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Nonlinear excitation of capillary waves by the Marangoni motion induced with a modulated laser beam

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We have studied the amplitude and phase relationships of capillary waves produced by modulated laser light on the surface of strongly absorbing liquids. For increasing laser fluences a strong nonlinear behavior is observed. This is connected with the convective motion of the liquid created by surface tension gradients known as the Marangoni motion. The abrupt changes in both the capillary wave amplitude and the phase are found to be due to an autoblocking effect of the heat flux from the region of the laser absorption as the result of the development of a closed circulating flow of the liquid.

Due to the creation of surface tension gradients on a liquid surface a convective motion develops known as the Marangoni motion.[1] The gradients appear as the result of some action on the liquid, for instance, during heating. In a liquid layer heated from below a cellular convection develops[2] which was explained in terms of interfacial surface gradients.[3]

During the action of laser radiation on a surface of an absorbing liquid the heating from above also leads to the development of the Marangoni motion and results in surface deformations.[4]

In the present work we consider the action of a modulated laser beam on a liquid surface. The created average heating leads to the development of a convective motion and the modulation of the laser beam provides periodical variations of the thermodynamical parameters of the liquid which are accompanied by the excitation of surface oscillations having at small scales the form of capillary waves.[5] This response of the liquid surface allows probing the liquid parameters and the processes in the interaction region. The interest and significance of the phenomena considered are determined by their general nature for all the processes connected with the radiational heating of a liquid from the surface.

It is shown that the Marangoni motion influences significantly the whole process of the heat transfer from the heated domain. At well-defined conditions even the formation of a closed convective motion is possible. The liquid then circulates back to the laser spot, thereby strongly reducing and even counteracting the heat conduction away from the region of the laser radiation absorption. We have labeled this effect *autoblocking,* since it develops as a result of a self-regulation of the system. This effect manifests itself in a characteristic nonlinear behavior of the amplitude and phase dependence on the laser intensity providing a new mechanism for capillary wave generation.

The influence of the Marangoni motion on the thermal regime is important in different problems dealing with the interaction of laser radiation with absorbing liquids: in the laser diagnostics of liquids,[6] in the laser generation of capillary waves,[7] and during the melting of a solid surface subjected to the action of a cw laser radiation which is used, for instance, in the welding of materials.[8] The heating and deformation of the liquid surface produce a dynamical optical lens in the surface layer, influencing the penetration of the laser radiation into the liquid.[9]

In our experimental setup the beam of a single-mode Ar^+ laser at $\lambda = 514$ nm was on-off modulated by a stabilized chopper and directed at normal incidence on the liquid surface. This pump beam heats the surface layer. The inclination of the liquid surface during laser irradiation due to the developing liquid motion was measured at various fixed distances from the center of the pump spot using a He-Ne laser as a probe beam. The deflection of this probe beam was registered with a two-plate position-sensitive photodiode. The sensitivity of the registration scheme which had a linear relation with respect to inclinations of the liquid surface was typically 80 V/deg and was used to evaluate the signal voltage in terms of a surface inclination. The angular resolution achieved was about 10^{-5} deg and was limited mainly by external vibrational noise. The measured surface inclinations corresponded for a typical value of the wave number $k = 10$ cm^{-1} to the capillary wave amplitude $\zeta_0 \sim (\theta/k)$ which was in the range of 2 to 2×10^3 Å. The signal was observed in two ways, directly on an oscilloscope and separately for each harmonic with a lock-in amplifier. A set of calibrated neutral density filters was used to vary the pump laser intensity.

The phenomena described occur only in sufficiently strong absorbing liquids. In particular, the results presented below were obtained for a solution of the dye LDS 751 (Exciton) in ethylene glycol with a concentration of 1.25 g per 1000 cm^3. The optical absorption coefficient for the mixture was separately measured to be $\mu = 70$ cm^{-1}.

Figure 1(a) depicts the signal amplitude versus the laser intensity. A magnification of the initial part representing low pump laser intensities is shown in the inset. The data were registered a few seconds after the laser beam had been turned on so that a stationary situation had established itself.

Consider at first the data registered at a modulation frequency of $f = 40$ Hz. With an increase of the laser intensity a gradual yet sufficiently rapid increase in the

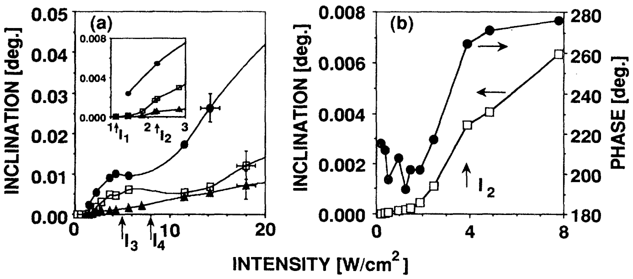

FIG. 1. (a) Signal amplitude versus the intensity of the laser radiation. The average surface deformation (●) and signals of the capillary wave excited at $f=40$ Hz (□) and at $f=60$ Hz (▲) are plotted. The thickness of the liquid layer is $h=14$ mm, the radius of the laser spot is $a=0.5$ mm, and the distance from the laser spot is $r=5$ mm. The arrows indicate particular values of intensity corresponding to the specific features observed. (b) Amplitude (□) and phase (●) of the first harmonic of the capillary wave versus the laser intensity for $f=40$ Hz, $h=14$ mm, $r=5.5$ mm, and $a=0.7$ mm.

slope of the dependence on laser intensity starts at $I_1 \approx 1.2$ W/cm^2 and occurs until a maximum of the slope is reached near the value $I_2=2.2$ W/cm^2. At I_2 a kink takes place and the amplitude increases with a smaller slope. Near the value $I_3=5$ W/cm^2 the curve reaches a local maximum followed by a small decrease in the amplitude. With a further increase of the laser intensity $I > I_4 = 8$ W/cm^2 a gradual approximately linear growth of the amplitude proceeds. At a modulation frequency of $f=60$ Hz the dependence is smoother; however, the kink at the intensity I_2 is still noticeable.

The interval of intensities around the value I_2 was examined in more detail by simultaneously registering the amplitude and the phase of the first harmonic of the capillary wave signal. The results are compiled in Fig. 1(b). A steep increase of the amplitude occurs simultaneously with an abrupt increase of phase. A change in the slope of the dependence is again observable. The value of the intensity I_2 corresponding to the kink is somewhat higher than in Fig. 1(a), which is due to the wider laser spot used. It should be noted that in the interval of intensities between 0.5 W/cm$^2 < I < 2$ W/cm^2 the signal was slightly unstable, a fact which is exhibited in the phase variations observed.

For both experiments we were able to visibly observe the liquid motion in the vicinity of the laser irradiation spot. For this purpose, micron-sized particles were poured on the liquid surface. The particles were quickly carried away from the center to the peripheral area of the laser spot. The characteristic speed of the particles was typically about 1 cm/sec.

The results obtained indicate a new nonlinear mechanism for capillary wave excitation which cannot be ascribed to a thermal nonlinearity due to a temperature dependence of the parameters of the liquid, nor to an evaporational mechanism, nor to an explosive boiling.[10] This follows since in our case the heating of the liquid is relatively small. In previous experiments with the liquid surface deformation due to the laser-driven Marangoni motion[2,11] typical intensities were about 1 W/cm^2 and the heating[11] of the liquid was only about 1 °C. Therefore we present in the following another interpretation of the results of our measurements. It is based on a model which includes the Marangoni effect and takes the pulsations of the liquid motion due to the formation of a closed circulation of the liquid into account.

The qualitative picture of the process is depicted in Fig. 2. The increase of the temperature T of the liquid due to the absorption of the laser radiation in the volume V, further referred to as the *active region,* leads to the creation of tangential forces on the surface proportional to the derivative $(\partial\sigma/\partial T)$ of the surface tension σ on temperature T. These forces initiate a convective liquid motion which is impeded by the viscous forces proportional to the shear viscosity coefficient η and the velocity v of the liquid. This process is approximately described by the following nonlinear system of two bounded averaged kinetic equations:

$$CV(\partial T/\partial t)=P-S_1\kappa(T-T_b)/l_T+T_bCvS-TCvS \; , \tag{1}$$

$$\rho V(\partial v/\partial t)=-\eta S_1(v/l_v)+(\partial\sigma/\partial T)Tl_a \; . \tag{2}$$

In these equations the heat capacity of the liquid per unit volume is C, the power of the absorbed laser radiation is P, and the heat conductivity of the liquid is κ. The active volume is $V=S_1 l_T$, with $S_1=\pi a^2$ being the cross section of the pump laser spot. The side area of the active region is $S=2\pi a l_T$ where the characteristic length l_T of the vertical temperature variation in the liquid flow counteracting the thermal conductivity is determined by the inverse value of the optical absorption coefficient μ as $l_T=1/\mu$. Other parameters are the density ρ of the liquid and the length $l_a=2\pi a$ which is the perimeter of the active region. The length l_v is the characteristic distance over which the velocity v decreases in the direction perpendicular to its flow. We also assume that $l_v=\mu^{-1}$.

A substantial feature of our model is that together with a major temperature increase in the active region there is

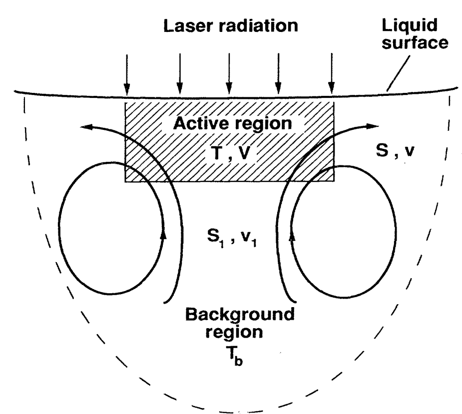

FIG. 2. Illustration of the proposed theoretical model.

also a smaller temperature increase T_b in the surrounding domain. This larger domain which surrounds the active region is therefore labeled the *background region.* For large optical absorption coefficients it is just the radius a of the laser beam which determines the spatial extent of the stationary temperature distribution in the laser heating of motionless matter.[12] We assume $\mu a \gg 1$, and therefore that the thickness of the active region is much less than that of the background region. It is also assumed that in the background region the variations of the temperature occur more slowly than in the active region.

The cross section of a stream tube passing through the active region will rapidly increase with distance, since a stream in a liquid originating from a small source experiences a divergence close to a spherical one. Consequently, the velocity of the liquid will rapidly decrease outside of the active region. We represent the power of the laser radiation, the velocity of the liquid, and its temperature in the active region as the sums of the time-averaged components and the alternating components caused by the modulation of the laser beam, i.e., $P=P_0+\tilde{P}$, $v=v_0+\tilde{v}$, and $T=T_0+\tilde{T}$. It is concluded that the pressure pulsations acting in the liquid surrounding the active region are on the order of $\tilde{p}=pv_0\tilde{v}$. This follows from Bernoulli's equation for the average pressure difference $p-p_0=\rho v^2/2$, where p_0 and p are the pressures in the stream tube inside and outside the active region. In this way the deformation of the surface is proportional to the average pressure difference.

Next it is assumed that the time of the velocity relaxation $\tau_v=\rho V l_v/\eta S_1$ is relatively small and that the oscillating components change harmonically with angular frequency ω and also that $|\tilde{T}| \ll T_0$ and $|\tilde{v}| \ll v_0$ hold. For the stationary regime we obtain then the averaged components

$$v_{0,S}=NT_{0,S}, \quad T_{0,S}=\alpha_0/(\Delta+\beta/2) , \tag{3}$$

where $N=2|\partial\sigma/\partial T|/(\eta\mu a)$, $\alpha_0=\mu I_0/C$, I_0 is the intensity of the laser radiation, $\beta=\mu^2\chi(I-\mathrm{Ma}/K)$, $\chi=k/C$, $\Delta=(\alpha_0\gamma+\beta^2/4)^{1/2}$, and $\gamma=2N/a$. In this problem the heat transfer from the active region into the depth of a liquid plays a significant role which is described by the coefficient β. It depends on the dimensionless Marangoni number $\mathrm{Ma}=|\partial\sigma/\partial T|T_b a/(\eta\chi)$ and the dimensionless geometrical factor $K=(\mu a)^3/4$ and characterizes the regime of heat transfer normal to the surface.

Similarly, we find for the stationary complex amplitudes of the oscillating components

$$\tilde{v}_a=N\tilde{T}_a, \quad \tilde{T}_a=\tilde{\alpha}\exp(i\varphi)/(\omega^2+\tau^{-2})^{1/2} , \tag{4}$$

where $\tilde{\alpha}=m\alpha_0$, with the modulation index $m=0.64$ for the first harmonic and for abrupt on-off modulation. Also we set $\varphi=\tan^{-1}(\omega\tau)$ and $\tau=(\beta+2\gamma T_{0,S})^{-1}$.

Surface perturbations are proportional to the variations of pressure. Therefore, summing all the expressions presented we obtain the stationary average deformation of the liquid surface ζ_0 and the amplitude of the surface capillary wave ζ_a as a function of the laser intensity

$$\zeta_0 \propto v_{0,S}^2, \quad \tilde{\zeta}_a \propto v_{0,S}\tilde{v}_a . \tag{5}$$

Now we have to take into account that with increasing laser intensity the temperature in the background region T_b also grows. When the convective motion is taken into account the estimate based on equations similar to Eqs. (1) and (2) gives for the temperature increase

$$T_b(^\circ\mathrm{C})=(I_0\eta/|\partial\sigma/\partial T|C)^{1/2}=[I_0(\mathrm{W/cm^2})/0.54]^{1/2} . \tag{6}$$

For a comparison of our model with the experiment we use the parameters for ethylene glycol at 25 °C and the experimentally measured values: $C=2.9$ J/cm^3 deg), $\chi=1.2$ cm^2/sec, $\partial\sigma/\partial T=0.13$ erg/(cm^2 deg), $\eta=0.7$ g/(cm sec), and $\sigma=54$ erg/cm^2. Using Eq. (3) we obtain for the velocity of the liquid motion $v_{0,S}\sim 2$ cm/sec and for the temperature increase $T_{0,S}\sim 2$ °C at the intensity $I_0=1$ W/cm^2 which is compatible with our observations.

The increase of the temperature T_b leads to a decrease of the coefficient β in Eqs. (3) and (4) which at $\mathrm{Ma}=K$ becomes zero. This is accompanied by an increase of the slope of the amplitude-on-intensity dependence in Fig. 1. The value of $\beta=0$ means that an autoblocking of the thermal flux occurs to the depth of the liquid by the developing liquid motion. This leads to an abrupt temperature increase in the active region and consequently according to Eqs. (3)–(5) to the rapid increase in the amplitudes of the average surface deformation and the generated capillary wave. This process implies that some of the stream tubes of the liquid motion are locked in the immediate vicinity of the active region. According to Eqs. (3) and (6) autoblocking, i.e., $\beta=0$, occurs at the intensities $I_1=1$ W/cm^2 and $I_2=4$ W/cm^2 for the conditions of Figs. 1(a) and 1(b). The corresponding experimentally observed values are $I_2=2.2$ W/cm^2 and $I_2=4$ W/cm^2 and are reasonably close to the calculated ones.

With a further increase of the laser intensity the transition to negative values of the coefficient β is possible. It is pointed out that large negative values cannot be produced since this means that the active region receives more energy from the background region than it gives to it. This is impossible for a stationary process. However, the system will maintain in some interval of laser intensities such a value of the temperature T_b as to provide a small negative coefficient β close to zero. With increasing laser intensity this is only possible if more stream tubes become unlocked. Thus, in the unlocking, the feedback loop gets broken. As a consequence the corresponding value of the coefficient β starts to increase, which leads in the proper intensity interval to a slowing down and even a decrease in the growth of the temperature and therefore the amplitudes of the surface perturbations. The qualitative behavior of the coefficient β is shown in Fig. 3(a). In this figure the value of β $(I=0)$ at low intensities is chosen to be equal to $\beta_0=\mu^2\chi$ and the characteristic values I_1, I_2, I_3, and I_4 are equal to those where the distinct features in the measurement were observed.

The amplitude dependences which were calculated for the frequencies $f=40$ and 60 Hz using Eqs. (3)–(5) and the dependence $\beta(I)$ presented in Fig. 3(a) are shown in Fig. 3(b). The calculated curves demonstrate the same specific features as observed in the experiment. Thus the

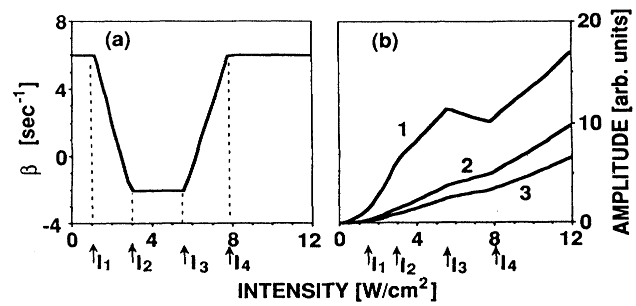

FIG. 3. (a) Intensity dependence of the heat transfer coefficient β. (b) Calculated amplitude of the surface perturbations for the average surface inclination (curve 1), and the amplitudes of the capillary wave at $f=40$ Hz (curve 2) and $f=60$ Hz (curve 3).

observable features in the amplitude dependence on laser intensity are directly connected to the special features in the process of the heat transfer reflected in the coefficient β. The change of phase in the capillary wave motion is due to the radial redistribution of the pressure pulsations after the formation of the closed circulating liquid motion. Indeed the maximum of the pressure pulsations is realized at some distance from the center of the laser spot depending on the divergence of the stream tubes. When the stream tubes are not locked this maximum lies farther from the center as compared to the case when a localized circulating motion in the vicinity of the laser spot has already formed. This spacial shift is order of magnitude wise equal to the radius a of the laser beam. Therefore the closing of the Marangoni motion is accompanied by the phase shift of the capillary wave signal $\Delta\varphi \sim ka$, where k is the wave number of the capillary wave. For the measurement represented in Fig. 1(b) the wave number corresponding to the frequency $f=40$ Hz is $k=10\ \text{cm}^{-1}$. In these estimates the influence of gravity and of the bottom of the container were taken into account.[13] Therefore, the closing of the stream tubes in the vicinity of the intensity will lead to a phase variation on the order of $\Delta\varphi \sim 60°$ which is slightly smaller but compatible with the phase variation observed in the experiment.

In summary, we have observed that the generation of capillary waves is strongly influenced by the Marangoni motion. The model developed is in good qualitative agreement with the experimental data. The observed abrupt increase in the capillary wave amplitude and the change of the capillary wave phase at a particular laser intensity were explained as a manifestation of the development of a closed convective motion of the liquid leading at the same time to an autoblocking effect of the thermal conductivity. Thus, the characteristics of the excited capillary waves reflect the processes in the interaction region enabling their diagnostics.

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