Residual thermal effects in laser ablation of metals

Anatoliy Y. Vorobyev and Chunlei Guo
The Institute of Optics, University of Rochester, Rochester, New York 14627, USA
E-mail: guo@optics.rochester.edu

Abstract. Thermal energy remaining in Sn and Ti samples following femtosecond and nanosecond laser ablation is studied as a function of laser fluence and ambient gas pressure. When the laser fluence is above a certain threshold value, we find a significant enhancement in residual thermal energy deposition in air but a decrease in vacuum, and these effects occur for both femtosecond and nanosecond laser ablation. In contrast to the previous belief, our study demonstrates that femtosecond and nanosecond laser ablation share similar residual thermal effects.

1. Introduction
Pulsed laser ablation of metals has been a topic of numerous studies [1-3]. However, only very few studies [4-10] have investigated the residual thermal effects in metals following laser ablation. These studies have shown that thermal energy remaining in an ablated sample undergoes an abrupt enhancement following single-pulse ablation with microsecond CO2 laser [4-7], nanosecond (ns) Nd:YAG laser [8], and nanosecond ruby laser [9]. More recently, an enhanced residual thermal energy deposition has also been seen following multi-pulse ablation with femtosecond (fs) Ti:sapphire laser [10]. For microsecond and nanosecond laser pulses, the energy transfer from laser-induced plasmas to the sample has been proposed as an explanation for the observed enhancement in thermal energy coupling to the sample [4,5]. This explanation, however, has later been shown to be incomplete [6]. In this paper, we study the residual thermal effects following single pulse ablation with both ns and fs lasers. We show that the enhanced residual thermal energy coupling occurs in both ns- and fs-laser ablation in a gas medium but does not occur in vacuum. The pressure of the ambient gas is found to be a critical parameter that determines the residual heating of metals following laser ablation.

2. Experimental setup
In laser ablation, a fraction of the directly absorbed laser energy will remain in the irradiated sample and produce a heat-affected zone. This residual energy, initially stored in the heat-affected zone, will dissipate into the entire bulk of the sample and finally lead to a residual heating of the entire sample. Besides the direct residual energy deposition, there are also different indirect mechanisms of residual heating, such as energy transfer from plasmas, re-deposition of ablated material, and exothermic chemical reactions. In this paper, using a calorimetric technique [4-10], we study the total thermal energy remaining in the metal...
sample following laser ablation. To characterize residual thermal energy deposition, we define a residual energy coefficient (REC) as \( K = E_R / E_I \), where \( E_R \) is the residual thermal energy remaining in the sample following ablation and \( E_I \) is the incident laser pulse energy.

In our experiment, we use a Ti:sapphire laser system that generates 65-fs laser pulses at a wavelength of \( \lambda = 0.8 \mu m \) with pulse energy of 1.5 mJ and a ruby laser that produces 45-ns pulses at \( \lambda = 0.69 \mu m \) with pulse energy of 0.6 J. The laser beam is focused with a lens onto a sample at normal incidence. A fraction of the incident pulse energy \( E_I \) is split off by a beamsplitter and measured with a pyroelectric joulemeter that allows \( E_I \) to be determined. The error of measuring \( E_I \) is estimated to be \( \pm 5\% \). The residual thermal energy \( E_R \) retained in the sample after ablation induces a bulk temperature increase, \( \Delta T \), of the sample. We measure \( \Delta T \) with a thermocouple sensor. Using known specific heat capacity \( c_p \) and the mass \( m \) of the sample, the residual energy can be calculated from \( E_R = mc_p \Delta T \). The error of measuring \( E_R \) is estimated to be \( \pm 10\% \). A more detailed description of this calorimetric method can be found in our previous study [10]. Having measured \( E_I \) and \( E_R \), we can deduce the residual energy coefficient \( K = E_R / E_I \). In this paper, we study REC of Sn and Ti in various ambient gas environments as a function of laser fluence \( F = E_I / S \), where \( S \) is the laser beam area on the sample. The surface of the bulk samples is mechanically polished. An X-Y stage is used to translate the sample to an undamaged spot after each laser shot. Ablation threshold, \( F_{abl} \), is determined as the onset of visible surface damage with subsequent examination of the damaged spot under an optical microscope. Plasma formation threshold, \( F_{pl} \), is determined by the onset of violet radiation from the irradiated area on the sample [11-13].

3. Results and discussion

The plots of REC versus laser fluence for Sn and Ti samples following femtosecond laser ablation in 1-atm air and in vacuum with a base pressure of \( P = 0.01 \) torr are shown in Figs. 1 and 2, respectively. For both metals, REC increases abruptly in 1-atm air above a certain threshold value of laser fluence, \( F_{enh} \), but decreases in vacuum. Our data show that the enhancement of residual thermal energy deposition in air is very large. For example, at \( F = 10 \) J/cm\(^2\) in air, about 80\% and 95\% of incident fs laser energy remains as residual thermal energy for Sn and Ti samples, respectively. Ablation and plasma formation thresholds are also determined for fs-laser ablation in 1-atm air, as shown in Figs. 1 and 2. If we compare \( F_{abl} \), \( F_{pl} \), and \( F_{enh} \), we see that the relationship \( F_{abl} > F_{pl} > F_{enh} \) holds for both Sn and Ti. For fs-laser ablation of Ti, the thresholds for ablation and plasma formation in vacuum are found to be identical to those in air within experimental uncertainty. The numerical values of \( F_{abl} \), \( F_{pl} \), and \( F_{enh} \) obtained in our experiments for fs-laser ablation are given in Table 1.

| Metal    | Ablation threshold (J/cm\(^2\)) | Plasma threshold (J/cm\(^2\)) | Threshold for enhanced thermal coupling (J/cm\(^2\)) |
|----------|---------------------------------|------------------------------|----------------------------------------------------|
|          | air    | vacuum | air    | vacuum | air             |
| Tin      | 0.042  | -      | 0.22   | -      | 2.0             |
| Titanium | 0.055  | 0.055  | 0.075  | 0.075  | 0.44            |

**Table 1.** Threshold values for fs-laser ablation, plasma formation, and thermal coupling enhancement for Sn and Ti.
Fig. 1. Residual energy coefficient of Sn versus laser fluence for single-pulse femtosecond laser ablation in air and vacuum. The thresholds of ablation and plasma formation are shown for femtosecond laser ablation in air.

Fig. 2. Residual energy coefficient of Ti versus laser fluence for single-pulse femtosecond laser ablation in air and vacuum. Within the experimental uncertainty, the thresholds for ablation and plasma formation are identical for air and vacuum.
Data in Figs. 1 and 2 show that air pressure affects the thermal energy coupling to metals significantly. To further understand the pressure effect on REC, we measure REC versus ambient air pressure at a fixed incident laser fluence and the dependence, \( K(P) \), for Sn and Ti is plotted in Fig. 3. We see that REC decreases slowly when air pressure is reduced from 760 to about 50 torr. An abrupt drop of REC occurs when pressure decreases from 50 to about 0.05 torr. Further decrease of pressure only induces a very slight decrease in REC. Therefore, the lower pressure range \( (P < 0.05 \text{ torr}) \) is the preferred condition to minimize thermal load in fs-laser ablation.

![Graph showing residual energy coefficient vs air pressure for Sn and Ti](image)

**Fig. 3.** Residual energy coefficient of Sn and Ti versus air pressure for single-pulse femtosecond laser ablation at \( F = 10.1 \text{ J/cm}^2 \).

Figs. 4 and 5 show dependence of REC versus laser fluence for ablation of Sn, and Ti with single nanosecond pulse in air and in vacuum. It is seen that for both Sn and Ti, an enhancement of thermal coupling occurs in 1-atm air but is absent in vacuum. This behaviour of REC is similar to that for fs-laser ablation. From Figs. 4 and 5, we can see that about 50% and 60% of incident ns-laser energy can be retained as residual thermal energy in Sn and Ti samples, respectively. For both Sn and Ti, \( F_{pl} \) is found to be approximately identical to \( F_{emb} \) for ns-laser ablation in air while \( F_{abl} \) has a smaller value, i.e., \( F_{abl} < F_{pl} \approx F_{emb} \). This relationship is different than that for fs-laser ablation \( (F_{abl} < F_{pl} < F_{emb}) \). The values of \( F_{abl}, F_{pl}, \) and \( F_{emb} \) for ns-laser ablation are summarized in Table 2.

**TABLE 2.** Threshold values for ns-laser ablation, plasma formation, and thermal coupling enhancement for Sn and Ti.

| Metal   | Ablation threshold (J/cm²) | Plasma threshold (J/cm²) | Threshold for enhanced thermal coupling (J/cm²) |
|---------|----------------------------|--------------------------|---------------------------------------------|
|         | air | vacuum | air | vacuum | air                          |                                            |
| Tin     | 0.6 | 2.0     | 1.0 | -       | 1.0                          |                                            |
| Titanium| 0.8 | 2.1     | 1.0 | -       | 1.0                          |                                            |
Fig. 4. Residual energy coefficient of Sn versus laser fluence following ablation with single 45-ns pulse from ruby laser in air and vacuum. In air, the thresholds for the enhanced thermal coupling and plasma formation are found to be identical within the experimental uncertainty.

Fig. 5. Residual energy coefficient of Ti versus laser fluence following ablation with single 45-ns pulse from ruby laser in air and vacuum. In air, the thresholds for the enhanced thermal coupling and plasma formation are found to be identical within the experimental uncertainty.
4. Conclusion

We have measured residual thermal energy deposition in Sn and Ti following both fs- and ns-laser ablation. In air, residual thermal energy deposition is higher for fs-laser ablation than ns-laser ablation. Therefore, residual thermal effects appear to play a greater role in fs-laser ablation despite the fact that the heat-affected zone is smaller for fs-laser ablation compared to ns-laser ablation [14]. For both fs- and ns-laser ablation in vacuum, residual energy coefficient reduces to a constant level of about 0.1-0.2 as laser fluence increases. In contrast to the previous belief, our study demonstrates that femtosecond and nanosecond laser ablation share similar residual thermal effects.

Acknowledgement

The research was supported by National Science Foundation.

References

[1] Bäuerle D 2000 Laser Processing and Chemistry 3rd edn (Berlin: Springer)
[2] Hubler GK and Chrisey DB eds 1994 Pulsed Laser Deposition of Thin Films (New York: Wiley & Sons)
[3] Miller JC and Haglund RF eds 1998 Laser Ablation and Desorption (New York: Academic)
[4] Marcus S, Lowder JE and Mooney DL 1976 J. Appl. Phys. 47 2966
[5] Maher WE and Hall RB 1977 J. Appl. Phys. 49 2254
[6] McKay JA, Bleach RD, Nagel D J, Schriemph J T, Hall R B, Pond C R and Manlief S K 1979 J. Appl. Phys. 50 3231
[7] Ursu I, Mihailescu IN, Apostol I, Dinescu M, Hening A, Stoica M, Prokhorov AM, Ageev VP, Konov V I and Tokarev V N 1984 J. Phys. D: Appl. Phys. 17 1315
[8] Golodenko NN, Vorobyev AV, Kuzminchev VM and Guzhva V G 1976 Radiotekhnika (Kharkov) 38 138 [In Russian]
[9] Vorobyev AV 1985 Sov. J. Quantum Electron. 15(4) 490
[10] Vorobyev AV and Guo C 2005 Appl. Phys. Lett. 86 011916
[11] Andreev SI, Verzhikoskii IV and Dymshits Yu I 1971 Soviet Phys.-Tech. Phys. 15 1109
[12] Maher WE, Nichols DB and Hall RB 1980 Appl. Phys. Lett. 37 12
[13] Rosen DJ, Mitteldorf J, Kothandaraman G, Pirri A N and Pugh E R 1982 J. Appl. Phys. 53 3190
[14] Le Harzic R, Huot N, Audouard E, Jonin C, Laporte P, Valette S, Fraczkiewicz A and Fortunier R 2002 Appl. Phys. Lett. 80 3886