Romanian Reports in Physics, Vol. 62, No. 3, P. 678–686, 2010

Dedicated to the 50th LASER Anniversary
(LASERFEST-50)

LASER BEAM IN THE SERVICE OF PAINTINGS RESTORATION

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(Received June 18, 2010)

Abstract. Laser cleaning as a particular application of laser ablation became in the last years a subject of interest due to the applications in cultural heritage restoration and conservation. For a safe application of laser cleaning in paintings cleaning and restoration analyses of laser radiation interaction with painting materials was realised and a laser cleaning system was developed. The work was realized in the frame of FP6 European Research Programme.

Key words: UV laser, laser cleaning, paintings, laser ablation, profilometry, microscopy, ablation threshold.

1. INTRODUCTION

Laser radiation interaction with painting layers was studied in an international consortium formed by research institutes and small and medium enterprises specialized in old paintings restauration as final users. The idea of this research sustained by the European Community's Sixth Framework Program was that based on the unique characteristics of the laser beam to offer a well controlled working tool for a very special and delicate activity, restoration of old paintings. The research was organized in two directions: to analyze the laser radiation interaction with painting layers and to develop a laser cleaning system sustained by diagnostic and control tools. Laser removal of thin layers of material is already demonstrated and it was recognized as a method of surface cleaning applied in industrial surface treatment (e.g. semiconductor cleaning in microelectronics, dye cleaning in plastic pressure casting, paint striping in the plastic maintenance and other [1-4]), in cleaning of buildings or stone and marble objects but also for cleaning of painted objects. The method is based on removal of a thin layer of material from a surface under the action of the laser beam, process which is generally known under the name "laser ablation". The main advantages of the laser ablation process are:

- no direct mechanical contact with surface to be processed;
- possibility to focus a great intensity in a precise localized point on the surface and no effects on the adjacent zones are present;
- localized action in depth of the material if the proper intensity and number of pulses is used, depending on the thickness of the covering material to be removed;
- it can be used for in-situ cleaning, without removal the subject object from the technologic flux;
- cleaning process is generally very fast;
- no foreign atoms are introduced to the surface as with other cleaning techniques;
- removal rate can be easily controlled by changing the beam fluence or pulse repetition rate.

The advatages of the laser action for surface cleaning are demonstrated, but must be observed that laser ablation is an irreversible effect which can be induced on optically absorbing materials or in their close proximity.

As the method was successfully demonstrated a deep analyse of the limiting parameters and of the adverse effects is needed in connection with the development of professional systems designed for a controlled treatment of painted surfaces.

Laser cleaning is a particular case of laser ablation where a specific material layer or substrate is uncovered through the removal of undesired layers or single particles. The first reported observation of the laser ablation related to cleaning phenomena dates back to the origins of laser technology [5]. Significant advancements on the understanding of the physical mechanisms through systematic phenomenological studies and the diagnostics of the laser-material interaction were achieved starting from beginning eighties [6-8]. The main results were provided by researches related with medical surgery and microelectronic industry, which resulted of fundamental importance also in other fields of application including the present one.

2. CONTROLLED REMOVAL OF OVERPAINTING AND PAINTING LAYERS UNDER THE ACTION OF UV LASER RADIATION

In this study we have analyzed selective removal of painting layers but also insoluble resins (e.g. highly oxidized varnishes, burnt varnishes and synthetic resins), oil-based over paints; deposits (e.g. dirt, soot, tar, grease, etc.).

Generally, the main parameters discussed to establish the working range for laser cleaning of different materials are: the ablation threshold defined as the lowest value of the incident fluence/intensity, to remove a significant quantity of material from a surface and the substrate damage threshold defined as the highest value for which the material is completely removed without the damage of the substrate. These are the limiting values for a working range which is recommended to be known or determined before the laser cleaning process begins. Another parameter which influences the laser cleaning process is the number of subsequent laser pulses which are incident at the same place. Therefore, to select the best cleaning practice, the ablation depth (as a function of incident laser fluence/intensity and pulse number) is an important parameter to be measured.

Laser cleaning experiments were realized on artificial samples (e.g. mock-ups) developed in order to simulate as better as possible different painting techniques and conditions. Therefore, we have considered two main categories: mural paintings (frescos) and easel paintings (oil and tempera based). Each category has its own specificity in respect of substrates, materials, and method of preparation, following traditional recipes. Generally, different color pigments for the subsequent layers were used. The support for the easel paintings was wood or canvas. In order to simulate old paintings the prepared samples were dried and artificially aged for a period of eight months in controlled conditions of temperature, humidity and especially of illumination.

Experiments were realized in conditions that respect the main requirements for a controlled and reproducible removal of painting layers from the mock-ups: selection and control of incident fluence and intensity, subsequent irradiation pulses or irradiation time, etc. In order to have an uniform transversal distribution of the laser fluence in the irradiation area an optical system was realized to deliver the laser beam on the mock-ups. Each mock-up sample was able to be translated in front of the laser spot to make irradiations in adjacent positions or in a continuous movement with a controlled translation speed.

The irradiation laser system was an Nd:YAG working on 1064, 532, 355 and 266 nm, with pulse length of 6 ns for 1064 nm and a frequency of 10 Hz (Fig. 1).

Custom software was developed to integrate the laser energy (and consequently the incident fluence/intensity, repetition rate, number of irradiation pulses or irradiation time) and implement it in the experimental set-up to ensure a centralized command. Also, the movement of the mock-up in front of the laser beam was commanded and controlled as step size or displacement speed (e.g. in the case of the continuous movement of the sample in front of the irradiation spot). The irradiated area was monitored with a CCD camera and the image displayed on a monitor in real time.

The irradiated area was analyzed from the point of view of the ablated surface and the thickness of the removed layer with optical microscopy, white light interferometry (WLI) and contact profilometry.

Fig. 1 – General view of the experimental set-up developed for laser cleaning experiments.

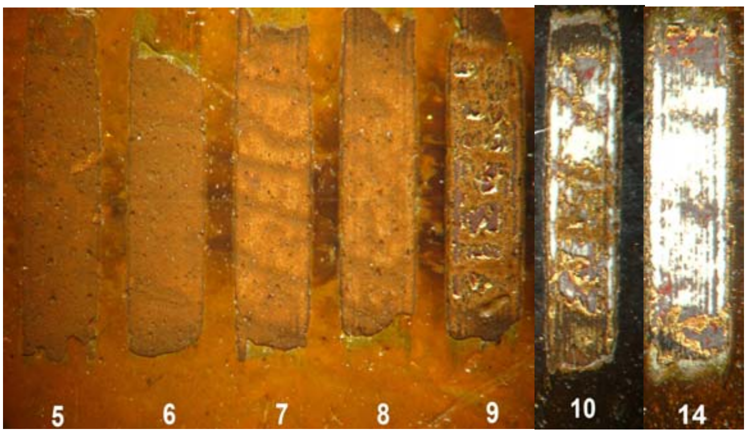

Fig. 2 – Sequence of laser removal of painting layers. Irradiation conditions: wavelength 355 nm; incident fluence 90 mJ/cm^2; subsequent irradiation pulses number (from left to right) 7, 10, 15, 20, 30; 5 sec., 60 sec.; the correspomding measured ablation depth was 15, 18, 21, 23, 25, not measured, 45 µm.

From the point of view of the thermal interactions *vs*. pulse length and UV wavelength are recommended because the heat affected zone for typical organic materials such as painting varnishes is estimated to be comparable to the optical penetration depth. As a consequence the ablation print after removal of a layer of varnish or painting material is free of melted material, as can be seen in Fig. 2. We present here experiences realized on a mock-up made on wood support, covered with ground powder of chalk with finings on which a gold leaf was displayed. The subsequent layers were ochre mastic, red bolus and black smoke (not uniform) all covered with dammar varnish colored with yellow pigment. The step-by-step removal of the subsequent layers was realized up to the gold layer. The laser action induces wrinkles on it and after more irradiation pulses the gold layer is removed also. Ablation depth was measured with a contact profilometer (Fig. 3) or with a white light interferometer (WLI). In Fig. 3 measured the ablation depth was about 12 µm and the roughness of the bottom of the ablation print was 0.7 µm The lowest measured ablation depth was 6 -7 µm for this sample.

In addition, the localized action of the laser beam can be seen in Fig. 3. The ablation print presents sharp borders, so the transition from the irradiated area is reduced on contrary to the painting layers removal with solvents. The roughness of the bottom of the ablation print indicates that no melted material appears in this irradiation conditions. The general characteristic of the presented irradiations was low incident fluence (in this case the ablation threshold was 60 mJ/cm^2), and the number of subsequent laser pulses or irradiation time controls the ablation depth or the thickness of the removed material.

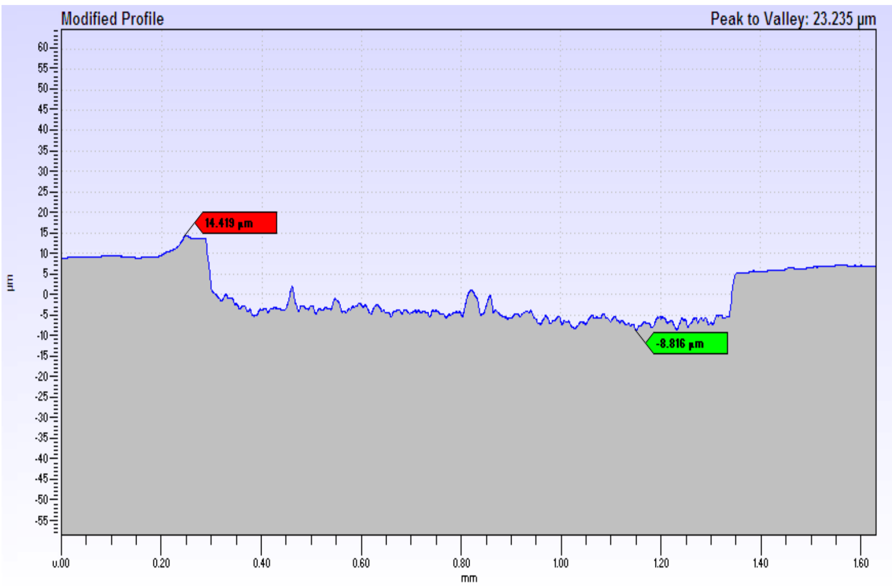

Fig. 3 – Ablation print profile. The measured ablation depth is about 12 µm.

a1 a2 90 mJ/cm2 | b1 b2 200 mJ/cm2 | c1 c2 280 mJ/cm2 | d1, d2 350 mJ/cm2 | e1, e2 400 mJ/cm2 | f1, f2 450 mJ/cm2

Fig. 4 – Laser (355 nm, 5 ns) cleaning tests on a mock-up on canvas support and painting layers: I. Ground-mixture of powder of chalk; II. Plaster + ochre oxide; III. Ultramarine; IV. Ultramarine+titan white; V. Varnish (cases noted 1 are irradiated with 120 pulses, noted 2 with 250 pulses.

If the irradiation fluence is too high the boiling of the not-removed painting layers can be induced (Fig. 4). The incident laser parameters which make the difference between a safe surface layer by layer removal without damages of the subsequent layers are the incident fluence/intensity, but also the number of irradiation pulses. These parameters have to be estimated for each painting layer before laser irradiation of a painting in order to have a safe treatment of the surface.

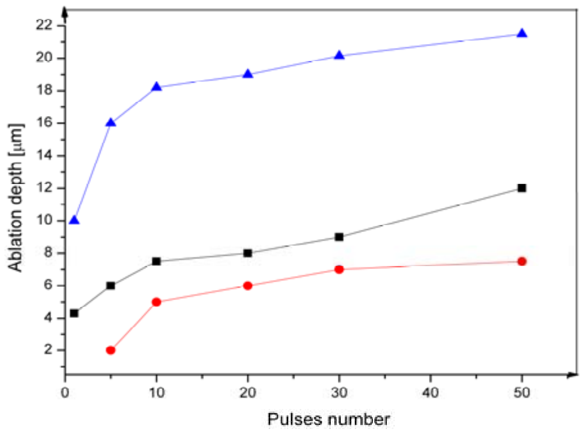

Fig. 5 – Ablation depth evolution as a function of subsequent irradiation pulse number for 3 incident fluences: 30 mJ/cm^2 – ●line, 90 mJ/cm^2 – ■ line, 200mJ/cm^2 – ▲ line (Mock-up: Support: WOOD; Substrate: ground powder of chalk with finings; layers: natural sienna iron rust, white and pink all in tempera egg emulsion 1/4, mastic varnish 12%, Patina and dummy dirt*: Opaque layer: Mixture of powder of chalk +plaster + black smoke + finings-4%).

Ablation depth measurements realized as a function of incident energy and/ or subsequent irradiation pulse number have evidenced that it is possible to select the film thickness to be removed in the micrometric range with a proper selection of irradiation parameters (Fig. 5).

Fig. 6 – Laser cleaning prototype general image.

From the extended set of tests we determined the incident laser parameters considered to be the milestones for the interaction of diferent painting layers with laser radiation and necessary to controll the laser cleaning process: safe irradiation maximum value, color change threshold, ablation threshold, efficient irradiation conditions, damage threshold.

3. LASER BASED SYSTEM AND TECHNOLOGIES FOR IN-SITU CLEANING OF PAINTING ARTWORKS

After the definition of the boundary conditions in which laser cleaning can be safely applied to a wide range of easel, mural painting materials and characteristic deposits the step from the laboratory tests to the controlled application of laser cleaning technology to painted objects was realized with the development of a new laser based system, mobile and flexible, to be used *in-situ*, for laser cleaning of paintings. The system is supported by diagnosis and control tools (e.g. Laser Induced Breakdown Spectroscopy technique for real time monitoring the cleaning operation, multispectral device for monitoring and artwork characterisation and colorimetry system for detection of chromatic changes upon laser cleaning), but also allowing human intervention through remote control of the cleaning system .

The laser cleaning prototype is composed by three major components:

– The cleaning cell, comprising the robotic manipulator, the optical laser delivery system and the scanning-head;
– The laser power source;
– The control cabinet, with the user interface.

In the cleaning cell the robotic system positions and delivers the laser beam from the laser source to the artwork as well it allows the positioning of all sub-systems present in the scanning head through its manipulator. A three linear axis Cartesian robot was specified and built during the project, in agreement with the project specifications.

The laser source is an Nd: YAG working on different wavelengths – from UV to IR – which demanded an accurate optical delivery system (based on broadband mirrors) in order to obtain a high homogeneity of the laser beam spots with each one of the wavelengths.

The scanning head mounted on the robot contains the laser beam delivering system and the distance sensor, and the systems for the monitoring and the control of the system operations (video control of the interaction region) the control tool (LIBS) and the multispectral and colorimetric cameras. The scanning head position is controlled by the end-user through the control system using a friendly interface (Fig. 6).

The general characteristics of the laser cleaning prototype are:

– Working area of ~ $1m^2$ (the displacement of the cleaning system allows higher areas to be treated);

– Nd:YAG laser source with interchangeable wavelengths, pulse length of 6 ns and repetition rates up to 10 Hz);

– Spot size on the artwork surface depending on the lens/mask combination (default configuration with ~ 3 mm × 3 mm);

– Scanning device with accurate radiation delivery of 0.1 mm;

– Live video for monitoring the cleaning process;

– Multispectral tools for imaging capture in the range of 300 nm to 1000 nm, with different imaging modes (UV reflection, visible reflection, UV fluorescence, false colours infrared, user-selectable wavelength imaging capture) and colorimetric analyses;

– Chemical identification and process control with LIBS (Laser induced breakdown spectroscopy) from 200 to 980 nm.

The laser based cleaning system integrates the control software, which allows the user to control all the available tools through a computer, having the following abilities:

– Definition and parameterisation of the area or areas to be cleaned or analysed;

– Control of the laser cleaning process allowing the configuration of the different laser parameters: wavelength, energy, repetition rate, number of pulses, etc.

4. CONCLUSIONS

This cleaning system will provide conservators/restorers with an alternative tool to complement the conventional methods. The developed device is capable of cleaning artworks that previously were unsatisfactorily treated by conventional techniques, avoiding at the same time the inherent problems related to the use of harmful chemical products. The laser-based system will provide SME end-user companies in Europe with an efficient and reliable advanced restoration tool that can be also integrated and combined with traditional methods. This new technology would increase their global competitiveness due to the new possibilities of restoring artworks that were not possible before.

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