Laser-Driven Corrugation Instability of Liquid Metal Surfaces

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During intense CO_2-laser irradiation deep corrugations build up on liquid metals such as Hg, In, Sn, Al, and Pb. Spacing, orientation, growth, and decay of the corrugations are studied, by visible light diffraction; support is found for a model of stimulated scattering where the incident light parametrically decays into both the surface corrugation and a surface plasmon. Thermal evaporation supplies the nonlinearity. The instability provides polarization-dependent absorption and can be expected in laser-metalworking and laser-plasma situations.

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Stimulated light scattering involving (electromagnetic) surface waves has recently been described.[1-3] In this process the free-space wave couples with a wave propagating along the surface (surface polariton[4]). Initially, surface imperfections serve as the scattering centers. By interference the total intensity attains a spatial modulation which causes the surface to deform. Subsequently the deformation itself contributes to the scattering and grows into a well-defined phase-matching corrugation structure. Rapid cooling after the irradiation can preserve the surface "ripples."[5] This Letter describes a time-resolved test of the model of "stimulated surface corrugation–surface wave scattering," at high laser intensity. Liquid metals were chosen as surface-polariton–active materials, thereby avoiding a solid-liquid boundary which complicates the typical laser melting situation.[1,3] Both the structure and the dynamics found are in support of the model.

The experiment simply uses a single-pulse transversely-excited-atmospheric CO_2-laser beam (polarized in the x direction) incident normally on the metal (the $-z$ direction). The near-fundamental-mode profile is collimated to a spot diameter of 1.7 mm. Longitudinal modes spaced at 46 MHz produce a spiky temporal shape. A visible probe beam with 0.9-mm diameter (polarized in the x direction) is shone on the sample at 0.5° angle of incidence. Its scattering measures the transient development and decay of any surface corrugation. Two observation modes were employed. The first used a dye-laser pulse of 3-ns duration (wavelength 605 nm, peak fluence 10 mJ/cm^2) and a translucent screen for taking photographs of the diffraction pattern. The second mode used a cw Kr^+ laser (wavelength 647 nm, peak intensity 30 W/cm^2) and fast-rise-time detectors (HP 5082) to monitor the evolution of the diffraction efficiency. No effects due to either probe beam alone were observable. In the second mode the use of 1.6° collecting apertures resulted in high sensitivity to plasma formation which occurred regularly with 80 mJ (but very seldom with 50 mJ) CO_2-laser "pump" energy.

Ripple formation on Hg was observed reproducibly when the pump energy exceeded about 30 mJ (i.e., peak fluence 2.4 J/cm^2), independent of the wavelength in the range 9.2 to 10.8 μm. The diffraction pattern consisted of up to a dozen concentric rings representing diffraction orders. The first two are displayed in Fig. 1. The half cone angle (approximately 3.5°) for the inner ring matches the ratio $\lambda_{probe}/\lambda_{pump}$ to better than 1%. Hence the surface is corrugated with a period equal to the pump wavelength. The nonuniform brightness along the circle shows that the corrugations run mostly orthogonal to the driving electric field. This tendency increases with higher pump power where furthermore a breaking up into isolated dots indicates the development of discrete corrugation packets. Similar findings were made with molten In, Sn, Al, and Pb, in laboratory atmosphere.

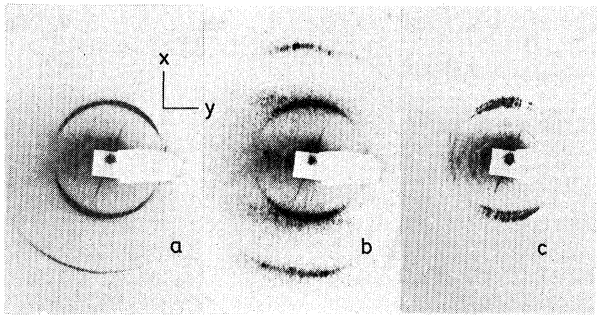


FIG. 1. Diffraction patterns of corrugated Hg surface, taken with visible probe pulse of 3-ns duration: (a), (b) immediately after the pump pulse, (c) 7 μs later. Pump wavelength 10.57 μm with fluence (a), (c) 4 J/cm^2 and (b) 5.6 J/cm^2.

Further time-resolved experiments were done with liquid Hg (in an evacuated cell), where readily available mechanical and optical data allow a quantitative test of the theory. Relative first- and zeroth-order intensities indicate a total scattering efficiency of at least 1% (note that the light square areas in Fig. 1 are from a $\times 30$ attenuator foil). The diffraction pattern remains visible for long times [Fig. 1(c)] with increasing blur indicating large-scale ($\gg 10$ μm) undulations. The diffraction is not observable before a considerable delay after the start of the pump pulse. I studied the onset phase with a 10-ns sampling interval [Fig. 2(a)]. The maximum occurs after about 700 ns when the pump power has dropped back to about 3% of its peak. Then the diffraction signal decays, with roughly a 3-μs exponential time constant [Fig. 2(b)] and strong modulation at a period $T \approx 1$ μs. This period is independent of the pump energy but scales with the $\frac{3}{2}$ power of the pump wavelength [Fig. 2(c)]. The oscillation can be understood easily if we assume that the surface corrugation decays into two counterpropagating capillary or surface Rayleigh waves.[6] These are in phase at the end of the pump pulse but cancel at further delays of $(n+\frac{1}{2})T$, with $n = 0,1,2,\ldots$, to give a momentarily flat surface with zero diffraction signal; T is half the capillary wave period. With the known dispersion $\omega^2 = k^3\sigma/\rho$ a measurement of T directly yields the capillary or corrugation wavelength Λ. in our experiment: $T = (\rho\Lambda^3/8\pi\sigma)^{1/2}$, where σ and ρ denote the surface tension and density of the metal. Such an analysis was performed [Fig. 2(c)], confirming that the corrugation period equals the pump wavelength.

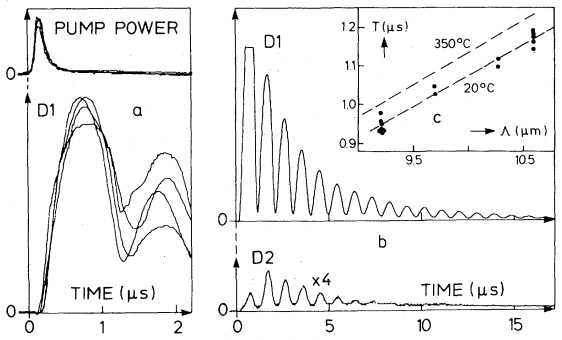


FIG. 2. Time evolution of 647-nm probe intensity diffracted from corrugated Hg surface, at pump wavelength 9.21 μm with fluence 4 J/cm^2: (a) first order (D1), four shots superimposed; (b) simultaneous first and second (D2) order. (c) Modulation period T of diffraction signals vs pump laser wavelength Λ (data points); the dashed lines represent dispersion characteristics of Hg capillary waves (T, half of the inverse frequency, versus Λ, wavelength), calculated for two temperatures.

Simple geometrical considerations account for the observed ripple spacing.[1] The pump field $E_L \exp[i(\omega t + k_L z)]$ has constant phase at the metal surface $z = 0$. By interference, a scattered surface wave $E_{sw}\exp[i(\omega t - k_{sw}x)]$, which propagates along x, leads to a standing intensity modulation $I_0 + \tilde{I}\cos(k_{sw}x)$ (with $\tilde{I} \sim E_L E_{sw}$). Its period is the wavelength of the surface wave. Surface corrugations are assumed to map directly this intensity modulation. Hence the equality of ripple spacing and laser wavelength means that the surface wave propagates at the speed of light. Such dispersionless behavior is expected for infrared surface plasmon polaritons[4] on metals (with plasma frequencies typically in the uv).

The crucial question arises of how the standing intensity pattern drives the buildup of the corrugation. There are two aspects, namely, the driving force itself and the pump scattering from the corrugation. The latter can be viewed as grating coupling[7] giving phase matching between the pump and the surface waves.[1] At normal incidence, two counterpropagating surface waves contribute. As long as the corrugation [assumed as $a\cos(k_{sw}x)$] has a small depth ($a \ll k_{sw}^{-1}$) the intensity fraction coupled into the surface waves is $I_{sw}/I_L = 2(ak_{sw}/2)^2$. The intensity modulation is $\tilde{I} \sim a$. Hence, growth of a increases $\tilde{I}$. Assume a simple linear relation $da/dt \sim \tilde{I}$ as in photochemical deposition.[2] The solution is exponential growth, $a \sim \exp(t/\tau)$. Such behavior can also be expected when a solid material vaporizes in proportion to the applied intensity. With heat conduction neglected the solid surface recedes with velocity $dz/dt = -(1-R)I_0/U$, where R is the reflectivity and U is the evaporation energy per unit volume, and thus $da/dt = (1-R)\tilde{I}/U$. Since $\tilde{I} \lesssim \sqrt{2}\,ak_{sw}I_L$ the rate constant becomes $\tau^{-1} \lesssim \sqrt{2}\,(1-R)k_{sw}I_L/U$. When we insert for liquid mercury $R = 0.85$, $U = 4\times10^3$ J/cm^3, and $k_{sw} = 2\pi/1000$ cm^{-1}, we obtain $\tau \gtrsim 110$ ns at $I_L = 3\times10^7$ W/cm^2 [peak intensity in the present experiment, Fig. 3(a)]. While the experimental observations are in accordance with the model's prediction of $a \sim \exp(t/\tau)$, we find $\tau \approx 45$ ns at $I_L = 3\times10^7$ W/cm^2, considerably faster than predicted from the simple evaporation picture above, thus indicating additional driving forces. These can be pressure effects. The surface tension decreases with temperature and thus leads to a

flow of material out of the high-intensity towards the low-intensity regions[8]; light pressure and ablation recoil both depress the surface in the high-intensity regions. These forces augment the action of the evaporation effect considered initially. Most likely, deviations from a simple exponential growth are introduced by the inertia of hydrodynamic response.

The visible diffraction signals yield further details. Minima above zero reveal an inhomogeneity, viz., a spatial variation in the starting time of the free decay of the corrugation. This is probably due to inhomogeneities in the pump beam. The envelope decay of the scattering signals comes from damping of the Rayleigh waves. The damping time is theoretically[6] $\tau_D = \rho\Lambda^2/8\pi^2\eta$, where η is the viscosity. With Hg and $\Lambda = 10$ μm, one obtains $\tau_D = 11$ μs at 20°C and $\tau_D = 16$ μs at 185°C. The observed decay is twice as fast, for a yet unknown reason. The depth of the corrugation can be estimated from the probe diffraction efficiencies d_1 (first order) and d_2 (second order). Scalar theory[7] predicts $d_1 = J_1^2(u)$ and $d_2 = J_2^2(u)$, where J denotes Bessel functions of the first kind, and $u = 4\pi a/\lambda$ for $\lambda \ll \Lambda$. For $u \lesssim 1$ one can approximate $d_1 \approx (2\pi a/\lambda)^2$ and $d_2 \approx 0.25(2\pi a/\lambda)^4$, and thus $d_2/d_1 \approx 0.25(2\pi a/\lambda)^2$. I observe $d_2/d_1 \approx \frac{1}{4}$ and thus have $2a \approx 0.2$ μm at the pump fluence of 5.6 J/cm² [Fig. 3(a)]. At such a corrugation depth, 10^{-3} of the pump power is coupled into the surface wave. With further increase of pump fluence the corrugation depth is expected to rise further. A critical depth is $2a \approx 2$ μm when full coupling between laser and surface wave is approached. Then a great part of the laser radiation, which is ordinarily reflected from the metal surface, becomes coupled to the surface wave and is totally absorbed. The transition has a thresholdlike character. Experimentally I find this to occur at about 6.5 J/cm², where strong plasma formation and splashlike disruption of the surface is obtained.

Quite remarkably the diffraction signals become nonsinusoidal at high pump power [Fig. 3(a)] indicating the appearance of spatial harmonics in the corrugation profile [as observed in a related experiment, Fig. 3(b)]. The absence of dispersive dephasing of the time harmonics in Fig. 3(a) points to a solitonic propagation.[9]

For practical applications the strong absorption by fully developed corrugations is of interest. Note that the instability occurs at arbitrary incidence angle.[1] The self-generated coupling structure can be viewed as *deus ex machina* to obtain near-unity efficiency in processing initially highly reflective materials. In the fusion case simulations show that light pressure (ponderomotive force) can suffice to ripple the critical density surface of the plasma.[10] In laser metalworking a steplike transition from a small (Fresnel) to near-unity process efficiency is observed at an intensity near 5×10^7 W/cm². Furthermore, the strong dependence on laser polarization found in the quality and attainable speed of steel cuts by CO_2-laser radiation indicates a role of stimulated surface plasmon scattering.

In summary, I have given evidence of the self-excitation of surface-plasmon coupling structures which develop on metal surfaces under high-intensity irradiation. The measurements of the spacing, orientation, and growth of these structures favor a model of "stimulated surface corrugation–surface plasmon scattering" which in this case encompasses an intimate interplay of thermal evaporation and surface plasmon excitation. Anomalously high absorption results when such a transient instability channels the applied laser power, via surface plasmons, into the target.

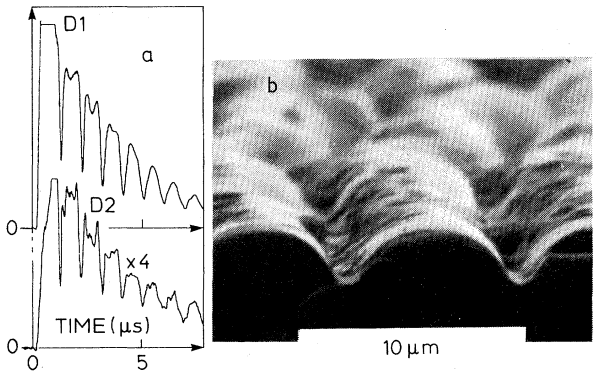


FIG. 3. (a) Diffraction signals as in Fig. 2(b), at pump fluence 5.6 J/cm². (b) Scanning electron micrograph of surface ripples frozen into CO_2-laser–melted quartz (Ref. 1), after fifty shots at 10.59-μm wavelength; viewing is from a slant angle of 2° above the surface.

[1]F. Keilmann and Y. H. Bai, in Proceedings of the CLEO Conference, Phoenix, 1982 (unpublished), Report No. WK5, and Appl. Phys. A 29, 9 (1982).

[2]S. R. J. Brueck and D. J. Ehrlich, in Proceedings of the CLEO Conference, Phoenix, 1982 (unpublished),

Report No. WK8, and Phys. Rev. Lett. 48, 1678 (1982); R. M. Osgood and D. J. Ehrlich, Opt. Lett. 7, 385 (1982).

[3]D. J. Ehrlich, S. R. J. Brueck, and J. Y. Tsao, Appl. Phys. Lett. 41, 630 (1982).

[4]For a recent review, see A. A. Maradudin, in *Festkörperprobleme*, edited by J. Treusch (Vieweg, Braunschweig, 1981), Vol. 21, pp. 62-116.

[5]Permanent surface rippling has been observed for a long time in laser-damage and in laser-annealing experiments: See Refs. 7–16 in Ref. 1; M. Birnbaum, J. Appl. Phys. 36, 3688 (1965); P. M. Fauchet and A. E. Siegman, Appl. Phys. Lett. 40, 824 (1982); H. M. v. Driel, J. E. Sipe, and J. F. Young, Phys. Rev. Lett. 49, 1955 (1982).

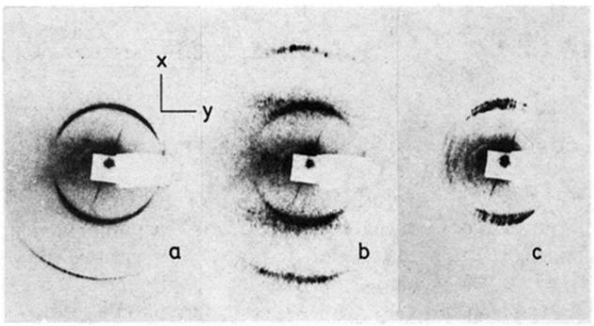
[6]L. D. Landau and E. M. Lifschitz, *Hydrodynamik* (Akademieverlag, Berlin, 1966).

[7]*The Electromagnetic Theory of Gratings*, edited by R. Petit, Topics in Current Physics Vol. 22 (Springer-Verlag, Berlin, 1980).

[8]J. C. Loulergue, P. Manneville, and Y. Pomeau, J. Phys. D 14, 1967 (1981).

[9]An exact solution of the nonlinear capillary wave problem, Crapper's wave, resembles the shape in Fig. 3(b); cf. B. Kinsman, *Wind Waves* (Prentice-Hall, Englewood Cliffs, N.J., 1965), p. 258. I thank H. C. Yuen (Redondo Beach) for this information.

[10]K. G. Estabrook, E. J. Valeo, and W. L. Kruer, Phys. Fluids 18, 1151 (1975).



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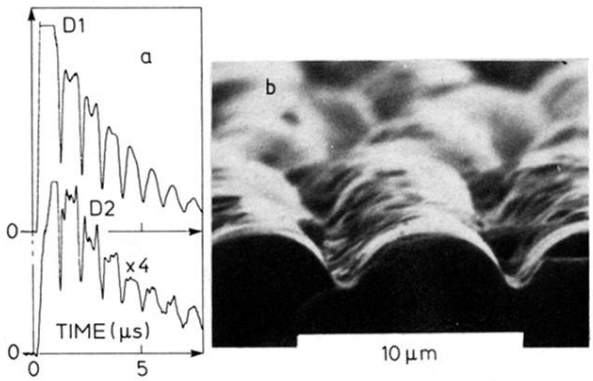

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