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In situ photography of picosecond laser ablation of nickel

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Abstract

This work experimentally investigated the time evolution of nickel ablation induced by high-energy picosecond laser pulses. A Nd:YAG laser with a 25 ps pulsewidth was used to perform a pump–probe microphotography experiment. The fundamental and second harmonic wavelengths were used for the pump and probe beams, respectively. The probe beam was delayed in time with respect to the pump beam and was reflected from the ablated surface into a camera to obtain time-resolved images. Photographs of the sample were obtained at the beginning of pump laser irradiation and continued until a time delay of 800 ps. Photographs revealed attenuation of the probe beam beginning during the pump pulse duration and lasting as long as the maximum time delay. Possible explanations of the probe beam attenuation are discussed, including homogeneous nucleation and the metal–dielectric transition near the thermodynamic critical temperature.

Keywords: Picosecond laser; Ablation; Heat transfer; Homogeneous nucleation; Phase explosion

1. Introduction

Ablation of materials using short pulsewidth lasers is a topic of much interest for applications such as laser micromachining and pulsed laser deposition (PLD) of thin films [1]. These applications use lasers with pulsewidth from tens of nanoseconds (ns) to as short as tens of femtoseconds (fs). Shorter pulsewidths are attractive for micromachining applications since the short laser pulse duration reduces thermal damage to the surrounding area. In order to optimize a laser ablation process, it is necessary to thoroughly understand the physical processes by which laser ablation proceeds. However, laser technology has progressed at a rate that has surpassed our understanding of laser–material interactions, and short pulse laser ablation is not well understood. Most ablation processes involve thermal transport and phase change, with phase change proceeding by normal vaporization at a free surface and volumetric boiling by homogeneous nucleation [2–4]. Recently, it was shown that the most likely mechanism by which mass is removed during picosecond (ps) laser ablation of metals is homogeneous nucleation in a superheated molten surface layer [5]. This is because normal vaporization will remove an insignificant amount of material on a picosecond time scale. Since heat conduction from the irradiated region will also remove little heat during this time duration, surface temperatures may become extremely high. When the molten surface is superheated to approximately $0.9T_{cr}$ (thermodynamic critical temperature), large fluctuations in liquid density result in a high rate of vapor nuclei formation (homogeneous nucleation) in the superheated liquid. This rate of nuclei formation increases by several orders of magnitude

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over a very short temperature span, resulting in a rapid liquid–vapor phase change (phase explosion) that will eject a mixture of vapor and liquid droplets [2]. There are still questions regarding phase explosion induced by picosecond laser pulses, since a finite amount of time is required for the formation of an equilibrium distribution of homogeneous nuclei in the superheated melt [4,5]. This time lag for nuclei formation has been calculated to be on the order of 1–10 ns [6], which makes the formation of homogeneous nuclei unlikely during heating with laser pulses less than 1 ns. This work addresses these issues by performing time-resolved imaging of a metal surface during and after pulsed laser heating in an attempt to observe the laser ablation phenomenon.

2. Experimental apparatus and procedure

The experimental apparatus is shown in Fig. 1. The laser source is a mode-locked Nd:YAG laser with a pulsewidth of 25 ps (FWHM). The laser beam is split to form two beams, the pump beam and the probe beam. The pump beam is used for ablation of the sample at the fundamental wavelength (1064 nm). The probe beam is used for imaging of the sample surface during ablation. After the pump and probe beams are split, the probe beam is directed through a second harmonic generator to convert a fraction of the light to a wavelength of 532 nm. Any light remaining at the fundamental wavelength is removed in an infrared absorbing filter. The probe beam then travels through a retroreflecting prism that is mounted on a micrometer stage. Moving the position of the prism changes the path length of the probe laser and the arrival time of the probe beam (with respect to the pump beam) at the sample surface. The probe beam is incident on the sample at an angle of approximately 25°. The reflected portion of the probe beam is viewed by a microphotography system at the same angle. Photographs are taken with 400 speed black and white film. Separate photographs were taken at different time delays, and multiple sets of photos were taken at a given laser fluence. The sample is moved after each photograph such that each photograph is of a region that was not previously ablated. The exact spatial irradiance distribution of the laser beam on the target was not known, but appeared to be semi-Gaussian from the

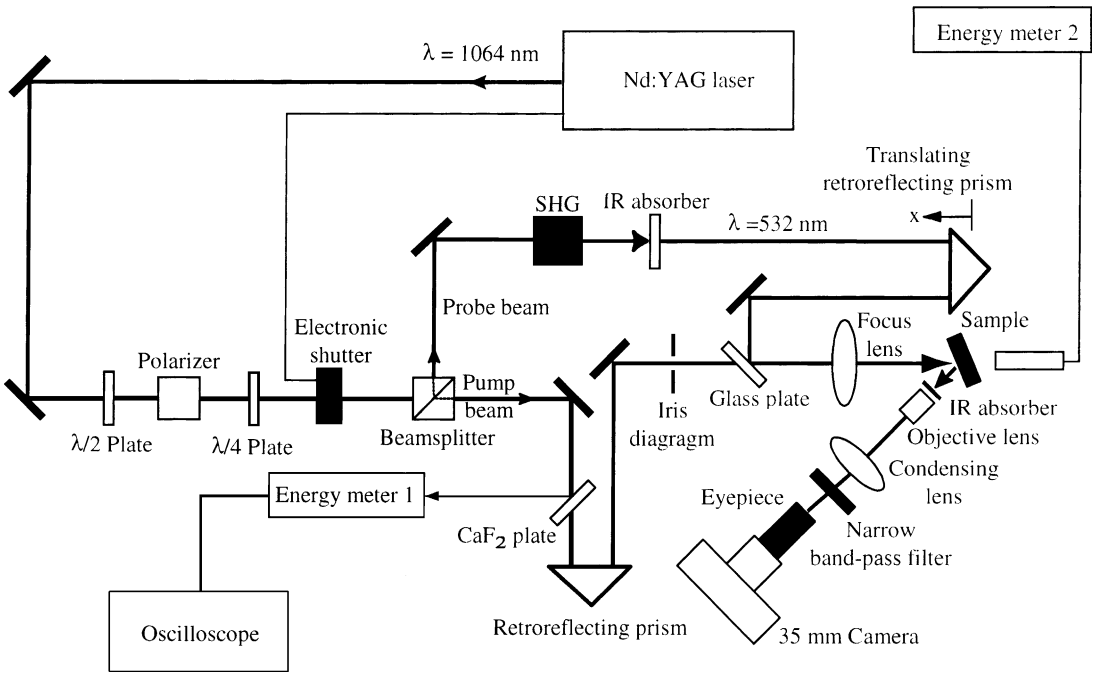

Fig. 1. Experimental apparatus for pump–probe microphotography.

observed damage pattern. The spot diameter was measured from visual observation of the damaged area at high fluence to be about 110 μm. Experiments were performed for average incident laser fluences ranging from 1.2 to 5.3 J/cm^2. It was determined by previous experiments that the threshold fluence for phase explosion in nickel is 2.0 J/cm^2 [4]. Time delays ranged from −30 ps to approximately 800 ps. A time delay of zero represents the time at which the peak irradiance of the pump and probe beams overlap in time. The first two experiments were performed below the phase explosion threshold, while in the remainder of the experiments the fluence exceeded that required for phase explosion. In all the experiments, the power density is not sufficient to cause the non-linear absorption effect [7].

3. Results and discussion

Microphotographs below the phase explosion threshold are shown in Fig. 2 for an average fluence of 1.2 J/cm^2. The fluence is noted below each photograph. The variation in fluence is a result of pulse-to-pulse variations in laser energy. The initial photographs at time delays from −30 to 10 ps show only reflected probe beam light from the nickel surface, which appears as white in the photographs. The square region shown is approximately 145 μm × 145 μm, much larger than the pump beam diameter. The reflected probe beam becomes attenuated in the central region of the photographs after a time delay of 20 ps. This attenuated region increases to a diameter of 64 μm at 100 ps, with some fluctuations in diameter due to changes in pump beam irradiance. The level of attenuation also increased between 20 and 100 ps. After 100 ps the diameter does not change significantly, although the level of attenuation appears to decrease gradually with time, as indicated by the increased amount of reflected light reaching the camera. The final photograph at infinity was taken several seconds after the pump pulse, and no surface damage is visible. This is to be expected since this fluence is below the phase explosion threshold.

It should be noted that the ablation threshold discussed here is a threshold for a significant amount of material being removed by a laser pulse causing visible damage to the target. Below the threshold, experiments showed that the target surface was roughened, but no net material removal resulted. If multiple pulses were allowed to reach the surface, only further

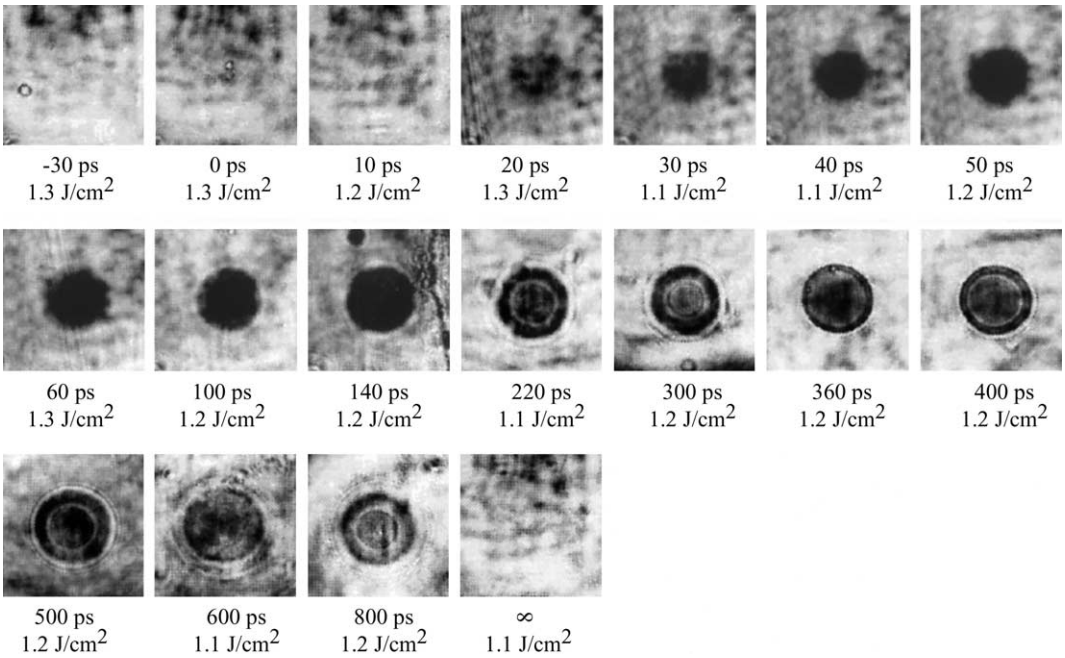

Fig. 2. Transient microphotographs of ablation at average fluence of 1.2 J/cm^2.

roughening was observed with no change in depth. It was found through a numerical calculation that surface evaporation during picosecond laser heating does not cause a significant amount of material removal (less than 1 Å) [5]. In other words, surface evaporation may still occur at the laser fluence used in this experiment (1.2 J/cm^2). The rings around the laser irradiated spot shown at 220–600 ps could be due to the gas dynamic effect from evaporation, though they can also be caused by sudden heating and expansion of the air adjacent to the heated surface.

The remainder of the photographs are for experiments above the phase explosion threshold fluence of 2.0 J/cm^2. Results of ablation at an average fluence of 2.3 J/cm^2 are displayed in Fig. 3. Probe beam attenuation began as early as 0 ps, with the diameter of the attenuated region increasing to approximately 81 μm at 50 ps. A distinct, sharp boundary to this region is visible after 140 ps. The level of attenuation again decreases very gradually with time, and at a time delay of 800 ps the entire ablated region has a reflectivity similar to that of the non-ablated region. This region also has a sharp boundary that coincides with the damaged region shown in the final photograph at infinity. Photographs at an average fluence of 3.1 J/cm^2 are shown in Fig. 4. Probe beam attenuation begins at −10 ps and expansion of the attenuated region continues until approximately 70 ps. The sharp boundary to the ablated region appears at approximately 100 ps, with a diameter of 93 μm. Reduction in probe beam attenuation is noticed after 300 ps. The final photograph at infinity has a diameter of approximately 93 μm, which is the same as the photograph at 800 ps. Similar results are seen in the photographs of Fig. 5 at an average fluence of 5.3 J/cm^2. At this fluence probe beam attenuation beginning at −30 ps and the attenuated region is fully expanded to a diameter of approximately 104 μm at 50 ps. Strong attenuation exists until 700 ps, although there is slight reduction in attenuation at the outer edges of the ablated region. The final photograph at infinity shows a damaged region of the same diameter as the attenuated region in the transient photographs.

The cause of the probe beam attenuation (darkening of the image) in the photographs needs to be discussed. The attenuation is present for laser fluences above and below the phase explosion threshold. Since it is present in the photographs below the threshold, it is not expected to be caused by a mass removal process at low laser fluences. The possible causes of the attenuation

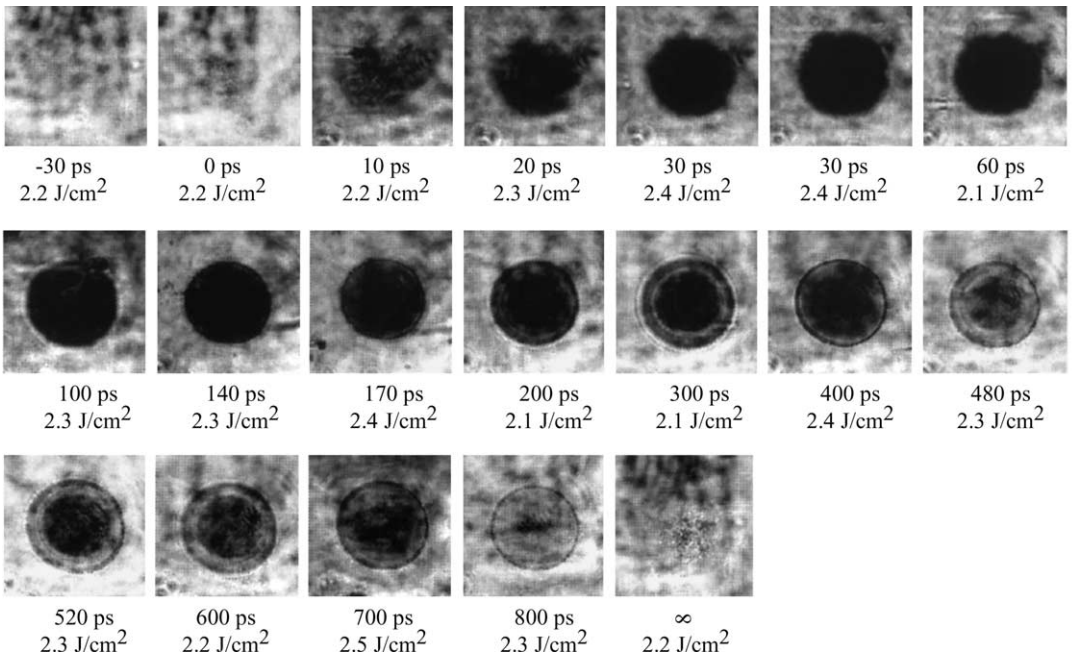

Fig. 3. Transient microphotographs of ablation at average fluence of 2.3 J/cm^2.

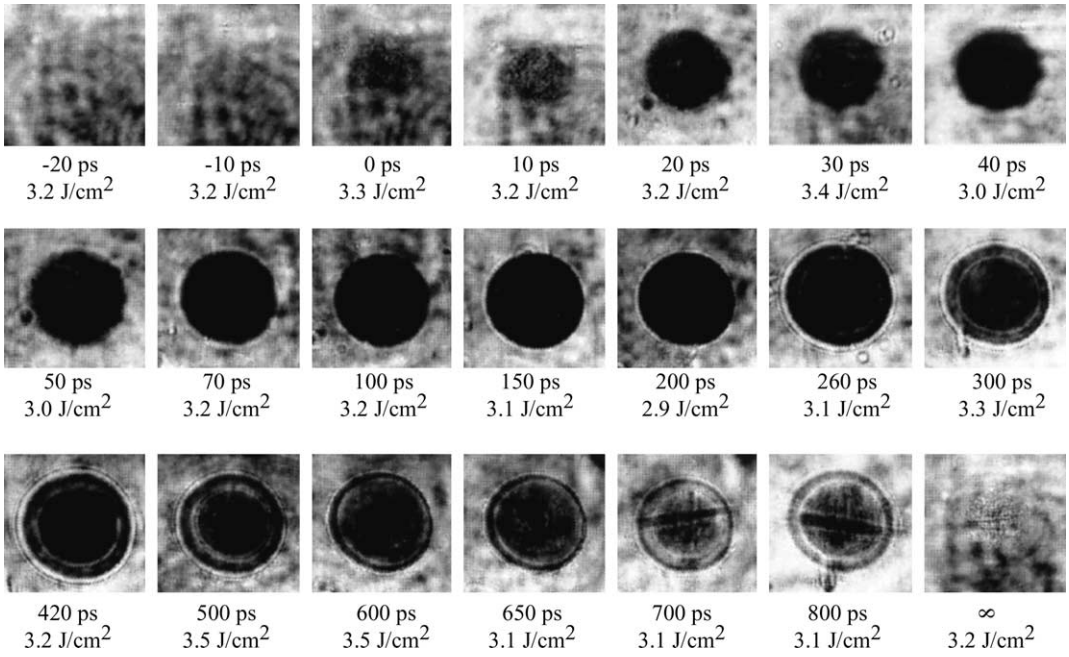

Fig. 4. Transient microphotographs of ablation at average fluence of 3.1 J/cm^2.

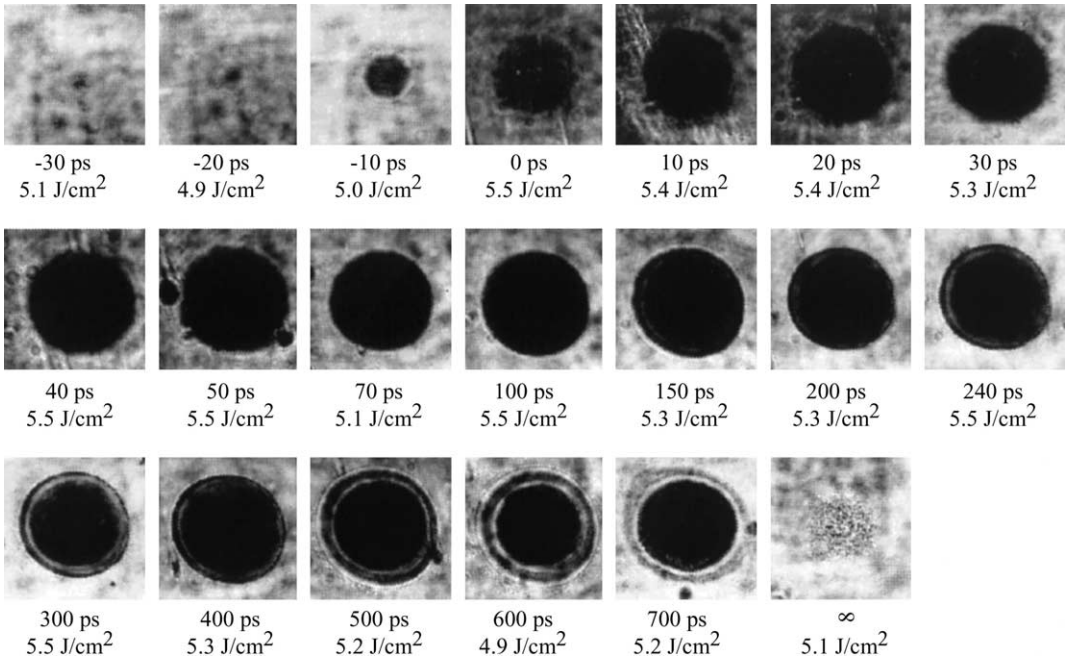

Fig. 5. Transient microphotographs of ablation at average fluence of 5.3 J/cm^2.

should indicate that the attenuation in all of the photos is a result of absorption or scattering processes that appear the same when imaged by the reflected probe beam. A very likely mechanism is the metal-to-dielectric transition when the surface temperature approaches the thermodynamic critical point. If the laser fluence, although below the threshold for phase explosion, is high enough for the surface to reach $0.8T_{cr}$, then large density fluctuations in the superheated liquid will result in a transition from a metal to a dielectric [2]. This transition to a dielectric material will result in increased transparency and scattering of the probe beam. It is expected that the reflectivity will be that of other dielectric materials, on the order of 10%. However, the target surface layer will no longer be a homogeneous media, as there will be many scattering centers, making the index of refraction and optical properties difficult to calculate and the reflectivity and transmissivity difficult to estimate. In addition to the scattering at $0.8T_{cr}$, if the laser fluence is high enough for the surface to reach $0.9T_{cr}$, then homogeneous nucleation will result in phase explosion, resulting in the ejection droplets of superheated liquid. Both of these processes will result in scattering and attenuation of the probe beam. On the other hand, the probing beam attenuation is seen as late as 800 ps at higher laser fluences, when the ablation process should have ended but the surface can still maintain a very high temperature. Therefore, the attenuation of the probing beam at this time indicates that the cause of attenuation is very likely to be the metal-to-dielectric transition.

4. Conclusions

This work presents time-resolved images of picosecond laser ablation of a metal, with picosecond time resolution. It was shown that attenuation of the reflected probe beam begins within the laser pulse duration and lasts for more than 800 ps. Possible causes of the change in surface reflectivity are scattering by fluctuations in the superheated liquid near $0.8T_{cr}$ and possibly also by scattering by ejected droplets near $0.9T_{cr}$. The high temperature state that sustains the metal-to-dielectric transition could last for hundreds of picoseconds after the laser pulse. Further studies are needed to distinguish between the metal–dielectric transition and phase explosion, so that to reveal the details of the laser ablation process.

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