


## Article

# Laser Cleaning as Novel Approach to Preservation of Historical Books and Documents on a Paper Basis

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**Abstract:** The purpose of this work is the study of laser cleaning of historical paper. The effect of laser exposure of the paper reflectance, fracture resistance and acidity was investigated. The paper surface roughness before and after laser treatment was analyzed by optical and scanning electron microscopy. It was shown that use of multi-pulse micromachining in combination with high-speed scanning of laser beams provides high safety for paper cleaning. The optimal parameters of laser radiation for effective and safe cleaning are a power density of about  $2 \times 10^5$  W/cm<sup>2</sup> at a wavelength of 1.06 μm, pulse repetition rate is 20 kHz; and a beam scanning speed of 200 mm/s–500 mm/s. The selective laser cleaning method for books and documents was proposed. Selective cleaning is achieved by means of high-precision control of the trajectory of movement of laser beams.

**Keywords:** laser cleaning; paper; books; documents; contaminants; cultural heritage; restoration



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## 1. Introduction

The use of laser technology in the field of preservation of cultural heritage (CH) is a new but rapidly developing scientific and technical direction. Most often, lasers are used to determine the chemical composition, structural diagnostics, and documentation of works of art. These applications are based on the use of laser spectroscopy, interferometry, holography, and 3D scanning [1–3]. The restoration exploits laser cleaning technology, which allows solving the problems of removing natural layers and anthropogenic pollution from the surface of CH objects. However, despite the fact that lasers were first used in restoration almost 50 years ago, to date, only the technology of cleaning the stone has been developed. Laser restoration of other, primarily organic materials is still in the process of conducting exploratory research [3–19]. This is due to the fact that in the process of laser treatment, damage to their surface may occur, which is unacceptable according to the canons of restoration.

This article is devoted to the development of technology of cleaning of books and documents in the paper base. Cleaning paper sheets of dirt is a necessary and very time-consuming operation in the restoration of books and documents. Wax stains, soot and dust pollution, biological lesions, the so-called “fly ash”, etc., are subject to removal. However, traditional methods of restoration cleaning do not always give satisfactory results. Thus, mechanical cleaning (based on the use of an eraser, rubber crumb or abrasive sandpaper with fine grain) violates the integrity of the surface structure of the paper, and reduces the thickness of the sheets, which ultimately negatively affects their mechanical strength. In addition, some of the removed dirt remains in the upper layer of the damaged paper surface in the form of dust. It should also be noted that there are often cases when the physical condition of books, especially the dilapidation of the lower corners of the book block sheets, in principle does not allow the use of mechanical cleaning. As an alternative to mechanical cleaning, water washing and chemical treatment with organic solvents and

enzymes are sometimes used. Although such methods may be effective, they cannot be used to clean handwritten documents, as well as books with unstable printing ink.

For these reasons, the search for alternative methods of restoration of books and documents is currently relevant. One possible approach to solving this problem may be the use of laser cleaning. The first experimental works on the use of lasers for this purpose was carried out in the late 1990s [4,5], but is still being carried out by a few scientific groups in Europe [6–18]. These works show the possibility of using a laser for cleaning, but the method is not yet widely used. For this, it is necessary to develop appropriate technologies and specialized laser systems suitable for a wide range of contaminants.

Therefore, the purpose of this work is to develop a method for laser cleaning of documents and books with different types of paper.

## 2. Experimental

Model samples of paper that differed by chemical composition as well as fragments of historical documents and books of XVI-XIX centuries were used in experiments (Table 1).

**Table 1.** List of samples.

Label	Paper Composition	Dating
Model samples		
B-2	100% sulphate cellulose	XX century
B-5	100% sulphate cellulose, kaolin filler, ash content–5.2%	XX century
B-6	100% Linen half-mass	XX century
B-19	100% sulphate cellulose, kaolin filler, ash content–4.6%	XX century
B-25	100% cotton half-mass, kaolin filler, ash content–9.3%	XX century
Pag paper	100% flax fiber, sizing-starch	XX century
Historical paper		
Russian typographic book		beginning of the XIX century
Russian handwritten book		XVII century
Arabic manuscript		Late of 19th century
Arabic manuscript		Medieval

A pulsed ytterbium fiber laser generating at the wavelength of 1.06  $\mu\text{m}$  was used for paper cleaning.

For all paper samples, reflection spectra were measured, whiteness of the paper, its acidity (pH) and fracture resistance were determined. Measurements were taken before and after laser cleaning. Reflection spectra were measured on a LAMBDA 900 spectrophotometer (manufactured by PerkinElmer LLC, Santa Clara, CA, USA) in the wavelength range of 300–2500 nm with a scanning step of 1 nm. The whiteness of the paper was measured on an FMSH-56 M ball photometer. An electron scanning microscope JSM-35 (JEOL Ltd., Tokyo, Japan) was used for surface study.

## 3. Results and Discussion

### 3.1. Choosing a Laser for Paper Cleaning

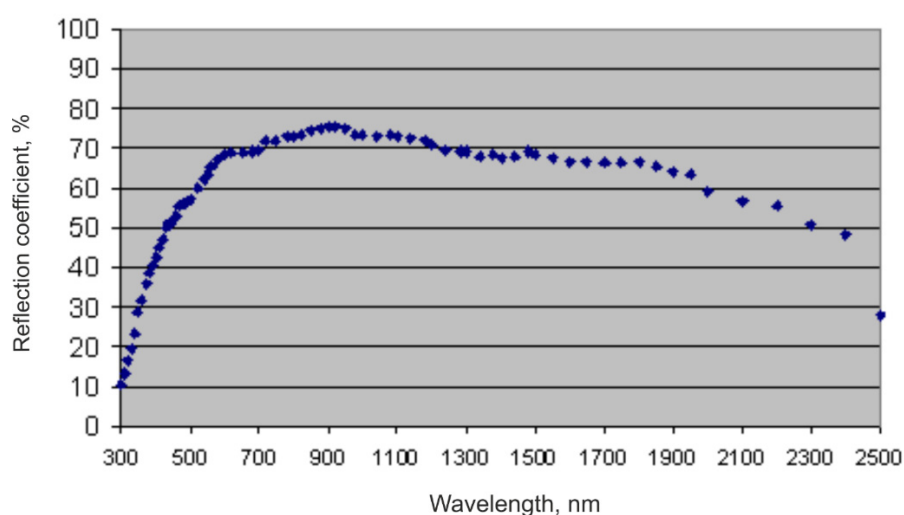
Laser cleaning of paper is based on the use of physical effects, which is called photoablation. Photoablation is the contaminant particles' ejection by photons as a result of their absorption by dirt material to be removed in the process of cleaning. The concept of the use of laser radiation for selective removal from the surface of an optically absorbing

different substance, for instance, a secondary encrustation, was first demonstrated in 1965 by Arthur Schawlow, one of the laser inventors, who evaporated selectively absorbing black pigments of printing ink from a strongly reflecting white sheet of paper [20]. Later, in 1972 John Asmus first demonstrated the laser cleaning of Cultural Heritage objects [21]. Nowadays, basic physical principles of laser cleaning are well known and described in the scientific literature (see, for instance [2,22,23]).

The first step towards the development of laser technology for paper restoration is the choice of such laser radiation parameters that allow for high efficiency in cleaning books and documents. However, laser output parameters must be so they do not cause any negative consequences (including carbonization as well as changes in color, acidity, porosity and other physical and chemical properties of the paper). When choosing the type of laser and its output characteristics, it is necessary to keep in mind that paper is a very fragile, easily damaged material. It is a porous-capillary body in the form of a “carcass” formed by cellulose fibers connected to each other by chemical hydrogen bonds. Under the influence of heat, mechanical stresses and chemical reagents, the paper fibers are easy to damage.

It is known from the scientific literature that various types of lasers were used for paper cleaning, including excimer lasers (in particular, the XeCl laser with a radiation wavelength of 308 nm and the KrF laser with wavelength of 248 nm), as well as a solid-state Nd:YAG laser operating either at the main wavelength or at the wavelengths of the second, third and fourth harmonics (with wavelength of 1064 nm, 532 nm, 355 nm and 266 nm, respectively) [2,4–13]. In all cases, pulsed laser radiation was used. In particular, when working with a solid-state Nd:YAG laser, a Q-switched generation regime with a pulse duration of 10 ns–30 ns is usually used. As for the pulse repetition rate, it is usually in the range 1 Hz–100 Hz (maximum—1 kHz).

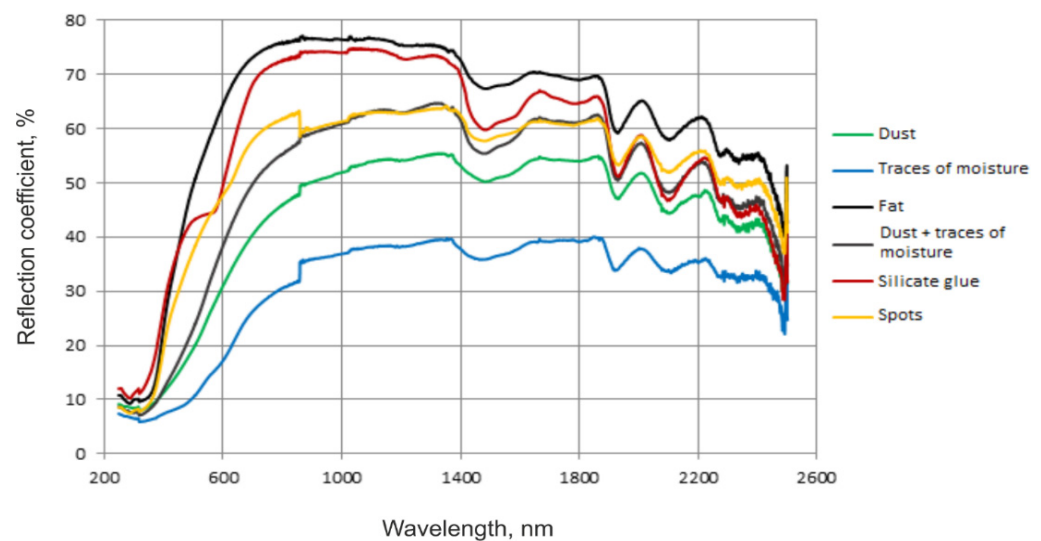
In order to choose the optimal laser wavelength for efficient and safe cleaning of books and documents, the spectral characteristics of the paper measured at the initial stage of our work. Measurements of the reflectance spectra give information on the spectral bandwidth where paper has maximum absorption. Since paper is a scattering medium, we used an integrating sphere. Results of these measurements are shown in Figure 1.



**Figure 1.** Reflection coefficient of paper versus laser radiation wavelength (for sheet of a Russian typographic book of the beginning of the XIX century made of wood pulp).

Then, we measured the reflectance of paper sheets of historical books and documents. These measurements were conducted using a LAMBDA 900 spectrophotometer (manufacturer—PerkinElmer LLC, Santa Clara, CA, USA). A diaphragm with a diameter of 10 mm was used to select the measurement area. The area included typical paper con-

taminations: traces of silicate glue and rust, dust and dirt stains, grease stains, moisture traces, etc. Obtained spectra are presented in Figure 2.



**Figure 2.** Reflection coefficient of paper with contaminants versus laser radiation wavelength.

From Figures 1 and 2, one can see that all the recorded spectra are identical to each other, and the differences are only in the absolute values of the reflection coefficient, which depend on the specific type of surface contamination. The most important finding is that the paper has the lowest reflectance values in the UV region of the spectrum. Consequently, the use of short-wave lasers for cleaning books and documents can be dangerous for their state of preservation due to the significant absorption of radiation energy by the paper. Indeed, the authors of some works (see, for example, [5]) reported damage to paper during its cleaning by an excimer laser with a radiation wavelength of 308 nm. From the graph in Figure 1 it is also clear that the maximum value of the reflection coefficient of the paper is in the range of 900 nm–1100 nm. Therefore, it is advisable to use lasers with a wavelength of radiation in this spectral region, and the best “candidates” for this role are solid-state neodymium lasers, as well as fiber ytterbium lasers with wavelength of about 1 micrometer. It is obvious that when using such lasers, the effect of their radiation on the surface of the paper cleaned of impurities will be insignificant, which ensures its safety during laser processing.

As for paper with contaminants on the surface from Figure 2, one can see that in the wavelength range of 900 nm–1100 nm, samples with dust contamination and under the lobes have lower reflection coefficients (about 35–60%) than samples with grease, silicate glue and stains of unknown origin (about 60–70%). In this case, the maximum value of the reflection coefficient (about 80%) has a sample with fat spots. It is clear that using lasers with a wavelength of about 1 micrometer with enough one can remove contaminants if the intensity of radiation will be enough high.

Another important parameter of laser radiation in the treatment of materials is the pulse repetition rate. Since at a fixed level of the average laser output power, an increase in the frequency leads to a decrease in the energy density of a single pulse, it is obvious that this allows for a higher “delicacy” of paper processing by reducing the thermal energy generated as a result of its absorption of laser radiation. It is for this reason that, unlike other scientific groups involved in study of laser paper cleaning, the authors of this work decided to check the possibility of using very high values of the repetition rate of laser pulses (at the level of tens of kilohertz). With all the above parameters, the effect of the laser on the paper becomes similar to the so-called multi-pulse laser micro-processing, which is widely used in industrial processing of materials. For the first time, the idea of using this

approach in the restoration of paper was expressed by one of the authors of this article in 2007 [24].

However, it should be noted that to ensure maximum safety of paper processing, it is important not only to reduce the pulse energy density, but also to eliminate the cumulative effect of the thermal effect of laser radiation. Local temperature increase, which can occur during prolonged exposure to the same area of the surface, may cause negative effects—color change and even charring of the paper. To avoid this, it is necessary to ensure a very fast and uniform movement of the laser beam on the surface of the paper sheet. This problem can be solved by using high-speed laser beam scanning systems. For this purpose, you can use, for example, 2-coordinate scanners based on galvanometric mirrors, which are used in lasers for marking and engraving. In this case, each time the laser beam passes over the surface of the paper sheet, very thin layers of dirt will be removed. If the cleaning level is insufficient, the processing can be continued by re-scanning the entire sheet or its individual sections.

Based on the above considerations, in the experiments on paper cleaning, a pulsed solid-state fiber ytterbium laser was used. It has the following output characteristics: radiation wavelength—1.06  $\mu\text{m}$ ; maximum average power—10 W; pulse duration—10 ns; maximum pulse repetition rate—100 kHz.

### 3.2. Characterization of Paper before and after Laser Cleaning

The scheme of the experimental setup is shown in Figure 3a. The laser beam was focused onto the surface of paper and the sample was scanned (the spot diameter of focused laser beam was about 50  $\mu\text{m}$ ). The general view of the experimental setup in the process of laser cleaning of paper is shown in Figure 3b.

Laser processing parameters have been determined to ensure the safe and efficient removal of contaminants from the paper surface. Safe cleaning does mean the absence of such negative side effects as charring (carbonization), discoloration and roughness change of the paper surface. The maximum removal efficiency without paper damage was obtained at an average power of 4 W, while the pulse repetition rate was 20 kHz. Thus, the power density in the spot was about  $2 \times 10^5 \text{ W/cm}^2$ . The beam scanning speed was varied in the range of 200 mm/s–500 mm/s, and the laser processing area was  $4 \times 6 \text{ cm}^2$ .

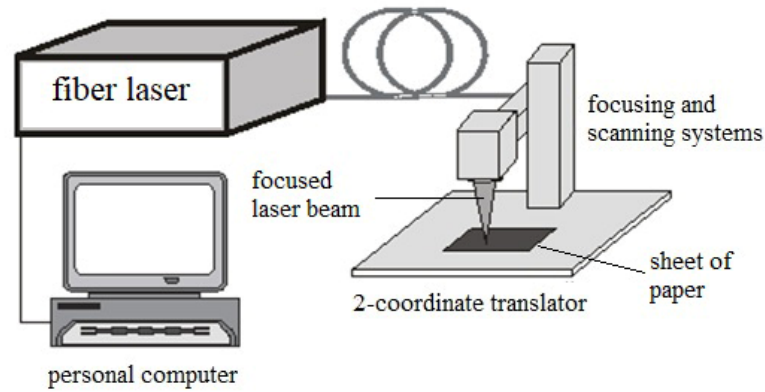
At the first stage, there was a study of the effect of laser processing on the physical, mechanical and chemical properties of paper and its wear resistance. The following parameters of paper properties were determined: fracture strength, pH value of aqueous extraction and whiteness. In the field of book restoration, exactly these parameters characterize the safety of any restoration treatment. Accelerated thermal aging of the studied samples (105 °C, 72 h) was carried out to study the effect of laser processing on the durability of paper. To reduce the thermal effect of the laser on the paper, high-speed scanning in two mutually perpendicular directions was used. The paper samples were lasered on both sides of the sheet. The values of the index of resistance to destruction of paper samples of various compositions after laser processing of its surface (on both sides) are presented in Table 2.

The maximum effect of laser cleaning on the mechanical strength of paper was in samples B-2 (from 100% sulfate pulp) and B-6 (from 100% half-mass of flax). For these samples, the average value of the number of double kinks (ndk) decreased by 12–20%, and the decrease in fracture resistance occurs only in the longitudinal direction. For other types of paper (B-5, B-19, B-25 and Pag paper), the fracture resistance index does not decrease after laser processing.

A slight increase in the ndk in the transverse direction was also observed. The nature of fracture resistance in mutually perpendicular directions is different, in the longitudinal direction it is determined by the strength and elasticity of the fiber, and in the transverse direction it depends on the strength of the interfiber bonds. Heating to a temperature of 220 °C leads to dehydration, which reduces the elasticity of the paper and leads to a decrease in the tear resistance of the paper. This is proved by a small weight loss associated with the dehydration of cellulose fiber, which was observed during DSC analysis in [25]. At



the same time, the removal of hygroscopic water should lead to strengthening of hydrogen bonds, which make the main contribution to the energy of interfiber bonds in paper. Laser exposure leads to heating of the paper and similar processes can be observed in this case.



(a)



(b)

**Figure 3.** Experimental setup for laser treatment of paper: (a)—diagram of experimental setup; (b)—general view.

**Table 2.** The effect of laser processing on the physical and mechanical properties of paper (index-fracture resistance).

Paper Composition		Fracture Resistance (Number of Permissible Double Kinks)					
		Control			After Laser Treatment		
		Longitudinal Direction	Transverse Direction	Average Value	Longitudinal Direction	Transverse Direction	Average Value
B-2	100% sulphate cellulose	208	160	184	150	174	162
B-5	100% sulphate cellulose, kaolin filler, ash content–5.2%	14	31	22	13	36	24

Table 2. Cont.

Paper Composition		Fracture Resistance (Number of Permissible Double Kinks)					
		Control			After Laser Treatment		
		Longitudinal Direction	Transverse Direction	Average Value	Longitudinal Direction	Transverse Direction	Average Value
B-6	100% Linen half-mass	630	274	452	469	284	376
B-19	100% sulphate cellulose, kaolin filler, ash content-4.6%	134	92	113	170	97	134
B-25	100% cotton half-mass, kaolin filler, ash content-9.3%	10	7	8	10	8	9
Pag paper	100% flax fiber, sizing-starch	9	9	9	9	9	9

The whiteness and acidity index values of paper samples before and after laser treatment (Table 3) show that paper properties (related to whiteness and pH) do not change.

Table 3. The effect of laser treatment on the whiteness of the paper and the value of the acidity index (pH).

Cipher of Sample	Control		After laser Treatment	
	Whiteness, %	pH	Whiteness, %	pH
B-5	73.0	5.1	72.0	4.8
B-19	72.0	7.2	72.0	7.0
B-25	83.0	7.3	82.0	7.6
Rag paper	58.0	4.9	58.0	4.9

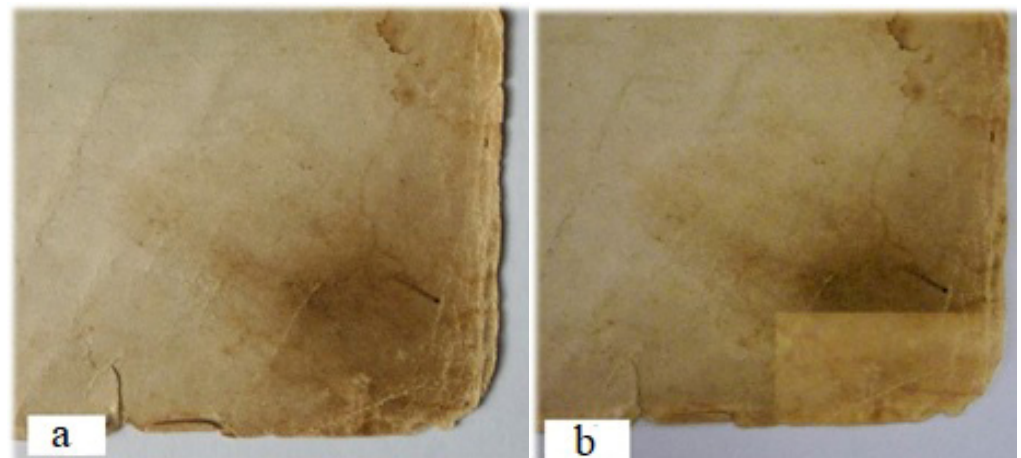
Analyzing the data of accelerated thermal aging of the samples, it can be concluded that laser processing did not affect the durability of the paper. Changes in fracture resistance, brightness, and pH values of the laser-treated and control samples were identical (see Table 4).

Table 4. Changes in the physical, mechanical, optical, and chemical properties of control paper samples and laser-treated paper samples after thermal aging (100 °C, 72 h).

Cipher of Sample	Thermal Aging									
	Control					After Laser Treatment				
	Fracture Resistance (n.p.k.)			White, %	pH	Fracture Resistance (n.p.k.)			White, %	pH
	Longitudinal Direction.	Transverse Direction	Average Value			Longitudinal Direction.	Transverse Direction	Average Value		
B-5	9	20	14	69.0	4.9	7	22	14	69.0	4.8
B-19	168	116	142	70.0	6.9	192	113	152	69.0	7.0
B-25	10	8	9	82.0	7.6	10	7	8	81.0	7.4
Rag paper	4	5	4	55.0	4.5	4	6	5	55.0	4.5

Visual inspection of the cleaning areas (using an optical stereomicroscope) showed that as a result of a single laser treatment, a significant part of the contamination is removed without disturbing the microrelief of the paper surface and its color characteristics. After completing a series of test tests on model images that demonstrated the safety of the

selected laser and its output characteristics on the properties of paper, experiments were conducted on laser cleaning of original historical documents—a sheet of a handwritten book of the XVII century, as well as a page of a Russian typographic book of the early XIX century with typical types of contamination. As a result of laser treatment, the main surface contamination was removed in both cases. The condition of the paper before and after laser cleaning, as well as when working with model samples, was monitored using an optical microscope. Additionally, an scanning electronic microscope (SEM) JSM-35 was also used. Optical microscopic and SEM images shown in Figures 4–6 allow us to conclude that laser processing does not lead to a violation of the surface morphology and structure of the cellulose fibers of the paper. In addition, there are no changes in its color. It was confirmed by means of colorimetric measurements including the original measuring technique described in one of our recent publications [17].

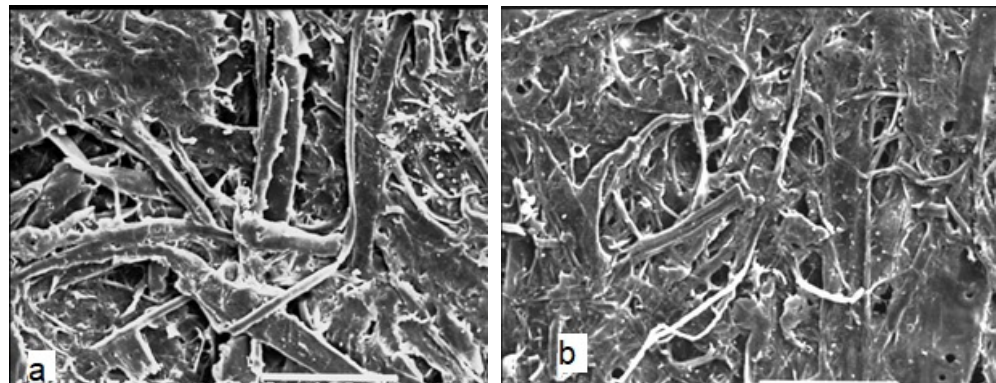


**Figure 4.** Flyleaf of a Russian typographic book (the beginning of the XIX century) with characteristic surface contamination: (a)—before laser cleaning; (b)—after laser treatment (light yellow rectangle in the lower right part of the sheet).



**Figure 5.** Page of a Russian handwritten book of the XVII century with characteristic surface contamination: (a)—the initial state; (b)—the lower right corner marked with a pencil—the experimental area for clearing with a laser; (c)—the same section of the page after laser processing.





**Figure 6.** Images of the surface of a sheet of handwritten paper book obtained on an electronic scanning microscope: (a)—the initial state, (b)—after cleaning with a laser.

It should be noted that Fourier Transform Infrared spectroscopy makes it possible to control the chemical changes of paper after laser treatment at the molecular level, but in our work such measurements have not been conducted since from scientific literature it is known (see, for instance [11]) that with the right choice of laser exposure, such a negative side effect can be prevented.

Therefore, our experiments confirmed high efficiency and safety of laser cleaning of paper.

### 3.3. Selective Laser Cleaning

Consider the security of such processing for the entire page, which may include text and pictures. Since the absorption of carcasses and ink differs from the absorption of paper, it is necessary to study the effect of laser cleaning on such objects.

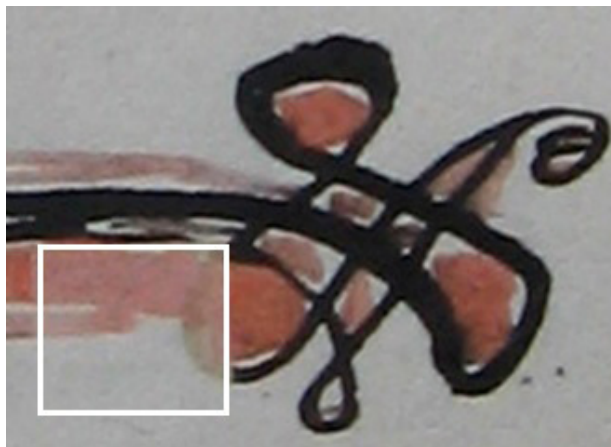
The preliminary experiments were carried out on a sheet of modern Russian strong paper “type 1000” on which typewritten text was printed in black ink. Laser cleaning removed the text from the paper, along with the dirt.

Then, a fragment of a sheet of a medieval Arabic handwritten book with text written in multi-colored ink (red and black) was cleaned. The text in red ink has not changed. Removal of surface dirt from the paper also led to partial removal of black ink (Figure 7).



**Figure 7.** Images of a sheet of medieval Arabic manuscript: (a)—a general view, (b)—a fragment (a light rectangle in the middle at the top) cleaned by a laser.

From a later Arabic manuscript (19th century), both black and red ink were partially removed by laser treatment (see Figure 8). That is, if a pencil, iron gall and soot ink were used to write books, then special protection measures must be taken during their laser cleaning. Aniline ink does not require any special protection procedures.



**Figure 8.** A fragment of an Arabic manuscript page after laser processing (the white square shows the area processed by the laser).

To ensure the safety of paper documents, it is necessary to exclude the effect of laser radiation on text and drawings applied by handwriting and printing. Thus, for the restoration of books and documents, it is necessary to use selective laser cleaning.

It should be noted that the idea of selective purification has already been described in [26]. Laser cleaning the BAM (Federal Institute for Materials Research and Testing, Berlin, Germany) prototype (designed by W. Kautek) was based on the use of a Nd:YAG laser (generating at 1064 nm or 532 nm) with a maximum repetition rate of up to 1 kHz and an energy density of  $\sim 10 \text{ J/cm}^2$ . Increasing the repetition rate to 35 kHz while maintaining a power density of  $10 \text{ J/cm}^2$  allowed us to reduce the energy of a single pulse, which is safer for paper heating.

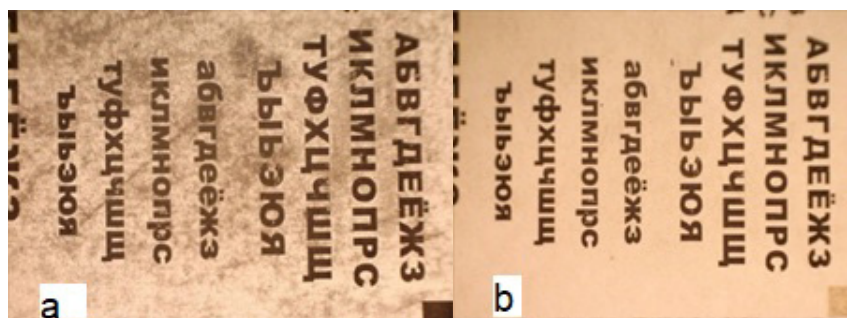
The method of selective cleaning by zones was developed on model samples, which were sheets of modern office paper with laser-printed text. Coal dust was applied to the surface of the paper to simulate contamination. To remove from the processing area areas containing text, their inverted vector image (mask image) was sent to the scanning system operation control program. The mask image was created by cropping the original sheet image with text and converting it using CorelDRAW X7 (designed by Corel Inc., Ottawa, ON, Canada).

With laser cleaning, it is necessary to ensure a very precise alignment of the processed area of the paper sheet with the scanning field in order to avoid the risk of damaging text and graphic information. For this, a combination of special fiducial marks (previously applied to the processed area of paper) and the so-called overall frame was made. Reference marks are applied to the treated surface to align it with the working area of the laser.

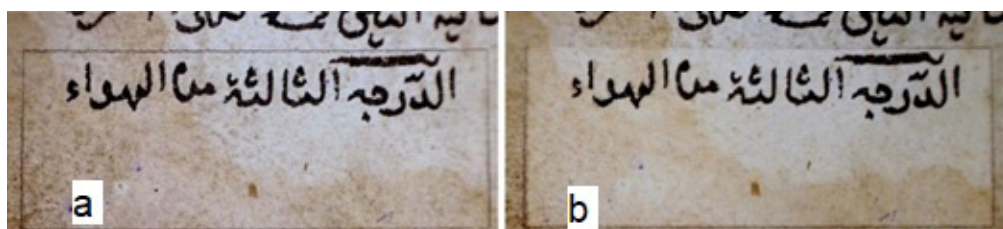
During the experiments, an artificially polluted sheet of paper was treated with a focused laser beam. The results of laser processing of the model sample sheet are shown in Figure 9. As one can see from these photos (on the left—a sheet of paper in its original state, on the right—after laser cleaning), the contamination was successfully removed, but the typographic text remained unchanged. This confirms the effectiveness of the proposed technical solution.

At the final stage of the work, experiments were carried out on the selective cleaning of authentic historical documents. In particular, a page of an early 20th-century Arabic book was used for this purpose. A small area of the text sheet was selected for cleaning. After completing all the preparatory technological stages (it is described in details in [17]), it was

necessary to accurately position the processing area selected on the book sheet relative to the laser working area. The processing area was a rectangle drawn in pencil (see Figure 10).



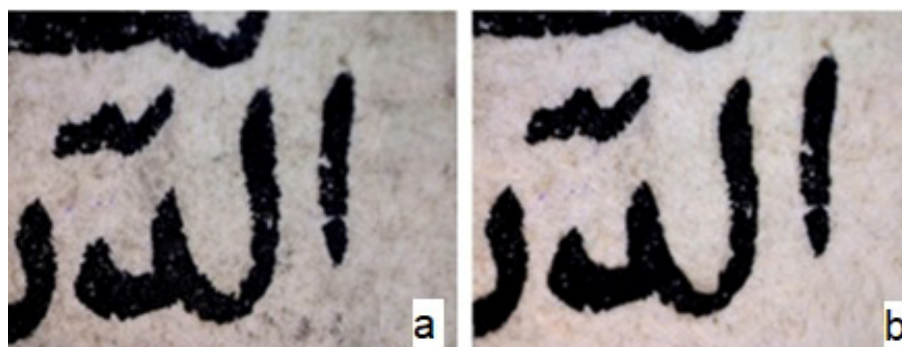
**Figure 9.** A model sample of paper with simulation of contaminants on the surface: (a)—before cleaning, (b)—after laser treatment.



**Figure 10.** A fragment of antique Arabic manuscript: (a)—before cleaning, (b)—after laser treatment.

As can be seen from Figure 10, during the experiment it was possible to successfully perform selective cleaning—the book sheet was cleared of surface contamination, but the original text was saved in the laser processing zone.

The analysis of the cleaning was evaluated visually. Figure 11 shows a photo of a small fragment of a book sheet with typographic text before and after cleaning. It clearly shows that the integrity and readability of the letters of the typographic text is well preserved during the laser processing.



**Figure 11.** A small fragment of antique Arabic manuscript: (a)—before cleaning, (b)—after laser treatment.

#### 4. Conclusions

Based on the experimental results obtained, the following main conclusions can be drawn:

1. Laser cleaning of paper using a pulsed IR laser with wavelength of about 1  $\mu\text{m}$  provides high efficiency and safety of divestment of historical books and documents.
2. The use of multi-pulse micromachining in combination with high-speed scanning of laser beam provides high safety for paper cleaning. The optimal parameters of



laser cleaning of paper are: pulse duration—about 10 ns; average power—of about 4 W (power density of about  $2 \times 10^5$  W/cm<sup>2</sup>); pulse repetition rate—about 20 kHz; scanning speed—200 mm/s–500 mm/s.

3. To ensure the safety of books and documents during laser cleaning, it is necessary to exclude the impact of laser radiation on the text and drawings to avoid their damage. It can be achieved using a high-precision laser beam scanning system that allows you to display typographic text and graphic information from the laser processing area.

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**Conflicts of Interest:** The authors declare no conflict of interest.

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