Dynamics of absorption in a laser-produced crack

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The radiation absorption dynamics in a growing laser-produced crack is investigated experimentally and theoretically, and an expression is derived for the rate of gas evolution in the crack as a function of the angle of incidence and intensity of the radiation. The growth rate is determined by the parameter σ/H, where σ is the absorption cross section of the gas molecules in the crack and H is the effective energy required to form a single gas molecule.

Damage to transparent polymers caused by intense laser pulses of length ~ 1 ms is known to be accompanied by the formation of disk-shaped cracks.[1,2] The damage process can be divided into three stages: 1) thermochemical instability develops at absorbing microinclusions, and a cavity of diameter 10–20 μm forms which is filled with damaged polymer material[3,4]; 2) the cavities rupture to form microcracks of length $\sim 100\,\mu$m (Ref. 5); 3) the laser crack then grows due to the "wedging action" of the gas formed by vaporization of the polymer at the phase interface.[6,7] The present paper is concerned with an experimental and theoretical study of the third stage, for which we have found it convenient (both for recording and for interpreting the data) to consider the energy absorbed by the growing crack.

To make some preliminary estimates, we note that in our experiments an average laser energy of 0.5 J was needed to form a single crack. Some of this energy was used to break down the polymer into gaseous products, while the rest heated the material near the crack or did work against the elastic forces. Roughly

$$\Delta Q_T \sim \varkappa T S_{cr} (\chi \Delta t)^{-1/2} \Delta t$$

of the absorbed energy was removed from the crack by heat conduction; here $\varkappa$ is the thermal conductivity of the polymer, $(\chi\Delta t)^{1/2}$ is the thickness of the layer heated by the crack, and T, S_{cr}, and Δt are the gas temperature in the crack and the crack area and growth times, respectively. For $T \approx 800$ K, $S_{cr} \approx 10^{-1}$ cm^2, and $\Delta t \approx 100\,\mu$m, we have $\Delta Q_T \approx 0.05$ J. We can estimate the work done by the gas against elastic and binding forces as $\Delta Q_p \sim PV$, where P is the gas pressure in the crack, $V \sim S_{cr} h$, and h is the width of the crack. Since $P \approx 10^2$ atm and $h \approx 3\,\mu$m (Refs. 8, 9), the work done by the gas accounts for only a very small fraction of the absorbed energy ($\sim 10^{-3}$ J). Thus, during laser cracking almost all of the absorbed light energy is used to break the polymer chains in the gas phase. This conclusion suggests the following simple model. If we let H be the energy required to form a single gas particle, then the formula

$$dN/dt = I S_{cr} \alpha h f(\theta)/H \qquad (1)$$

should give the rate at which gas particles are formed in a growing crack. Here I is the intensity of the laser light, S_{cr} is the instantaneous area of the crack, $\alpha = \sigma N/V$ is the absorption coefficient for the gas in the crack, and the factor $f(\theta)$ depends on the crack orientation. For small cracks, we readily find from (1) that

$$N(t) = N_0 \exp(t/\tau), \quad \tau = [\sigma I f(\theta)/H]^{-1}, \qquad (2)$$

if the optical thickness is αh.

The amount of vaporized polymer, and hence also the absorption of the laser light, thus grows exponentially with time during the early stage of cracking. The ratio σ/H, where σ is the effective absorption cross section of the vaporized polymer and H is the effective energy defined above, is a characteristic of the polymer and determines the cracking rate.

We will next find how the growth time τ depends on the angle of incidence of the light. If we model the crack as a plane-parallel gas layer of thickness h, then we can write

$$\alpha h f(\theta) = \cos\theta (1 - |r|^2 - |t_t|^2)$$

for the fraction of the laser energy absorbed per unit area of crack; here θ is the incident angle of the light on the plane of the crack and r and t_t are the amplitude coefficients of reflection and transmission of the layer, which acts like a Fabry-Perot interferometer. For a thin layer such that $|(\omega/c) h (\varepsilon_0)^{1/2} \cos\theta_0| \ll 1$, where ε_0 and θ_0 are the permittivity and incident angle inside the layer, the familiar formulas for Fabry-Perot interferometers[10] yield the result

$$f(\theta) = 1 + \frac{1}{2}\left(\left| \frac{\varepsilon}{\varepsilon_0} \right|^2 - 1 \right) \sin^2\theta - \frac{\alpha h}{2}\left(1 + \cos^4\theta + \left| \frac{\varepsilon}{\varepsilon_0} \right|^4 \sin^4\theta \right) \Big/ \cos\theta \qquad (3)$$

for depolarized radiation to first order in $h/\lambda \ll 1$. Here ε is the permittivity of the polymer surrounding the crack. For $\alpha h/\cos\theta \ll 1$ the function $f(\theta)$ increases monotonically with θ, so that cracks should grow faster for obliquely incident light. However, as θ approaches θ_{TIR}, the angle for total internal reflection, the energy absorption should drop abruptly due to the decreased transmission of the steeply inclined boundary.

Analogous results were derived in Ref. 11 for the energy absorbed by optically thick cracks ($h \geqslant \lambda$). Among other things, it was shown there that the specific absorption (the fraction of energy absorbed per unit area) is inversely proportional to

$$\left| 1 - r^2 \exp\left(2i \frac{\omega}{c} h \cos\theta_0 - \frac{\alpha h}{\cos\theta_0} \right) \right|^2,$$

which oscillates as a function of the crack thickness h. These oscillations are caused by destructive interference and by an

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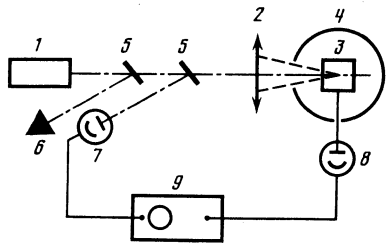

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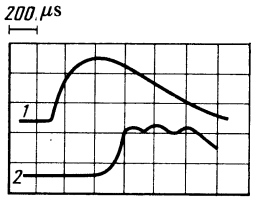

b

FIG. 1. a) Sketch of experimental apparatus. b) Typical trace, recorded by a storage oscilloscope: 1) incident radiation pulse; 2) difference signal.

increase in the amplitude of the radiation wave inside the plane-parallel layer.

The predictions of this model regarding the absorption dynamics of laser cracks may be summarized as follows. Initially, the absorbed energy rises exponentially, and the growth rate increases with I, θ, and σ/H. Later, when the crack has grown beyond the irradiated region, the absorbed energy oscillates with time due to the dependence on the crack width h.

Figure 1a shows a sketch of the experimental system. Light from a free-running neodymium laser 1 (energy 5 J, pulse length 2 ms, beam divergence 10^{-2} rad) was focused by a lens 2 into a polymethylmethacrylate (PMMA) polymer 3. The PMMA was placed inside an integrating photometer 4 (Ref. 12), which measured the intensity of the light scattered by the polymer. The plates 5 diverted part of the laser beam to an energy meter 6 and photocell 7. The radiation emerging from the photometer was recorded by another photocell 8, and the signals from 7 and 8 were fed to the input of a differential amplifier in the storage oscilloscope 9. Since the two signals were identical when no polymer was present in the photometer, the trace of the difference signal when the photometer was loaded was proportional to the energy absorbed in the polymer. Figure 1b shows a typical trace. We see that the absorbed energy does indeed increase nearly exponentially during the early stage, after which pronounced oscillations develop.

The dark circles in Fig. 2 show the experimental dependence of the growth time τ in PMMA as a function of the incident angle θ of the laser beam on the crack for a fixed laser energy. We see that τ (θ) is closely approximated by the dependence $f(\theta)$ in (3), which is also shown. Figure 3 plots the experimental dependence τ (I^{-1}), where I^{-1} is the reciprocal of the laser energy. As predicted by (2), $\tau(I^{-1})$ is linear; its slope yields the estimate $\sigma/H = 0.28 \cdot 10^{-7}$ cm^2/erg.

The oscillations found in the experiments were used to

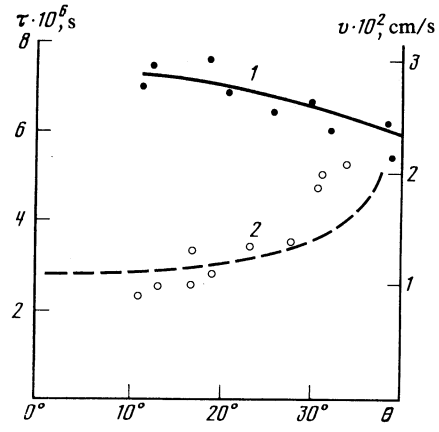

FIG. 2. Absorption growth time τ (●) and crack velocity v (○) as functions of the incident angle of the radiation: 1) calculated from (3); 2) specific energy absorption from Ref. 11.

deduce the velocity (widening) v of the crack during an oscillation period Δt_{osc}:

$$dh/dt = \Delta h/\Delta t_{\text{osc}} \approx \lambda/2 \cos\theta_0 \Delta t_{\text{osc}}.$$

$dh/dt = v$ remained roughly constant for $0 \leqslant t \leqslant 300\,\mu s$, after which it decreased slowly. Figure 2 shows $v(\theta)$ for constant laser power; we see that $v(\theta)$ is even steeper than the angular dependence of the specific energy absorption in the crack,[11] and v rises abruptly as θ approaches θ_{TIR}. The velocities v deduced from Fig. 2 for typical growth times $3 \cdot 10^{-4}$ s give final values $h \approx 5\mu$m, in agreement with the experimentally measured width.[13] This suggests that the oscillations in the absorbed energy are due to interference.

We have thus studied the time behavior of the energy absorbed during the growth of a laser crack and found that the mass of vaporized polymer increases exponentially during the initial stage. For optically thick cracks, the absorbed energy varies quasiperiodically due to interference effects in the absorbing gas layer. The absorption growth time τ and crack velocity v were analyzed as functions of the incident angle of the laser beam on the crack plane, and the experimental results are accurately accounted for by the proposed model. We have found that for transparent polymers, the parameter σ/H determines the growth rate and can be used

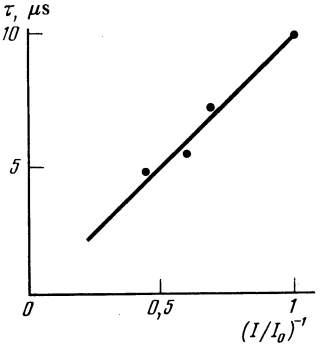

FIG. 3. Growth time τ as a function of the reciprocal $(I/I_0)^{-1}$ of the normalized intensity.

to predict if cracking will occur for various laser pulse lengths and intensities. For example, the value $\sigma/H = 0.28 \cdot 10^{-7}$ cm^2/erg found above for PMMA indicates that a giant pulse ($t_p \sim 10^{-8}$ s) of intensity $I \approx 3.6 \cdot 10^8$ W/cm^2 (well above threshold) will not cause cracking in PMMA, because $\tau \leqslant t_p$. This explains why short pulses do not cause cracking in polymers.

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