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Laser Treatment of Tattoos: Basic Principles

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Abstract

Tattooing has become very popular worldwide during the past decades, and millions of people have one or many tattoos at different anatomical sites. The color of tattoos is mainly black, followed by red, green, blue, and other colors. A part of the tattooed people regret tattooing or have permanent problems with tattoos and therefore seek for tattoo removal. Tattoos consist of solid pigment particles in the skin. Thus, tattoo removal requires fragmentation of these permanently incorporated particles. The gold standard of tattoo removal is laser therapy. Short light pulses at high intensities are applied to the tattooed skin surface. The laser light penetrates the skin and is selectively absorbed in the pigment particles. The absorbed laser light leads to heat-up and fragmentation of the particles. Due to the complex chemistry of the various tattoo pigments, the efficacy of this fragmentation process is frequently unpredictable. Due to the short and intense pulses, nonlinear effects of light and thermal properties of tattoo particles may play a role, and the assumptions of selective photothermolysis may not reflect the real process of tattoo particle fragmentation as a whole. In case fragmentation occurs, the concentration of pigment particles in the skin decreases, yielding a fading of the tattoo color in the skin. Laser therapy is most effective in black tattoos and less effective for colored tattoos. The rate of side effects is low due to the selectivity of the treatment. Laser light may change the chemistry of the tattoo pigments and hence provoke toxic decomposition products.

Tattooing is an ancient procedure to stain the skin of humans, and its history goes back thousands of years. In the past, people used various colorants and devices to stain the skin permanently [1, 2]. Nowadays, the colorants are usually injected as a suspension into the skin by the use of solid needles, which are actuated by tattoo machines. A fraction of the colorant stays in the dermis as particles causing the color of the tattoo, while an unknown fraction of the injected colorant is removed from the skin via the lymphatic system. As a result, tattoo colorants can be found in the lymph nodes located next to the tattoo [3–7].

From time to time, tattooed individuals ask for tattoo removal stating various reasons, such as change of social status, dissatisfaction with the tattoo, or medical problems. A survey in German-speaking countries revealed a rate of about 5% of tattooed individuals who desire tattoo removal [8]. Several treatment procedures were applied in the past decades to remove tattoo colorants from skin. Surgical excision may remove a tattoo completely but leaves scars and is usually limited to small tattoos. Laser treatment raised hope of selective destruction of tattoo particles in the skin without leaving scars. In 1983, Rox Anderson described such a selective laser application and called it 'precise microsurgery by selective absorption of pulsed radiation' [9]. The so-called selective photothermolysis was born in laser medicine.

Prior to this, T.H. Maiman published the first report on laser emission from a ruby crystal in 1960 [10]. Five years later, one of the first laser treatments of tattoos was published by Goldman and coauthors in the *Journal of Investigative Dermatology* [11]. The report was entitled 'Radiation from a Q-switched ruby laser. Effect of repeated impacts of power output of 10 megawatts on a tattoo of man'. The authors summarized their findings as follows: 'Repeated experiments with Q-switched ruby laser impacts produced transient, edematous blanched areas in a dark tattoo of the forearm but not in the white skin. As yet, the mechanism of this change in the skin reactions of the tattoo to Q-switched ruby lasers is not known, but the differences were probably not due entirely to thermal factors'.

Since then, Q-switched lasers evolved to standard laser technology, which has been applied for many years to destroy tattoo particles in skin, yielding a lightening of the tattoo color [12–15]. Meanwhile, scientific investigations revealed the mechanisms associated with Q-switched lasers and tattoo particles in the skin. In comparison to other targets in laser medicine, like blood vessels, however, the knowledge about the processes is still incomplete with regard to the lightening of tattoo colors in the skin when using short and intense laser pulses.

Tattooed Skin

During the process of tattooing, tattooists perforate the skin surface by using solid needles and thereby place a certain amount of tattoo colorant in the skin. Part of the injected colorant is removed from the skin by the lymphatic system weeks and months after tattooing. Due to limitations of lymphatic transportation, a certain amount of injected colorant stays in the dermis which yields a long-lasting tattoo color of the skin [16, 17]. The major component of tattoo colorants are small solid state particles consisting of pigments with a complex chemistry [2]. Thus, when tattooed people undergo laser treatment of their tattoos, the targets for the laser radiation are such small solid state particles lying randomly distributed in the dermis [18, 19].

Chemistry of Tattoo Colorants

The colorants used for tattoos are frequently called ink, a term that is somewhat misleading because it usually means 'colored water'. However, such a colorant would not stain the skin permanently. The English word 'ink' descends from the old French word *enque* which means dark writing fluid. *Enque* descends from the Latin word *encaustum*, which is what was used by the Roman emperors to sign their documents.

Colorants are chemically classified as either pigments or dyes, whereas the chemical structure of pigment molecules and dye molecules are frequently the same. In contrast to dyes, pigments are practically insoluble in the medium in which they are incorporated. Pigments are inorganic or organic, colored, white, or black materials.

In the past decades, tattooists used inorganic pigments that contained heavy metals such as mercury, chromium, or cadmium. Typical colors were yellow (cadmium sulfide), red (mercury sulfide), or green (chromium oxide). Two important inorganic pigments are still in use: titanium diox-

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Table 1. Selection of colored pigments, which are classified by their chemical nature and identified either by chemical index (C.I.) or pigment shortcut (Px.y)

Trade name	Color	Pigment number
Canary Yellow	yellow	PY.14
I7	reddish yellow	PY.55
Zitronengelb	greenish yellow	PY.74
Golden Luv, Dunkelgelb	reddish yellow	PY.83
Sunset Yellow	reddish yellow	PY.87
Orange, Navel Orange, Melon, I3, P7	yellowish orange	PO.13
Dunkelrot	carmine	PR.5
I8	yellowish red	PR.9
Cardinal Red, Dragon Red, Spanish Red, P8	yellowish red	PR.22
Ruby Red, Red Velvet	red	PR.112
P1	red	PR.170
Burgandy, I5	bluish red	PR.122
I6, Pur Purple, True Purple, P3	bluish violet	PV.23
Permanent Blue, Permanent Blau, Navy Blue, Fezan Lt. Blue, I1	blue	PB.15
Permanentgrün, Waldgrün, Forest Green, I4, Fezan Blue Green, Permanent Green, P2	green	PG.7
Black inks	black	carbon black
White pigments	white	titanium dioxide

Black tattoo inks mainly consist of carbon black. Titanium dioxide is frequently applied to reduce the color strength of pigment solution.

ide for tempering and carbon black for black tattoos. About 60% of tattoos are either completely or partly black [8]. Nowadays, the colored tattoo colorants mainly consist of organic pigments like azo or polycyclic pigments, which are usually taken from the chemical industry [2].

In view of the variety of tattoo pigments on the market, a list of tattoo pigments, such as presented in table 1, is definitely incomplete. Pigments are tiny particles with diameters of a few tenths of nanometers (fig. 1). Tattooists usually purchase ready-to-use suspensions which contain the respective pigment, solvents, emulsifier, anti-foam agents, preservatives, and other substances. The absorption spectra provide evidence of the high color strength of the pure pigments (fig. 2). Therefore, the pigments are often mixed with titanium dioxide to change the color strength.

The Basics of Lasers

The acronym LASER is derived from the most important elements: Light Amplification by Stimulated Emission of Radiation. The process of spontaneous emission of photons describes the emission of radiation from a usual light source. After excitation, atoms or molecules can emit photons at different times, in any direction, and usually with different wavelengths. Therefore, such a radiation is incoherent, divergent, and spectrally broadband.

In contrast, a laser medium consists of identical atoms or molecules which are excited by different means. Such photon emitters are placed in a substrate like a solid state. After excitation, which is called pumping, the atoms or molecules return from the excited to the ground state and thereby

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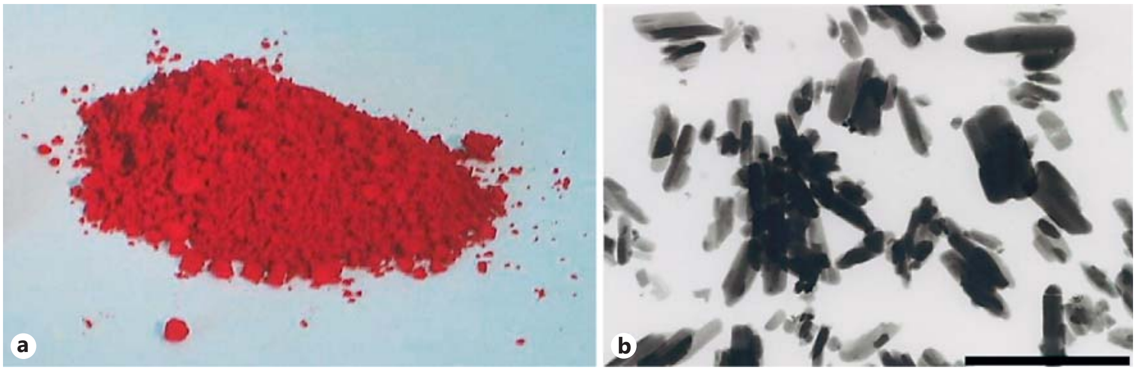

Fig. 1. a Azo pigment PR.22 powder. **b** Electron microscopy image of the disazo-diarylide pigment PY.82 powder. Scale bar, 1 µm.

emit photons with a specific wavelength λ that is determined by the energy difference ΔE of the excited and the ground state, with ΔE = hc/λ.

The combination of such identical transitions and photons with the same wavelength λ in a confined volume called laser medium (e.g. glass rod) allows another process to come to the fore: the stimulated emission of photons, as described and published by Albert Einstein in 1917 (4). A laser medium consists of up to 10^{20} photon-emitting atoms or molecules, which provides an immense number of stimulated photons. For example, a laser pulse (energy: 1 J) in the visible spectrum contains about 10^{19} photons. Thus, the light amplification is actuated by the stimulated emission of radiation, as described by the acronym LASER. The process of light amplification does not change the energy of photons but increases their number exponentially.

To promote the stimulated emission of radiation and hence optimize the process of light amplification, the laser medium is placed in an optical resonator that consists of two or more mirrors. Due to the reflection of laser photons inside the resonator, the number of photons continues to increase until the maximum is reached. The temporal behavior of the laser emission depends on the temporal behavior of the excitation (energy supply) of the atoms or molecules. When the energy supply is limited to a certain time interval by using, e.g. a flash lamp, pulsed excitation take place and the laser emits pulses accordingly, usually in the range of milliseconds to microseconds.

Pulse durations in the nanosecond range (10^{-9} s) can be achieved by switching the quality of the resonator (Q-switch) by using an electro-optical device. The device contains an optically nonlinear crystal, which prevents photons from running back and forth inside the resonator (low quality of the resonator) as long as the crystal is connected to high voltage. When the voltage is switched off within nanoseconds, the switch abruptly becomes transparent, increasing the quality of the resonator and yielding a very intense emission peak.

The mode-locking of the longitudinal modes in the laser resonator may result in ultrashort pulse duration in the picosecond (10^{-12} s) or femtosecond (10^{-15} s) range. Active mode-locking appears when devices such as optoacoustic modulators are placed in the resonator. Also, passive mode-locking is possible by applying a saturable absorber like dye molecules.

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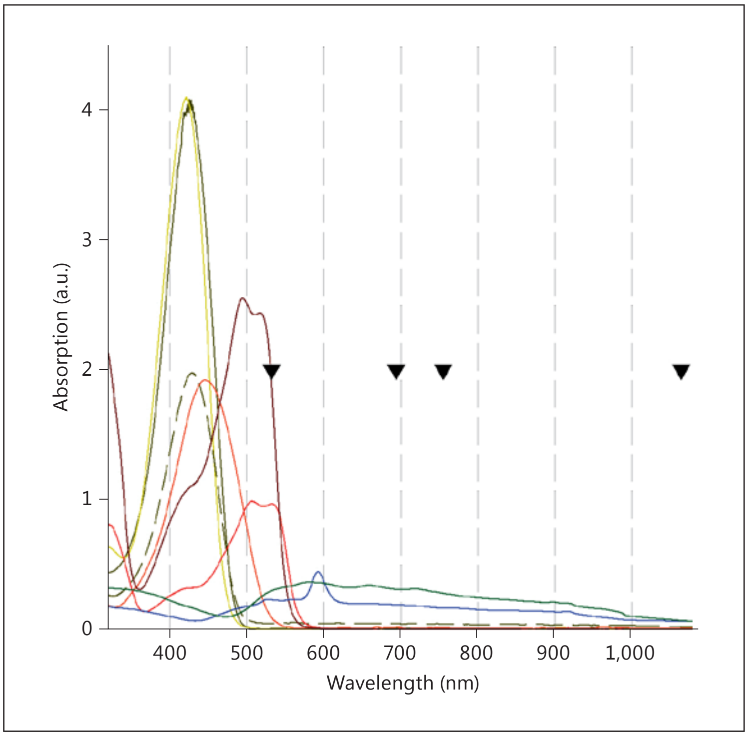

Fig. 2. The image shows the different light absorption (absorption cross-section) of some colored pigments dissolved in appropriate solvents. The four black triangles indicate the wavelengths of typical Q-switched lasers used for tattoo removal: frequency doubled Nd:YAG laser (532 nm), ruby laser (694 nm), alexandrite laser (755 nm), and Nd:YAG laser (1,064 nm).

Laser Tattoo Removal

Background
It remains unclear how many people desire the removal of their tattoos. In a survey, about 5% of people indicated that they plan tattoo removal [8]. The reasons for tattoo removal are either health problems or dissatisfaction with the tattoo. Tattoo colorants in the skin can be removed via surgical excision or laser therapy [20, 21]. When using the CO_2 laser, the tattoo pigments are removed unselectively together with the skin area treated, which leads to unwanted scar formation. Contrarily, when using Q-switched lasers, the ultrashort and intense laser pulses are suitable to destroy only the pigment particles in the skin via selective photothermolysis with harming the surrounding tissue [9]. This principle requires the proper selection of wavelength, light energy, and pulse duration of the laser pulses applied.

Selective Photothermolysis

Wavelength of Laser Pulse
The color of organic pigments is caused by light absorption in the delocalized electrons of a conjugated system in the pigment molecule. The pigment molecules strongly absorb light in a specific wavelength range. The unabsorbed light is reflected to viewers' eyes and hence represents the color of a given pigment. This provides a first indication of which wavelength could be appropriate in case of using laser light for tattoo removal. The light absorption maximum of different pigment molecules may be located anywhere in the UV and visible spectrum (200–700 nm), yielding a huge variety of colored pigments. This, however, hampers the selection of appropriate laser wavelength for tattoo removal, because neither tattooists nor patients usually know the identity of the pigment used for tattooing.

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The lasers used in practice are frequency-doubled Nd:YAG laser (532 nm), ruby laser (694 nm), alexandrite laser (755 nm), and Nd:YAG laser (1,064 nm). Depending on the respective pigment, the laser light does not necessarily match the absorption spectra of tattoo pigments (fig. 2). Moreover, tattoos having similar colors may contain different pigments and therefore exhibit a different absorption behavior. Physicians, who treat tattoos with lasers, therefore apply wavelengths according to the published clinical studies and the clinical experience. However, the numerous different pigments face a small number of laser wavelengths available, which are limited due to technical and commercial reasons.

Energy of Laser Pulse

After tattooing, the pigment particles are exclusively found intracytoplasmatically, lying in membrane-bound structures, identified as secondary lysosomes. This is due to active phagocytosis into dermal cells (macrophages, fibroblasts) [6]. Then, the pigment particles show diameters up to a few micrometers being visible when using normal light microscopy in histology. Now, the goal of tattoo treatment with Q-switched lasers is the fragmentation of these tattoo particles in the skin allowing transportation of the pigment material away from skin. Consequently, the color of the tattoo in the skin should fade.

Thus, the light energy absorbed in the pigment particle must be sufficient for particle fragmentation. When treating tattoos with lasers, the first mechanism is the absorption of light in the pigment and hence the conversion of absorbed light energy to any excitation energy inside the particle (e.g. heat). Assuming a simple heat-up process, the absorbed light energy correlates to the produced heat energy. This energy must be sufficient to heat up the entire pigment particle.

Duration of Laser Pulse

The pulse duration must be in the order of nanoseconds at least for the following two reasons. Firstly, such short pulse duration allows very high laser light intensities that enable rapid heat-up of the tattoo particles, which is necessary for particle fragmentation. Secondly, the heated particle starts to transfer the heat energy to the adjacent tissue by means of thermal conductivity. This effect should be limited to a sufficiently short time to avoid heating of adjacent tissue, reducing the risk of damaging the surrounding tissue.

Laser Tattoo Removal

Penetration of Light into the Skin

After tattooing, the pigment particles are randomly distributed inside the dermis of the skin. When treating tattoos with lasers, the laser light is applied to the skin surface with different spot sizes with diameters of a few millimeters. Then, the light penetrates tissue to reach the pigment particles, which are located in different depths of the dermis.

Propagation of optical radiation through normal media such as air or glass can be described in a straightforward fashion, in particular for monochromatic and collimated laser light. Reflection, refraction, and absorption dominate the propagation. However, when optical radiation penetrates turbid media like the skin, scattering of radiation inside tissue must be considered, which hampers light propagation in a complex manner.

When optical radiation strikes the skin, part of the photons is reflected back due to the different refractive index of air and skin. Photons inside the skin do not propagate straight on but frequently and abruptly change their direction due to collision with skin constituents (scattering). The mode and the extent of scattering depend on the size of the scattering objects and the wavelength of the radiation. The size of the scattering objects ranges from a few nanometers (small cell organelles, cell membranes) to a few micrometers (large cell organelles, cells, collagen) and to hundreds of micrometers (hair follicle, sweat glands).

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The extent of scattering decreases with increasing wavelength. With increasing wavelength, photons are less deflected on their path into skin. Thus, the longer the wavelength, the higher is the penetration depth of radiation in the skin. UVB radiation (around 300 nm) penetrates the skin up to a few tenths of millimeters only, whereas infrared radiation (e.g. Nd:YAG, 1,064 nm) achieves a penetration depth of up to a few millimeters. However, the increase in penetration depth with increasing wavelength reverses for wavelengths longer than about 1,100 nm because radiation is increasingly absorbed by water in the skin. Unfortunately, the scattering changes the beam geometry in the skin and thereby substantially affects the dosimetry. It is complex to determine the number of photons that reach a target (e.g. a vessel) in the skin.

The higher the number of photons reaching the target, the more heat can be produced inside the target. Thus, the deeper tattoo particles are located in skin, the less photons reach the particle and the less is the fragmentation efficacy that limits the therapeutic effect of lasers for tattoo removal.

Mechanisms of Action

The current assumption regarding the mechanisms of action is heat production in the tattoo particle by the absorbed light energy. So far, it is unclear whether and to which extent the heat-up process alone describes the entire process of tattoo particle destruction. Other processes like release of electronic bonds in the particle might also play a role.

Nonetheless, as a response to this laser impact with appropriate parameters, a multitude of mechanisms may occur at the same time. Large aggregates and agglomerates break down into smaller particles. Molecules can break up, resulting in hazardous decomposition products like hydrogen cyanide and aromatic amines [22–24]. Due to fragmentation of the tattoo particles, small pigment particles, unknown decomposition products, and newly generated chemical compounds may then be removed from the skin via the lymphatic system. This mechanism could induce a decrease of the color strength of the pigments responsible for a noticeable lightening of a colored tattoo.

The heating of the particle should be processed in a very short time yielding high temperatures. Then, the extremely hot surface of the particle may cause ultrashort heating-up of a thin shell of surrounding tissue. The following expansion of this water-containing shell may induce a negative pressure and a shock wave near to the surface of the pigment. These shock waves may help to fragment the tattoo particles mechanically. Such an anomalous photoacoustic effect was reported when a suspension of small particles in water was irradiated by a Q-switched Nd:YAG laser (532 nm), where high-temperature reactions and gas expansions induced shock waves [25].

Histological investigation of tattooed skin samples directly after laser impact showed that these shock waves occurred upon treatment with Q-switched lasers. Small vacuoles were exclusively formed in the dermis, which caused a clinically visible whitening of the skin. These vacuoles usually resolve within a few hours after laser impact [19].

Laser Tattoo Removal and Selective Photothermolysis

So far, little is known about the removal of tattoos by using Q-switched lasers. Laser treatment of many skin disorders has been performed under the terms of selective photothermolysis for decades [9]. Shortly again, the energy of laser pulses applied to the skin is selectively absorbed by a target that undergoes thermal damage, while the damage is confined to the target by choosing the appropriate pulse duration. The pulse duration correlates to the thermal relaxation time, which can be estimated when knowing the size of the target. Tattoo pigment particles show a small size requiring a pulse duration of a few nanoseconds.

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However, selective photothermolysis is frequently based on simplified models assuming linear optics and linear thermodynamics. In addition, the density, the composition, as well as the optical and thermal properties of the heated target (e.g. tattoo particle) are considered homogenous. It is additionally assumed that these properties do not change during the heat-up process. This is an appropriate assumption to begin with. However, the assumption may not reflect the real and complex process of tattoo particle fragmentation as a whole.

Laser Tattoo Removal

Linear and Nonlinear Effects
At low light intensities, e.g. nonlaser sources, the properties of materials remain independent of the light intensity. Light waves can pass through materials or be reflected from boundaries and interfaces without interacting with each other.

However, laser sources can provide sufficiently high light intensities to modify the optical properties of materials. Then, light waves can interact with each other causing nonlinear optical effects. This interaction of light waves can result in the generation of optical fields at new frequencies (wavelengths) including optical harmonics of incident radiation or frequency mixing (e.g. frequency-doubling effects, two-photon absorption). Nonlinear optics is the area where the response of the material system to the applied electromagnetic field is nonlinear in the amplitude of this field.

With regard to the short laser pulses (10^{-9}–10^{-12} s) used in tattoo removal, the intensity of such laser pulses should be high enough (up to hundreds of MW/cm^2) to allow nonlinear processes such as frequency doubling and two-photon absorption. Numerous dye molecules show light absorption at half of the wavelength that is applied for excitation [26, 27].

Regarding tattoo removal, so far, the assumptions of the selective photothermolysis are based on the linear absorption of laser photons in a pigment molecule. The extent of photon absorption (A) is described by the Beer-Lambert law: $A = 1 - exp\,(-\mu_a d)$, where μ_a is the linear absorption coefficient and d the thickness of the light absorbing sample. Due to the high intensities of Q-switched lasers, the simultaneous absorption of two photons might occur. That is, two photons of a ruby laser (694 nm) can absorb like one photon at 347 nm. At present, however, it has not been investigated whether and to which extent such a nonlinear process of two-photon absorption might contribute to the effect of light absorption in tattoo pigments.

Complex Properties of Tattoo Particles
As mentioned above, tattoo pigments are usually industrial pigments consisting of inorganic or organic molecules. Inorganic pigments such as carbon black (black tattoos) show numerous and complex morphologies. Commonly, the carbon black particles can be divided into four different types of morphologies, spheroidal, ellipsoidal, linear, or branched, which are caused by different production and finishing processes [28]. Organic pigments such as azo or polycyclic pigments (colored tattoos) are closer to the amorphous state than to the idealized crystal. However, pigments are frequently treated with different methods to enhance crystallinity [29, 30]. When used for tattoos, it is usually unknown which pigment particles in which crystallinity state were applied. In addition, the small particles may agglomerate after tattooing into the skin by different mechanisms of phagocytosis. Thus, the particles of black and colored tattoos in the skin may show various particle structures and sizes that clearly influence any interaction with light. In particular when heated up by laser radiation, such variations may lead to different fragmentation mechanisms and efficacies.

Ho et al. [31] investigated whether the mechanism of the breakup process can be identified via computer simulations and proposed a treatment strategy that can potentially minimize the collateral damage to the surrounding tissues. The au-

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thors calculated the magnitude of the tensile stress generated inside graphite tattoo particles as functions of laser pulse length and particle size. The authors concluded that the breakup of tattoo particles is photoacoustic and the optimal pulse length is approximately 10–100 ps to minimize the laser fluence and the collateral damage. However, these calculations are based on homogenous graphite particle keeping the respective values for density, specific heat, and thermal conductivity constant.

In contrast to that, it has recently been shown that the values for density and thermal conductivity of carbon black decrease and the value of specific heat increases with increasing temperature [32]. That is, these values of carbon black might also change when heated up by intense laser pulses during tattoo removal. As mentioned above, tattooists usually apply black pigments that contain no graphite particles but carbon black with various morphologies and hence various optical and thermal properties. In addition, the interaction of intense laser pulses with carbon black and carbon-based material may produce nonlinear effects. Lim et al. [33] have recently reported that under intense laser excitation, thin films and suspensions of graphite and its nanostructure, including carbon black, nanotubes, few-layer graphenes and graphene oxides, exhibit multi-photon absorption and induced transparency due to saturable absorption. When using nanosecond pulses, the authors reported the detection of microbubbles and microplasmas that causes nonlinear light scattering.

Another computer-assisted model (finite element method) was applied to simulate the fragmentation of tattoo pigment particles. Interestingly, the authors concluded that an increased spot diameter and beam fluence improve the lightening response of tattoos. This correlated to the authors' observations from the simulation. Conversely, variations in the pulse width are shown to have little influence on the fragmentation response [34].

The situation should be even more complex when laser light interacts with the various morphologies of colored pigment particles based on azo or polycyclic compounds. On one hand such modeling work is important, but the results might not reflect the reality when a laser pulse hits a tattoo particle of unknown origin and morphology in skin.

An animal study was published in 1999 showing better clearance of black tattoos when using pulse duration of 500 ps (titanium:sapphire laser, 795 nm) as compared to 50 nanoseconds (alexandrite laser, 752 nm) [35]. The question remains whether and to which extent the different wavelengths possibly affected the results. In 1998, a preliminary clinical study showed that picosecond laser pulses yielded better results than nanosecond pulses [36]. The authors used a Nd:YAG laser with a pulse duration of 35 ps, which was considerably shorter than that used in the few recent studies (350–900 ps) [37–40].

Conclusion

At first view, the treatment of tattoos appears to be a rather simple process that has been well understood in the past decades. However, on closer inspection, the fragmentation of complex morphological structures such as tattoo pigment particles, consisting of various molecules, is a rather complex process that involves known and many unknown facts. This includes nonlinear optical effects as well as unknown thermal properties of various pigment particles with different morphologies. This might hamper the application of the usual theory of selective photothermolysis or lead to the conclusion that this theory might not be appropriate in its current form. In view of all these unknown facts, the simple change of a single laser parameter such as shortening of the pulse duration – as frequently recommended – might be a premature and oversimplified recommendation.

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