

Doctoral School
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**Laser cleaning of roman fresco fragments and
holographic interferometric testing of the mechanical effects of the method**

Theses of DLA dissertation

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Introduction

Lasers are used in different areas of everyday life, and several examples of their application in art conservation have been published since the 1970s. Following their huge development, by now, the different laser based techniques increasingly gain ground in several areas of restoration, such as surface cleaning, materials analysis, and documentation using 3D imaging.

By means of lasers it is possible to remove different contamination types of various compositions and origin from artworks made of different materials, and excellent results can be achieved, if the numerous variable laser parameters are set correctly. Obviously laser cleaning is not a universal solution for all restoration problems either, therefore it is necessary to study its possible negative effects.

My master's thesis dealt with excimer laser cleaning of roman fresco fragments. That was the time I came in contact with the University of Pécs, Institute of Physics, where the possibility of laser applications in restoration interventions has been investigated since 2005. The topic of my doctoral dissertation is the sequel of that work. I extended my experience about the usage, advantage and disadvantage of the laser cleaning using different lasers (with different wavelength or length of pulse) on new samples.

It was my objective to investigate the potential negative photomechanical effects of laser cleaning too. Digital holographic speckle pattern interferometry (DHSPI) was used for this purpose. Using DHSPI, I came to know such a non-destructive method which has not been applied yet by the Hungarian conservators, in spite of the fact that it could be very useful in numerous cases (e.g. condition survey or observation of wall paintings, wooden panels).

In my dissertation, first I describe the physical background of laser surface cleaning based on literature data, and compare the traditional cleaning methods to laser cleaning. Then the parameters which affect the efficiency of the latter are discussed in detail, and also its possible negative effects are considered.

A method, called digital holographic speckle pattern interferometry (DHSPI) is introduced through literature data in the next chapter. This method is suitable for detection of the possible negative impact of the mechanical effects occurring during laser cleaning.

Thereafter, my own work will be discussed in detail. After presenting the objective, the selected or prepared samples are described, and the results of the laser cleaning and the DHSPI measurements are shown. Finally, conclusions are drawn regarding the applicability of laser cleaning.

In the masterwork belonging to this dissertation I investigated the practical applicability of the laser, as a conservator tool. First I show laser cleaning of roman wall-painting fragments, and then I describe the restoration of a completely new statue which underwent a serious paint attack.

Theses of the dissertation

1. Laser cleaning of excavated wall-painting fragments

In my doctoral research I investigated the possibility of laser cleaning of such excavated wall-painting fragments on which there is a hard, calcareous encrustation.

From the results of my experiments I concluded that - with carefully chosen parameters - laser cleaning can work effectively, so that it does not harm the paint layer of the fragment. Therefore, this method can be a good alternative or additional method of traditional interventions such as mechanical cleaning (scalpel, micro sand-blasting), solvent cleaning or poultices.

In the case of both 248 nm and 1064 nm wavelengths, the following sensitivity scale can be established from the most sensitive to the most resistant investigated pigment: cinnabar, coal-black, red iron-oxide, yellow iron-oxide, terre verte, Egyptian blue and lime white. The yellowing of the iron-oxide red was observed on certain samples, which effect had not been published in the literature before.

2. Demonstration of the effect of different consolidants on the laser cleaning of wall-painting fragments

Through my research, I demonstrated that the presence of the different consolidants effected the result of the laser cleaning. In many cases, the original fragments can be so fragile that their previous consolidation is necessary, although this intervention carries the risk that the contamination will be bound to the surface.

During the laser cleaning (KrF 248 nm) of the original wall-painting fragments we could find examples for the positive effect attributed to the consolidant (Syton X30¹, MfCO²), when it protected the paint layer from splitting off or damage. However, it also occurred that

¹ aqueous silica acid dispersion

² finely dispersed, aqueous dispersion of an acrylic copolymer

the consolidant (Remmers KSE 300³) made the elimination of the contamination by laser (248 nm and 1064 nm) more difficult.

The KrF laser cleaning with 248 nm wavelength of the consolidated model samples (Remmers KSE 300) was more effective than the Nd:YAG laser cleaning with 1064 nm. Furthermore, I found for both wavelengths, that the necessary number of pulses for the intact model samples cleaning is less in the case of consolidated samples than in the case of not consolidated samples, therefore for these samples it is more effective to ablate the consolidant and the contamination together, than the encrustation itself.

Based on all the above, it is recommended to consider the possibility of laser cleaning for those samples which need consolidation and cleaning at the same time.

3. Mechanical tests of original wall-painting fragments and fresco models

Mechanical tests were taken for the characterization of the original and model wall-painting samples. The test results proved that the density of the bulk of the custom-made model fresco samples was uniform ($\sim 1,65 \pm 0,03 \text{ g/cm}^3$) and was in a reasonable agreement with those of the original excavated Roman fresco fragments, the density of which was measured to vary between $1.69\text{-}2.41 \text{ g/cm}^3$. By the ultrasound velocity measurements in consolidated and non-consolidated samples and by the results that the density does not change due to consolidation, I experimentally demonstrated that the consolidant strengthened the bonds in the bulk of the fresco. I found, that the surface hardness determined by Duroskep rebound values was increased by consolidation (with Remmers KSE 300) on the reverse side of the samples, while consolidation did not significantly affect the hardness measured on the front, encrusted side of the samples. From this fact I concluded that the consolidant reacts with the bulk of the fresco and with the material of the model crust differently.

4. The applicability of Digital Holographic Speckle Pattern Interferometry (DHSPI) for the detection of photomechanical effects

Based on the results of the experiments testing the mechanical effects of the laser cleaning by an interferometric method, I concluded that it was possible to detect the photomechanical effect of the laser cleaning on the original wall-painting fragments and model samples equally by the DHSPI system.

³ solvent-free stone strengthener on a silicic acid ethyl ester base.

The possible consequences caused by the movement of the original wall-painting between the laser cleaning and the DHSPI measurements can be avoided by performing the cleaning and the testing simultaneously. I excluded the accumulated influence of subsequent cleaning events by separate tests on uniform custom-made model samples.

There were two samples among the model samples, on which the fringe patterns revealed unintentional defects that had been present in the samples before cleaning (shown by the reference fringe patterns), and the joints of the broken samples were also visible well on the interferograms. Based on these results, I concluded that the DHSPI method is suitable for the examination of the mechanical effects of laser cleaning.

5. Comparison of the surface cleaning by KrF (248 nm) and Nd:YAG (1064 nm) lasers by DHSPI

The DHSPI tests were carried out on eight different groups of model samples detecting the effect of KrF (248 nm) and Nd:YAG (1064 nm) cleaning lasers. From the laser cleaning tests I concluded that the number of necessary pulses is almost equal for KrF and Nd:YAG laser cleaning within the groups of not consolidated intact and broken samples. On the other hand, as I already mentioned in the 2. thesis, in the case of consolidated samples, KrF cleaning was found to be more efficient.

The number of pulses necessary for the laser cleaning of the broken samples is higher for both the KrF (248 nm) and Nd:YAG (1064 nm) lasers than the number of those needed for the intact samples. In my opinion, this follows from the fact that the surface of the broken samples is less plain after the re-cementation, therefore the thickness of the encrustation above the joint can be locally different. The laser cleaned area was positioned there, which explains the higher number of necessary pulses in the case of broken samples.

I also observed that in those cases when the paint layer behind the crust was damaged by the Nd:YAG laser, it was more severe and extended than the damages caused by the KrF laser. This occurs because in these series of experiments cleaning was done in dry conditions, because the wetting caused noisy interferograms. Given that the effect of wetting is significant in the IR cleaning regime its absence resulted in non-satisfactory results.

6. DHSPI measurements do not show new structural defects attributed to the laser cleaning

From the results of the DHSPI experiments I found that the fringe patterns of either the intact or the accidentally or intentionally weakened (broken and re-cemented) samples do not show additional defects that could be attributed to the laser cleaning. Furthermore, according to the shape and density of the curved fringes, neither accidentally, nor intentionally produced defects were changed significantly by the laser cleaning.

I observed that on the fringe patterns of the broken samples, the centre of the fringes is usually over the joint or over the intersection of the joints, showing that these are the regions where the displacement of the sample is the greatest.

By comparing the relative displacements of two points being 5 cm apart on different samples, it was concluded that the changes in the samples' response to the thermal excitation are not larger than the differences that had been present between the samples in their reference state. It was also seen that the samples respond to the excitation more uniformly after 1 month of relaxation than in their reference state and 1h after the cleaning session.

The laser intervention increased the average relative displacement (in response to $\sim 3^{\circ}\text{C}$ increase in temperature) only to a negligible extent, and even this negligible effect was undetectable after 1 month of relaxation.

7. Practical experiences of laser cleaning based on the masterwork

In my masterwork I used different lasers and parameters as a potential alternative or additional method of the traditional techniques on different artworks. They significantly differed in their age, size, components and also in the type of contamination on them.

Using KrF (248 nm) and Nd:YAG (1064 nm) lasers, I successfully eliminated the well attached, calcareous encrustation of variable thickness from the original, roman wall-painting fragments and the filmy Plextol layer appearing on the paint surface in a way that the underlying paint layer was not harmed. The reactions of the pigments were in good agreement with my previous experiences thus the Egyptian blue and lime white did not show any discoloration. Neither the iron-oxide yellow changed colour, but the iron-oxide red paint layer darkened when the Nd:YAG (1064 nm) laser was applied, so I used KrF laser (248nm) for the cleaning of that.

In order to achieve the best cleaning result, I combined laser cleaning with different conservation cleaning methods when eliminating the polluting paint layer from the István Károlyi statue from Fót. I reduced the thickness of the polluting paint layer by micro sand-blasting and with solvent poultices, then I removed the rest of the paint by a Nd:YAG laser (at 532 nm and 1064 nm) so that the characteristic, original scoring of chisels were preserved. To eliminate the traces of the paint layer left in the hollows, cracks, hard-to-reach areas, I used graffiti-shadow remover gel (Tensid AGS 60).

During the laser cleaning of the wall-painting fragments I observed that the wetting with distilled water caused yellowing of the paint layer, therefore I applied the Nd:YAG (1064 nm) laser without wetting. However, it was possible to remove the Plextol layer from the surface by wet Nd:YAG laser cleaning without damaging the paint layer. In the case of the laser cleaning of the statue, wetting of the surface with alcohol accelerated the process at both wavelengths (532nm, 1064nm), and it was more effective than wetting with distilled water.

The size of the objects determined the laser setup. I fixed the articulated arm and I moved the samples when I performed laser cleaning of the few cm wall-painting fragments. On the contrary, during the laser cleaning of the statue, I took advantage of the mobility of the laser and the feasibility of the articulated arm and I moved the device around the artwork.

Based on the extensive experience gained during this work, I found that it is worth to consider the possibility of laser cleaning for the planning of the restoration process. As long as we can vary the parameters in a quite broad scale, we can hope that the laser offers an effective solution for the cleaning problems that cannot or can hardly be solved by traditional methods.

Publications related to the doctoral research

Zs. Márton, I. Kisapáti, Á. Török, V. Tornari, E. Bernikola, K. Melessanaki, P. Pouli: *Holographic testing of possible mechanical effects of laser cleaning on the structure of model fresco samples*, NDT&E International, vol. 63, pp. 53–59, 2014.

Kisapáti I., Márton Zs.: *Római kori falkép-töredékek lézeres tisztítása*, Műtárgyvédelem, vol. 37-38, pp. 185-191, 2012-13.

Kovács-Mravik P., Galambos É., Kisapáti I., Márton Zs., Sajó I., Sándorné Kovács J., J. Schultz, Tóth A.: *Az Iparművészeti Múzeum Damaszkusz szobája*, Műtárgyvédelem, vol. 37-38, pp. 7-37, 2012-13.

Zs. Márton, I. Kisapáti, P. Pouli, E. Bernikola, V. Tornari: *Laser cleaning of excavated fresco fragments; testing and optimization of laser parameters and structural monitoring by means of Digital Holographic Speckle Pattern Interferometry in Lasers in the Conservation of Artworks IX*, ed: Saunders et al. Archetype Publications, London, 2013

J. Marczak, M. Strzelec, R. Ostrowski, A. Rycyk, A. Sarzynski, W. Skrzeczanowski, Koss, R. Szambelan, R. Salimbeni, S. Siano, J. Kolar, M. Strlic, Z. Márton, I. Sánta, I. Kisapáti, Z. Gugolya, Z. Kántor, S. Barcikowski, P. Engel, M. Pires, J. Guedes, A. Hipólito, S. Santos, A.S. Dement'ev, V. Švedas, E. Murauskas, N. Slavinskis, K. Jasiunas, M. Trtica: *Advanced laser renovation of old paintings, paper, parchment and metal objects in Lasers in the Conservation of artworks VII*, ed: Castillejo et al. Taylor and Francis Group, London, 2008

Szörényi Bella: *Graffiti eltávolítása kőfelületekről*, supervisor: Kisapáti Ivett, consultant: Dr. Márton Zsuzsanna, MKE, 2014.

Participation on conferences, presentations related to the doctoral research

40. Nemzetközi Restaurátor Konferencia, Budapest, 2015., oral presentation:
Kisapáti I., Márton Zs.: A főtí Károlyi István szobor lézeres tisztítása

LACONA X, Sharjah, 2014., poster:

Zs. Márton, I. Kisapáti, V. Tornari, E. Bernikola, K. Melessanaki, E. Tsiranidou, K. Hatzigiannakis, P. Pouli: Holographic evaluation of the structural condition of laser-cleaned model fresco samples

ICOM-CC: Heritage Wood: Research & Conservation in the 21st century, Varsó, 2013.:
P. Kovács-Mravik, É. Galambos, Zs. Márton, I. Kisapáti, J. Schultz: A polychrome wooden interior from Damascus: a multi-method approach for the identification of manufacturing techniques, materials and art historical background

Laserlab User Meeting, Marseille, 2013.:

I. Kisapáti, Zs. Márton, I. Bernikola, E. Tsironidou, C. Hatzigiannakis, P. Pouli, V. Tornari: Laser-cleaning of excavated fresco samples and DHSPI testing of model fresco samples for differentiating the photomechanical effects induced by the laser cleaning

CHARISMA Itinerant course on stone conservation, Amsterdam, 2013., oral presentation:

Kisapáti I., Márton Zs., P. Pouli, K. Melesanaki, E. Bernikola, E. Tsiranidou, K. Hatzigiannakis, V. Tornari: DHSPI testing of laser-cleaned model fresco samples for monitoring the photomechanical effects induced by laser cleaning,

II. Interdiszciplináris Doktorandusz Konferencia, Pécs, 2013., oral presentation:

Kisapáti I., Márton Zs., Pouli P., Melesanaki K., Bernikola E., Tsiranidou E., Hatzigiannakis K., Tornari V.: Freskó modelleken végzett lézeres tisztítás által okozott fotomechanikai hatások vizsgálata digitális holografikus szemcskép interferometriával

LATKÖV - Lézeres és analitikai technikák a kulturális örökség védelmében, Pécs, 2013. február 8-9., oral presentation:

Márton Zs., Kisapáti I., Tóth T.: Lézeres felület-tisztítás és LIBS analízis műtárgyakon. Fizikai alapok és gyakorlati alkalmazások

Kisapáti I.: Holográfia alkalmazása műtárgyak vizsgálatára

CHARISMA – Advanced laser-based techniques in art conservation, diagnostics and analysis, FORTH, Institute of Electronic Structure and Laser, Kréta, 2012., poster: I. Kisapáti, P. Pouli, I. Sajó, K. Kovács, Zs. Márton: Laser induced yellowing of red ochre pigments

Magyar Képzőművészeti Egyetem, Budapest, 2012., oral presentation:

Márton Zs., Kisapáti I., Tóth T.: Lézeres felület-tisztítás és LIBS analízis műtárgyakon. Fizikai alapok és gyakorlati alkalmazások

LACONA IX, London, 2011.,

Zs. Marton, I. Kisapati, P. Pouli, E. Bernikola, V. Tornari: Laser cleaning of excavated fresco fragments; testing and optimization of laser parameters and structural monitoring by means means of Digital Holographic Speckle Pattern Interferometry

XXXVI. Nemzetközi Restaurátor Konferencia, Budapest, 2011., oral presentation:

Márton Zs., Kisapáti I.: Különböző típusú lézerek alkalmazhatósága freskótöredékek tisztítására

XXXV. Nemzetközi Restaurátor Konferencia, Budapest, 2010., oral presentation:

Kisapáti I., Márton Zs., E. Bernikola, P. Pouli: Római kori freskótöredékek lézeres tisztításának ellenőrzése holografikus interferometriával

XXXIV. Nemzetközi Restaurátor Konferencia, Budapest, 2009., poster:

Kisapáti I., Márton Zs.: Római kori freskótöredékek lézeres tisztítása

Lasers in the Conservation of Artworks (LACONA) VII, Madrid, 2007., poster:

J. Marczak, M. Strzelec, R. Ostrowski, A. Rycyk, A. Sarzynski, W. Skrzeczanowski, A. Koss, R. Szambelan, R. Salimbeni, S. Siano, J. Kolar, M. Strlic, Z. Márton, I. Sánta, I. Kisapáti, Z. Gugolya, Z. Kántor, S. Barcikowski, P. Engel, M. Pires, J. Guedes, A. Hipólito, S. Santos, A.S. Dement'ev, V. ©vedas, E. Murauskas, N. Slavinskis, K. Jasiunas, M. Trtica: Advanced laser renovation of old paintings, paper, parchment and metal objects