bright and then dark rings arose around the ablation crater. The dark zone area amounted to 60% of the entire irradiated area.

In the case of uncoloured fabric, destruction with the formation of a crater occurred under the same irradiation conditions as for the coloured one but started at lower fluences $(Q > 0.7 \text{ J cm}^{-2})$. Fabric decolourisation (occurrence of white hue) was observed around the ablation crater in a fibre (the initial uncoloured fabric was grey-beige).

The degree of decolourisation of coloured and uncoloured fabric samples in the area irradiated by femtosecond laser



Figure 6. Dependences of the degree of cotton fabric decolourisation in an area irradiated by femtosecond pulses (a) on Q at N = 200 for coloured and uncoloured fabric samples and (b) on N for the coloured fabric at different Q values.

pulses was determined by the above-described colour estimation method. The dependence of the degree of fabric decolourisation in the irradiated area on Q for N = 200 is presented in Fig. 6a. At Q = 0.2 J cm⁻² the coloured fabric is decolourised to natural colour ($L_{\text{CIE}} \approx 63$). With an increase in Q to 0.3 J cm⁻² the degree of decolourisation of coloured fabric increases by a factor of about 1.3. The parameter bchanges under these conditions from 15.47 (for Q = 0.2 J cm⁻²) to 11.12 (Q = 0.3 J cm⁻²), a value corresponding to pronounced yellow hue, close to the natural fabric colour. With a further increase in Q the degree of decolourisation of coloured fabric changes only slightly, and the parameter b does not exceed 3. As can be seen in Fig. 6b, the degree of decolourisation of coloured fabric increases by about 5% with an increase in N.

3. Laser destruction of the second model object (pig skin with dye)

To perform efficiently laser tattoo removal, it is of prime importance to determine the fraction of the incident laser radiation that reaches the dye location depth under the skin, i.e., to measure the skin sample transmission spectrum. It is known that tattoo ink is introduced into dermis to a depth from 1.1 to 2.9 mm, depending on the part of body, gender, and age of recipient [12].

Experiments were carried out with a pig skin. A skin sample was taken from the abdominal area of a pig. Prior to experiment the skin was stored in a physiological solution. The hair covering and cellular subcutis were preliminary carefully removed. The ex vivo thickness of pig skin sample was 1.9 ± 0.2 mm. The pig skin transmission spectrum was measured with an SF-56 spectrophotometer (OKB Spectr, Russia) (Fig. 7).

Averaging over ten measurements showed the transmittance of a pig skin sample clamped between two object glasses to be $1.3 \pm 0.3\%$ at $\lambda = 800$ nm. A measurement of the pig skin transmittance at $\lambda = 800$ nm with a microprocessor energy meter Gentec-EO SOLO2 (Gentec Electro-Optics, Inc., Canada) gave a similar result: $1.1 \pm 0.3\%$.

Experiment 1. The object of study was a pig skin sample (thickness 1.9 ± 0.2 mm), placed on grey-beige cotton fabric coloured with black tattoo ink True Black (World Famous Tattoo Ink, United States). The sample was irradiated by a



Figure 7. Transmission spectrum (averaged over ten measurements) of a pig skin sample with a thickness of 1.9 ± 0.2 mm.



Figure 8. Optical images of pig skin samples irradiated by femtosecond laser pulses under the following conditions: (a) $Q = 13.0 \text{ J cm}^{-2}$, N = 200 (the laser beam is focused at a distance of ~ 0.5 mm above the skin surface); (b) $Q = 4.0 \text{ J cm}^{-2}$, N = 400 (the beam is focused at a depth of ~ 1.5 mm under the skin surface); and (c) $Q = 1.0 \text{ J cm}^{-2}$, N = 600 (the beam is focused at a depth of ~ 3 mm under the skin surface).

femtosecond laser TiF-100-F4 at $\lambda = 800$ nm and $E_p = 0.87$ mJ. The laser spot size and number of pulses were varied (Fig. 8).

Destruction (ablation) of skin occurred under all irradiation conditions.

Experiment 2. The object of study was a pig skin layer (thickness 1.9 ± 0.2 mm) coloured with black tattoo ink True Black (World Famous Tattoo Ink, United States) from the rear side. A femtosecond pulsed laser beam ($E_p = 0.9 \text{ mJ}$, N =600) was directed onto the front (uncoloured) sample surface. The focused spot diameter was changed in each measurement by varying the distance between the objective and sample surface by 0.5 mm. Several measurements were performed under these conditions (Fig. 9): measurement 1 (the laser beam was focused above the skin surface at a distance of 0.5 mm), measurement 2 (the beam was focused on the skin surface), and measurements 3-6 (the beam was focused under the skin surface at depths from 0.5 to 2 mm). A crater formed in the sample bulk was observed on the front side of skin layer in all irradiation modes. When the laser beam was focused above the sample surface (Fig. 9a, point 1), the crater diameter on the surface was $\sim 90 \,\mu\text{m}$. When focusing the beam on the skin surface, the crater diameter was minimum: $\sim 60 \,\mu\text{m}$ (Fig. 9a,





Figure 9. (a) Reflected- and (b) transmitted-light images of a pig skin sample coloured with black tattoo ink from the rear side, after irradiation with femtosecond pulses at Q = (1) 13.4, (2) 27.2, (3) 11.3, (4) 4.7, (5) 4.1, and (6) 3.5 J cm⁻²; N = 600.



Figure 10. Optical images of the (a) front and (b) rear sides of a pig skin sample coloured with black tattoo ink from the rear side, after irradiation with femtosecond pulses at Q = 3.5 J cm⁻² and N = 600.

point 2). However, no damage of coloured layer was found in both measurements. Beginning with measurement 3, one could observe the formation of a hole on the rear side with a dark ring around it, whose external diameter increased from 170 (Fig. 9b, point 3) to 370 μ m (Fig. 9b, point 6).

Figure 10 shows enlarged (as compared with Fig. 9) optical images of the front and rear (coloured) sides of the pig skin sample irradiated by femtosecond pulsed laser with Q =3.5 J cm⁻² and N = 600 (measurement 6).

This experiment did not reveal any decolourisation (destruction of the dye layer with the pig skin retained intact) but showed that laser radiation must be focused into the sample bulk, because in this case the fluence on its surface decreases, increasing inside the skin, due to which experimentally observed destruction of the dye layer on the rear side of the sample occurs.

Experiment 3. The object of study was a pig skin sample (thickness 1.9 ± 0.2 mm), coloured with True Black tattoo ink (World Famous Tattoo Ink, United States) from the rear side. The laser beam ($E_p = 0.9$ mJ, $\tau = 100$ fs, f = 10 Hz, N = 600) was directed to the front (uncoloured) side of the sample and focused on its rear (coloured) side so that the spot diameter on the front surface was ~530 µm, with $Q \sim 0.4$ J cm⁻² (Fig. 11). It was found that black tattoo ink can be destroyed without skin damage under these conditions.



Figure 11. (a) Reflected- and (b) transmitted-light images (fivefold magnification) of a pig skin sample coloured with black tattoo ink from the rear side, after irradiation with femtosecond laser pulses at $\lambda = 800$ nm, f = 10 Hz, $E_p = 0.9$ mJ, N = 600, $D = 530 \mu$ m, and Q = 0.4 J cm⁻².

4. Discussion

As was mentioned above, tattoo ink penetrates into the bulk of cotton fabric during its colouring; this feature is also characteristic of skin tattooing. Thus, the processes occurring when dye is removed from coloured cotton fabric are similar to those taking place when a tattoo is removed from the skin bulk (with exception of the biological processes leading to removal of fragmented dye particles from the skin bulk after laser irradiation). It was indicated in [27] that the destruction threshold of uncoloured cotton fabric exposed to nanosecond pulses (at $\lambda = 1064$ nm, $E_p \le 450$ mJ, $\tau = 5-10$ ns, $f \le 10$ Hz, N = 100) is 2.5–3 J cm⁻², i.e., lies in the range of fluences (2–12 J cm⁻²) of nanosecond lasers used for tattoo removal [7,14].

It was found experimentally that the fluence corresponding to the ablation threshold of coloured cotton fabric decreases with decreasing laser pulse duration (Q = 3.3, 3, and 0.8 J cm⁻² for nano-, pico-, and femtosecond pulses, respectively). This result is quite natural. A transition from one time scale of irradiating laser pulses to another is accompanied by an increase in the laser power density $q = Q/\tau$ ($q \approx 7 \times 10^7$, 10^{11} , and 8×10^{12} W cm⁻² for nano-, pico-, and femtosecond pulses, respectively); as a result, the probability of multiphoton light absorption in the ink material (which determines its heating) increases.

Decolourisation of cotton fabric without its destruction did not occur under irradiation with nanosecond pulses, although the latter are widely used in laser tattoo removal. The point is that the ink destruction temperature is much higher than the fabric (skin) destruction temperature; therefore, the thermal conditions under which the dye is destroyed without skin damage cannot be implemented. This result is in agreement with the experimental data on tattoo removal [1].

Decolourisation of fabric without destruction was observed under its irradiation with pico- and femtosecond pulses. This is most likely related to an increase in the probability of multiphoton absorption, which leads to an increase in the temperature of solid ink particles up to the evaporation point. An evidence is the presence of a dark ring around the ablation zone observed during the laser treatment of coloured cotton fabric and skin. In this case, conditions for sublimation of organic materials (i.e., their destruction without carbonisation) can be provided.

Decolourisation without destruction was implemented using femtosecond irradiation of ink through a skin layer 1.9 ± 0.2 mm thick, when the laser beam was focused in the bulk of pig skin sample, with a fluence on the sample surface of ~ 0.4 J cm⁻². This result is a good premise to search for optimal modes of dye removal without skin destruction using ultrashort laser pulses.

Thus, the study of irradiation of model objects with nano-, pico-, and femtosecond laser pulses made it possible to analyse a number of specific features of laser decolourisation of tattoo ink in dependence of the laser pulse duration, radiation energy, laser spot size, and the number of pulses. Indeed, the treatment of skin with nanosecond laser pulses is accompanied by its destruction, which is confirmed by the practice of bulk tattoo removal, whereas the laser treatment using ultrashort pulses, due to the change in the mechanism of interaction between the laser radiation and material, is promising for implementing dye decolourisation without destruction of the surrounding biological tissue. High laser beam intensities, characteristic of pico- and femtosecond ranges, reduce the fluence necessary for dye destruction at ultrashort pulse durations. No size effect (influence of laser spot size on the irradiation result) was observed. However, smooth variation in the spot size on the sample surface by focusing radiation into the sample bulk made it possible to determine the irradiation conditions under which the ink deposited on the rear side of skin sample is decolourised. It was also shown that, in the case of multipulse treatment, an increase in the number of irradiating pulses may cause skin ablation as a result of heat accumulation.

In sum, the investigation of the modes of laser removal of black tattoo ink from cotton fabric revealed that, using picoand femtosecond pulses, one can remove the dye from this fabric without any fabric damage traces that can be seen in microscope (with a fivefold magnification). It was shown experimentally that, when the threshold fluence is exceeded, cotton fabric decolourisation depends only slightly on the number of pico- and femtosecond pulses. It was established that a pig skin sample can be purified from ink deposited on its rear side without damaging skin using irradiation with femtosecond pulses through a skin layer with a thickness of 1.9 ± 0.2 mm.

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