Surface laser damage thresholds determined by photoacoustic deflection

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The technique of intensity dependent photoacoustic probe beam deflection has been applied to the determination of surface damage thresholds. We take advantage of an unambiguous correlation between the degree of laser damage and the energy in the generated acoustic pulse. The high sensitivity of this method, cross checked by measuring scatter losses in reflection, is independent of any surface optical properties. As an example for optical materials, damage thresholds for MgF_2 and CaF_2 have been determined to be about 1.4 GW/cm^2, and for LiF to be about 0.2 GW/cm^2.

The increasing use of high-power lasers in technical applications and fundamental research has brought about the problem of optical damage, in particular on surfaces. This has initiated extensive research in recent years.[1] Nevertheless, the situation remains unsatisfactory since the results of damage threshold measurements performed at different laboratories do not necessarily agree. In fact, they may differ by an order of magnitude or more.[2–5] Hence there is still a need for a reliable method to determine damage thresholds *in situ*, which is not achieved by the common technique of microscopic inspection, e.g., using Nomarski micrographs.[6] The drawback is overcome when the scatter of an auxiliary HeNe probe laser beam, directed onto the interaction spot, is used as an indication of damage.[7] Up to now this seems to be agreed on as being the most sensitive method.[6] It is not universally applicable, however. For example, it cannot be used for strongly scattering surfaces.

Rosencwaig and Willis[8] developed a method of observing the occurrence of laser damage by transducer detection of damage-generated acoustic waves inside the sample. It is well known that in dielectric breakdown a shock wave is generated,[9] traveling in the opposite direction to the laser beam[10] and leading to a modulation of the refractive index of the surrounding medium. This can be monitored optically by virtue of the associated deflection of a probe beam passing perpendicular to the shock wave.[10–13] Since particle emission from the surface has been observed already at laser intensities far below the onset of macroscopically visible damage,[14–16] we became interested in the dependence of acoustic pulses on laser intensity right around the damage threshold. We expected the energy contained in these pulses to be related to the degree of damage at the surface. In this letter we show that probe beam deflection for measuring the acoustic pulse energy as a function of incident laser intensity yields a reliable *in situ* determination of damage thresholds, applicable to any kind of surface and material.

In order to detect the acoustic pulse, a single-mode HeNe probe laser beam runs parallel to the surface a few mm apart and passes, at a certain distance, a 40 μm slit which only transmits a portion of the beam profile around the steepest slope, as sketched in Fig. 1. A photodetector behind this slit measures the change of the transmitted probe beam intensity, which is directly related to the deflection of the beam in either direction. In order to compare our measurements with the scatter method, a chopped second HeNe laser is focused under 45° incident angle onto the interaction spot and monitored in reflection under a restricted solid angle, using lock-in detection. A change in reflected intensity is then taken as an indication of scatter.

Damage is produced by a Nd:YAG pumped pulsed dye laser (wavelength 530 nm, pulse width 3 ns, pulse energy a few mJ) which is focused onto the sample mounted on a motor-driven *x-y* positioning stage. The relative laser intensity is computer controlled by a combination of a polarization-rotating Fresnel rhomb and a linear polarizer, so as not to change the spatial beam characteristics when varying the intensity. In order to obtain the power density, the spatial laser profile was measured by a burn pattern method at the position of the sample. Idle reflections of the beam are used for each laser shot to monitor both the temporal pulse shape (with subnanosecond resolution) and the pulse energy, the latter using a pyroelectric detector calibrated against a calorimeter at the sample site. All signals are digitally recorded shot by shot and then processed by a personal computer.

A typical signal caused by the deflection of the probe beam is shown in Fig. 2. As proof that this signal indeed originates from an acoustic pulse, we have measured the time interval between dye laser pulse and signal as a function of distance between surface and probe laser beam. The result is displayed in Figs. 3(a) and 3(b) for two different laser intensities. From these data we have derived the pulse velocity as a function of distance from the sample [Figs. 3(c) and 3(d)]. The fact that the pulse travels at large distances with

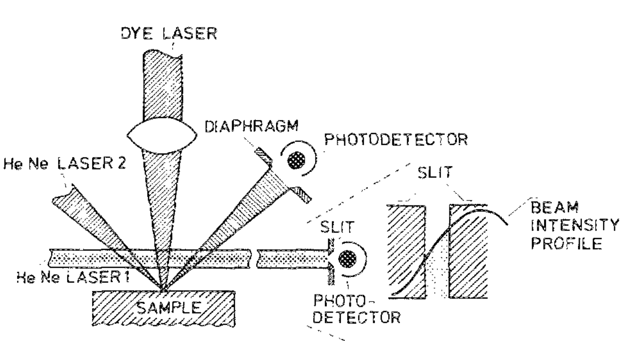

FIG. 1. Principal scheme of the experimental arrangement. The inset to the right shows the intensity distribution of the probe laser relative to the detection slit.

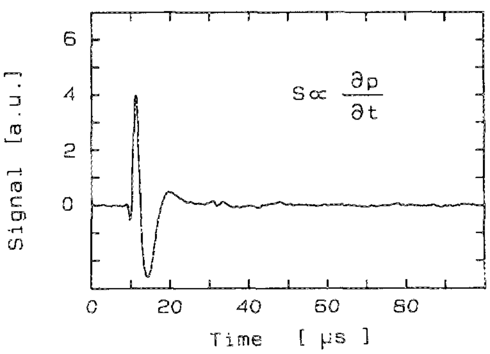

FIG. 2. Typical photoacoustic deflection signal from CaF_2 for a single laser pulse of 530 nm and an intensity above the damage threshold.

the normal speed of sound confirms that we are dealing with an acoustic phenomenon. Close to the surface, a shock wave is observed. From Figs. 3(c) and 3(d) it is evident that its range spreads out with increasing laser intensity, in agreement with previous reports.[17,18]

The amplitude S of the photoacoustic signal outside the shock wave range is proportional to the gradient of the refractive index[19] generated by the acoustic pulse and hence to the derivative of the pressure p with respect to the direction z perpendicular to the surface. This is equivalent to the time derivative divided by the speed of sound v:

$$S \propto \frac{\partial p}{\partial z} = -\frac{1}{v}\frac{\partial p}{\partial t}. \tag{1}$$

In order to obtain the energy E contained in the acoustic pulse, we have to consider the time integrated intensity I

$$E = \int I\, dt \tag{2}$$

with

$$I = [(c_p/c_v)p_m\rho]^{-1/2}(\Delta p)^2. \tag{3}$$

Here, $\Delta p = p - p_m$ is the deviation from the mean pressure p_m and ρ is the density of air. Consequently, we have to numerically integrate our measured signal twice in time in order to obtain the energy of the acoustic pulse:

$$E \propto \int dt \left(\int_0^t S\, dt'\right)^2. \tag{4}$$

The span over which the inner integration in Eq. (4) is performed extends from 8 to 30 μs (cf. Fig. 2). The integral returns to zero after the acoustic pulse has completely crossed the probe beam, which confirms the strictly linear response of the entire detection system.

That the obtained photoacoustic energy is indeed correlated to the degree of damage is proved in Fig. 4. Here E, defined by Eq. (4), is plotted as a function of the damage spot diameter, measured by microscopic inspection. The data obey approximately a third power law, indicating a proportionality to the volume of material blown away.

As an example, the technique was applied to determine damage thresholds of polished windows of calciumfluoride, magnesiumfluoride, and lithiumfluoride, at a wavelength of 530 nm. For each sample the intensity dependence of the acoustic signal was measured by evaluating more than 500 shots on virgin sites of the crystal (1-on-1 method). In order to avoid systematic errors, the laser intensity was varied randomly from pulse to pulse. The intensity range was subdivided into 50 intervals, and the corresponding photoacoustic signals were averaged and then integrated, according to Eq. (4). Typical sets of data are shown in Fig. 5 denoted by stars. Simultaneously, the results of the parallel scatter measurements are displayed as circles. For each material there is a distinct onset of the acoustic signal at a certain laser intensity which more or less coincides with a change in reflectivity. Even though there is significant noise in the acoustic data below that onset, the slope is sufficiently steep to define the damage threshold unambiguously. Except for MgF_2, the acoustic signal sets in at somewhat lower laser intensities

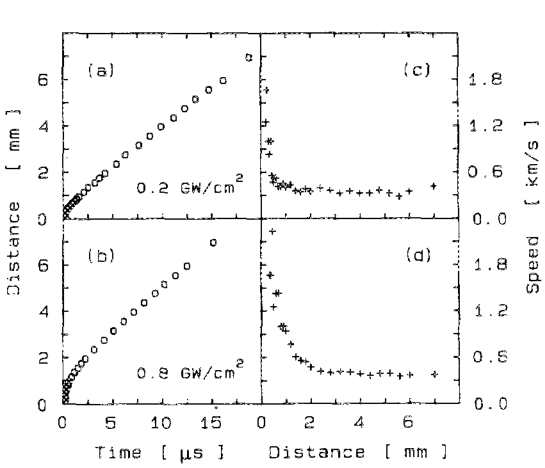

FIG. 3. Propagation of the acoustic pulse generated by 530 nm radiation on cleaved BaF_2 for two different laser intensities. (a),(b) Delay time of the deflection signals with respect to the laser pulse as a function of distance between probe beam and surface. (c),(d) Speed of the acoustic pulse as a function of distance from the surface.

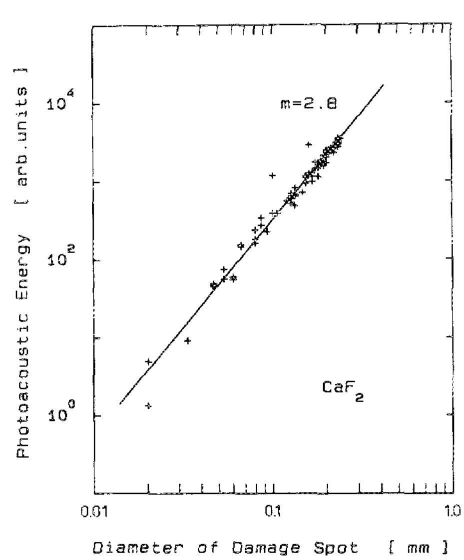

FIG. 4. Double-logarithmic representation of the acoustic energy [Eq. (4)] as a function of the diameter of visible damage spots on polished CaF_2 surfaces. The solid line of slope $m = 2.8$ is a least-squares fit to the data.

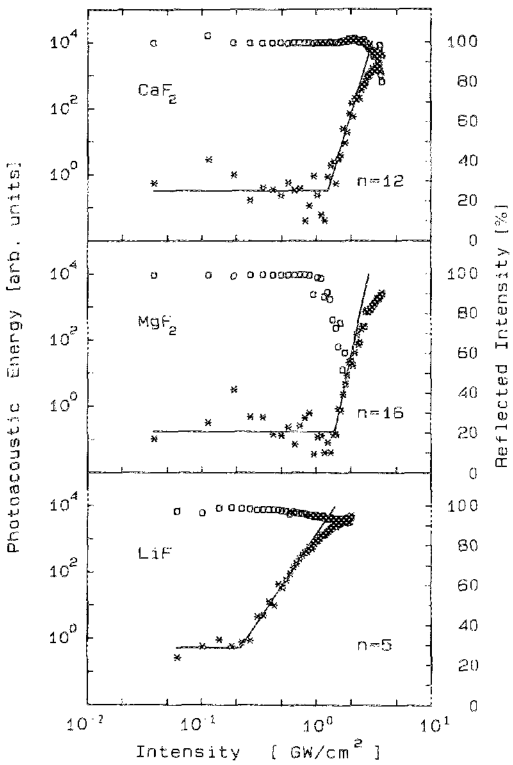

FIG. 5. Acoustic energy as a function of incident laser intensity at 530 nm from polished surfaces of three different materials. The solid lines of slopes n are least squares fits to the data in a selected intensity range.

than the attenuation of the scatter probe beam, indicating that the technique presented here may be somewhat more sensitive than the scatter method.

The damage thresholds obtained from the data in Fig. 5 are about 1.4 GW/cm^2 for MgF_2 and CaF_2, consistent with previous investigations.[2-5] For LiF we find a considerably lower damage threshold of 0.2 GW/cm^2, similar to what is usually observed for alkali halides. It should be noted, however, that due to difficulties in measuring the focal properties of the damaging laser beam, there may be an uncertainty of up to 25% in our absolute intensity scale.

In conclusion, we have shown that the intensity-dependent photoacoustic deflection is a sensitive technique for reliable *in situ* determination of damage thresholds. In principle, our data must contain information about the basic mechanisms of laser surface damage. Up to now, however, the actual origin of the acoustic signal near the threshold is not fully understood. The result for LiF in Fig. 5 indicates that multiphoton absorption across the optical gap may be the fundamental mechanism, similar to findings in ultrahigh vacuum experiments.[15,16,20] On the other hand, the much steeper slopes for the alkaline earth halides point toward avalanche gas-breakdown[21] near the surface. The macroscopic damage observed on the surface could then be the result of plasma sputtering.[22]

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