Analysis of laser absorption on a rough metal surface

L. K. Ang, Y. Y. Lau,[a),b)] R. M. Gilgenbach,[b)] and H. L. Spindler
Department of Nuclear Engineering and Radiological Sciences, Intense Energy Beam Interaction Laboratory, The University of Michigan, Ann Arbor, Michigan 48109-2104

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We have developed a simple model to estimate the cumulative absorption coefficient of an ultraviolet laser pulse impinging on a pure metal, including the effects of surface roughness whose scale is much larger than the laser wavelength λ. The multiple reflections from the rough surface may increase the absorption coefficient over a pristine, flat surface by an order of magnitude. Thus, as much as 16% (at room temperature) of the power of a 248 nm KrF excimer laser pulse may be absorbed by an aluminum target. A comparison with experimental data is given. © *1997 American Institute of Physics.* [S0003-6951(97)00806-1]

The laser ablative technique is of considerable current interest for deposition of thin films.[1–3] Laser pulses are incident on metals or semiconductors to produce a plume of plasma and neutral atoms for deposition. The laser-solid interaction is crucial, particularly the amount of laser energy that is absorbed. In this letter, we use a simple model to evaluate the fraction of the laser light absorbed on a metal surface with large scale surface roughness. The depth and width of such surface roughness are much larger than the laser wavelength, and they are observed when a series of KrF excimer laser pulses (λ=248 nm, 40 ns, <1.2 J, 6.4 J/cm^2) is used to ablate a pure solid aluminum target.[1] The surface roughness then causes multiple reflection and multiple absorption of the laser light, and may enhance the absorption by an order of magnitude over that on a perfectly flat surface. By virtue of the large scale in the roughness, diffraction of light becomes unimportant.

Considerable work already exists on the formation of periodic surface structure on a wide variety of materials by an incident laser. These works concentrate mostly on a different regime from the present study. Specifically, microroughness of height, h, less than laser wavelength λ, and lateral wavelength of order λ, is usually considered. Such a regime is prevalent when the laser fluence is below the damage threshold, typically 0.12–0.55 J/cm^2,[4] beyond which permanent damage would occur. These periodic structures are caused by inhomogeneous energy deposition associated with the interference of incident beam with a surface scattered field.[5,6] Excitation of surface plasmon can produce anomalous absorption under high fluence irradiation,[7,8] and the laser-driven corrugation instability leads to a strong coupling when the critical depth reaches the pump laser wavelength ($\lambda \approx h$).

What we consider here, then, is the case where the surface has already suffered permanent damage. The scale of roughness, in both height and width, is much larger than the wavelength. We set aside the physical processes that lead to such large scale deformation, such as the positive feedback mechanism,[4] the slow decay[8] mechanism (due to surface tension) of laser-driven corrugation instability, and other mechanisms explored recently.[2] We attempt to establish a scaling law that accounts for the enhancement in the laser absorption when such large scale roughness is present. Thus, the estimates given here may also be applied to keyhole formation in the continuous high power laser welding.[9]

Let us first record the absorption coefficient in a single reflection. In response to the high frequency laser field, we treat solid aluminum as a lossy plasma with dielectric constant[10] (SI units)

$$\varepsilon = \varepsilon_0 \left[1 - \frac{\omega_p^2}{\omega^2} - i\Delta \right] \equiv \varepsilon_0 n^2, \tag{1}$$

where ω_p is the electron plasma frequency, ω is the frequency of the laser light, ε_0 is the free space permittivity, n is the index of refraction, and $\Delta(\Delta \ll 1)$ is the dielectric resistive loss term, given by

$$\Delta = (\omega\varepsilon_0/\sigma)(\omega_p/\omega)^4. \tag{2}$$

In Eq. (2), σ is the electrical conductivity of the metallic target. Equation (2) is valid when $\omega\varepsilon_0/\sigma \ll 1$, and $\omega_p/\omega > 1$; its derivation follows a standard procedure.[10] For our case, pure aluminum has $\sigma = 3.72\times 10^7$ $(\Omega\ \mathrm{m})^{-1}$ at room temperature, and $\omega = 7.6\times 10^{15}$ rad/s. Numerically, if we associate the electron plasma frequency with the surface plasmon energy (E_s)[11] in aluminum $(\hbar\omega_p = E_s)$, then we have, with $E_s = 10.3$ eV,[11] $\omega_p/\omega = 2.06$, $\omega\varepsilon_0/\sigma = 0.00181$, and $\Delta = 0.032$ (Ref. 12).

By matching the electric field and magnetic fields across the vacuum-metal interface,[13] we can calculate the absorption coefficient (in power),

$$A(\theta_i) = \frac{2\Delta \cos\theta_i}{(\omega_p/\omega)^2 \sqrt{(\omega_p/\omega)^2 - \cos^2\theta_i}}, \quad [s \ \text{polarization}] \tag{3}$$

and

$$A(\theta_i) = \frac{2\Delta \cos\theta_i}{(\omega_p/\omega)^2 \sqrt{(\omega_p/\omega)^2 - \cos^2\theta_i}} \times \left\{ \frac{(\omega_p/\omega)^2 - \cos 2\theta_i}{(\omega_p/\omega)^2 \cos^2\theta_i - \cos 2\theta_i} \right\} \quad [p \ \text{polarization}] \tag{4}$$

for $\Delta \ll 1$ and $\omega_p/\omega > 1$, where θ_i is the incident angle. Note that if $\Delta = 0$ there is no absorption $(A=0)$. This means that the incident wave is totally reflected even though the field inside the lossless ''dielectric'' decays exponentially with a

[a)]Electronic mail: yylau@umich.edu
[b)]Also at: Applied Physics Program, University of Michigan, Ann Arbor, MI 48109.

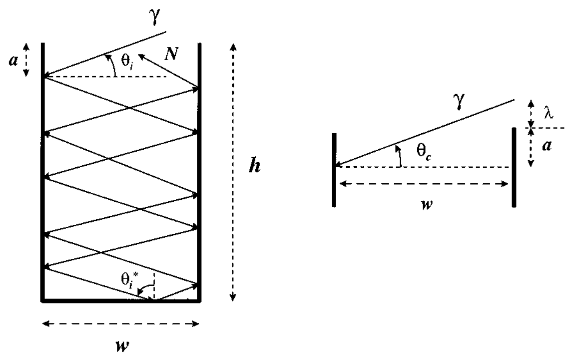

FIG. 1. Surface roughness representation with a rectangular well that has height h, and width w. The incident position is a, θ_i is the incident angle, and θ_c is the minimum incident angle that a photon can enter the well at position a.

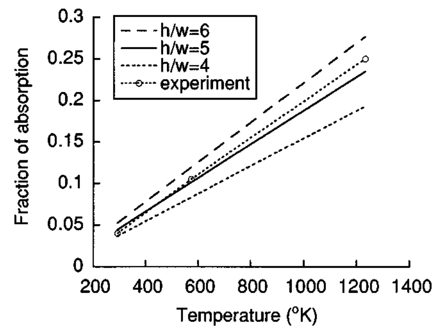

FIG. 3. Fraction of absorption by a 690 nm laser pulse impinging on solid silver (Ag) as a function of temperature (room temperature to melting temperature), for three different degrees of surface roughness [$\Omega=4.83$, $\mu=0.964$]. Experimental data were taken from Ref. 15.

scale of the ''plasma skin depth'' $\delta=(c/\omega)/[(\omega_p/\omega)^2-1]^{1/2}$, but no energy is dissipated within δ. Therefore, the laser energy is absorbed only through a nonzero resistive loss ($\Delta\neq 0$). From Eqs. (3) and (4), p polarization (with a large absorption coefficient) has a lower laser damage threshold than s polarization, in agreement with the experiment.[14] In this letter, we will use only s polarization formula [Eq. (3)]. Under normal incidence ($\theta_i=0$) condition, we have $A=8.51\times 10^{-3}$. Thus, less than one percent of laser light energy is absorbed in a single reflection. This energy is absorbed mostly within the plasma skin depth δ.

We now include the effects of the surface roughness that cause the laser light to undergo multiple reflections and, therefore, enhanced absorption. To calculate the cumulative absorption coefficient, A^*, we have to estimate the number of bounces of a photon on a rough surface. We model the rough surface with a distribution function of rectangular wells with height h, and width w. As shown in Fig. 1, a photon strikes the well at incident position a with incident angle θ_i, bounces back and forth between the walls, and finally leaves the well after N bounces, where N is a function of (h, w, a, θ_i):

$$N(h,w,a,\theta_i)=2+\frac{2h-a}{w}\cot\,\theta_i. \qquad (5)$$

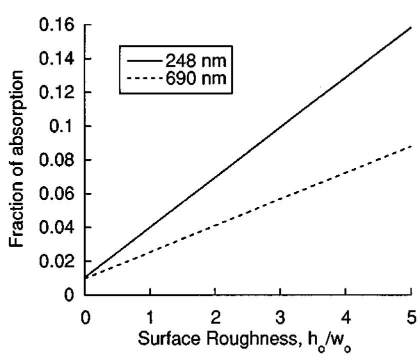

FIG. 2. Cumulative absorption coefficient, $\langle AN\rangle$, of two different wavelengths of laser radiation ($\lambda=248$, 690 nm) on a solid aluminum target, where h_0/w_0 is degree of surface roughness, $\Omega\equiv\omega_p/\omega$, $\mu=w_0/(\lambda^2+w_0^2)^{1/2}$ and $\alpha\equiv\omega\varepsilon_0/\sigma$ (at $T=20$ °C). [$\lambda=248$ nm: ($\Omega=2.06$, $\mu=0.995$, $\alpha=1.81\times 10^{-3}$) and $\lambda=690$ nm: ($\Omega=5.75$, $\mu=0.972$, $\alpha=6.5\times 10^{-4}$)].

The absorbed energy E_a, on the surface after N bounces can then be expressed as

$$E_a=\left\langle \sum_{j=1}^{N} A(\theta_i)[1-A(\theta_i)]^{j-1}E_0\right\rangle \equiv A^*E_0, \qquad (6)$$

where E_0 is the incident laser energy, and $\langle\ \rangle$ denotes the average over the distribution in (h, w, a, θ_i). Note that even at normal incidence, A is much less than one ($A=8.51\times 10^{-3}$), so we can approximately write $A^*\approx\langle A\cdot N\rangle$. Note also that all bounces in the well have the same incident angle, θ_i, except for the one which bounces off the base of the well. It has a different incident angle, θ_i^*, which can be related to θ_i by $\cos\theta_i^*=\sin\theta_i$. Thus, we can write AN as $A(\theta_i)[N(h,w,a,\theta_i)-1]+A(\theta_i^*)$. To calculate A^*, we average over the incident angle, incident position, and the distribution functions of height and width:

$$\langle AN\rangle=\frac{\int_0^\infty g(w)dw\int_0^\infty f(h)dh\int_0^h P(a)da\int_{\theta_c}^{\pi/2}\{AN\}d\theta_i}{\int_0^\infty g(w)dw\int_0^\infty f(h)dh\int_0^h P(a)da\int_{\theta_c}^{\pi/2}d\theta_i}, \qquad (7)$$

where $\theta_c\equiv\tan^{-1}[(a+\lambda)/w)]$ is approximately the minimum incident angle that the laser light can enter the well at position a, without suffering diffraction [Fig. 1], λ is laser wavelength, and $P(a)$ is the probability of laser light entering at position a. For simplicity, we assume that the laser light impacts on the top of the wells, so that $P(a)=\delta(a)$ where δ is the Dirac delta function. We further assume $\theta_c=\tan^{-1}[(a+\lambda)/w_0)]$, where w_0 is the characteristic width to be defined below. We next consider various distribution functions of h and w, characterized by indices m and n which measure the sharpness in the distributions:

$$f(h)=C_m h^m\exp(-h^2/\overline{h^2}), \qquad (8a)$$

$$g(w)=D_n w^n\exp(-w^2/\overline{w^2}). \qquad (8b)$$

Here, C_m and D_n are normalized so that $\int_0^\infty f(h)dh=1$ and $\int_0^\infty g(w)dw=1$, and ($\overline{h}$, $\overline{w}$) are related to the (h_0, w_0) by ($\overline{h}=h_0\sqrt{2/m}$, $\overline{w}=w_0\sqrt{2/n}$), with ($h_0$, w_0) defined as the value (h,w) at the peak of the distribution function. Under these assumptions, Eq. (7) gives

$$\langle AN\rangle = \frac{4\,\alpha\,\Omega^2}{\pi - 2\cos^{-1}\,\mu}\Bigg\{\ln\left|\frac{\Omega+1}{\sqrt{\Omega^2-\mu^2}+\sqrt{1-\mu^2}}\right| + \ln\left|\frac{\sqrt{\Omega^2-1+\mu^2}+\mu}{\sqrt{\Omega^2-1}}\right| + 2G_{mn}\frac{h_0}{w_0}\left[\frac{\ln|2(1-\Omega^{-2})/(1-\mu)|}{2\sqrt{\Omega^2-1}} - \sin^{-1}\left(\frac{\mu}{\Omega}\right)\right]\Bigg\}, \tag{9}$$

where

$$\Omega \equiv \omega_p/\omega > 1, \quad \mu \equiv \cos\,\theta_c = w_0/(\lambda^2+w_0^2)^{1/2},$$

$$\alpha \equiv \omega\varepsilon_0/\sigma,$$

$$G_{mn} = \sqrt{\frac{n}{m}}\left(\frac{\Gamma\left(\frac{m+2}{2}\right)\Gamma\left(\frac{n}{2}\right)}{\Gamma\left(\frac{m+1}{2}\right)\Gamma\left(\frac{n+1}{2}\right)}\right), \tag{10}$$

and $\Gamma(x)$ is the gamma function. For all $m \geqslant 1$, $n \geqslant 1$ and up to 30% accuracy, $G_{mn} \approx 1.3$. In the limit $m \to \infty$, $n \to \infty$, we obtain $f(h) \to \delta(h-h_0)$ and $g(w) \to \delta(w-w_0)$ from Eq. (8) and $G_{mn} \to 1$ from Eq. (10).

Equation (9) gives an estimate of the cumulative absorption coefficient, $\langle AN\rangle$ of a photon incident on a rough surface as a function of h_0/w_0, which can be defined as the degree of roughness. Note that the laser wavelength and material dependence of $\langle AN\rangle$ enter through the parameters Ω, μ, and α. Figure 2 shows two different wavelengths ($\lambda = 248$, 690 nm) of laser radiation on aluminum target at room temperature ($T = 20\,°C$), where w_0 is assigned to be 2.5 μm. As can be seen, with the same h_0/w_0, the aluminum target absorbs more power from 248 than 690 nm laser radiation. This figure shows that a rough surface may increase laser absorption by an order of magnitude over a flat surface. As a target is normally preheated to a higher temperature in laser deposition process, in Fig. 3 we compare the temperature dependence on the parameter $\sigma(T)$ of a 690 nm laser impinging on an Ag target with experimental value.[15] The increase of absorption with increasing temperature (up to melting temperature) has also been shown in the high-intensity optical radiation experiment.[16]

The above estimates were based on the classical theory of electrical resistivity applied to a much simplified model of rough surfaces. Validity of this model awaits further comparison with reflectivity measurements on a rough surface such as those displayed in Ref. 1. The simple formulas derived in this letter provide an immediate assessment of the relative importance of surface roughness on laser absorption, when the roughness scale is much larger than the laser wavelength.

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[1] H. L. Spindler, R. M. Gilgenbach, and J. S. Lash, Appl. Phys. Lett. **68**, 3245 (1996).
[2] S. I. Anisimov and V. A. Khokhlov, *Instabilities in Laser-Matter Interaction* (Chemical Rubber, Boca Raton, 1995).
[3] A. Peterlongo, A. Miotello, and R. Kelley, Phys. Rev. E **50**, 4716 (1994).
[4] J. F. Young, J. E. Sipe, and H. M. van Driel, Phys. Rev. B **30**, 2001 (1984).
[5] H. M. van Driel, J. E. Sipe, and J. F. Young, Phys. Rev. Lett. **49**, 1955 (1982).
[6] J. E. Sipe, J. F. Young, J. S. Preston, and H. M. van Driel, Phys. Rev. B **27**, 1141 (1983).
[7] S. R. J. Brueck and D. J. Ehrlich, Phys. Rev. Lett. **48**, 1678 (1982).
[8] F. Keilmann, Phys. Rev. Lett. **51**, 2097 (1983).
[9] A. Poueyo-Verwaerde, R. Fabbro, G. Deshors, A. M. de Frutos, and J. M. Orza, J. Appl. Phys. **74**, 5773 (1993).
[10] See, e.g., M. A. Lieberman and A. J. Lichtenberg, *Principles of Plasma Discharges and Materials Processing* (Wiley, New York, 1994); R. P. Feynman, *Lectures on Physics* (Addison-Wesley, Reading, MA, 1964), Vol. 2, pp. 32-10.
[11] See, e.g., Chap. 10 in C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1976).
[12] If instead we calculate ω_p by assuming that there is one free electron for each aluminum atom, as done in Feynamn [Ref. 10], we arrive at a similar value of $\omega_p/\omega = 1.82$. Feynman, in addition, suggested a method to evaluate Δ, with the result being given in Eq. (2) following his procedure. More serious are the effects of oxide layer and of polycrystallinity in the metal in the evaluation of Δ and ω_p.
[13] See, e.g., Chap. 7.3 in J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1990).
[14] J. F. Young, J. S. Preston, H. M. van Driel, and J. E. Sipe, Phys. Rev. B **27**, 1155 (1983).
[15] A. M. Bonch-Bruevich, Y. A. Imas, G. S. Romanov, M. N. Libenson, and L. N. Mal'tsev, Sov. Phys. Tech. Phys. **13**, 640 (1968).
[16] T. E. Zavecz and M. A. Saifi, Appl. Phys. Lett. **26**, 165 (1975).