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Thresholds for dielectric breakdown in laser-irradiated diamond

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We report on picosecond laser-induced damage experiments that were carried out on a natural type-IIa diamond and a thick specimen of high-quality chemically vapor-deposited (CVD) diamond. In conjunction with earlier measurements performed elsewhere on an "optically thick" single crystal, it is shown that for spot sizes (2ω) ranging from 3 to 60 μm, the breakdown field strength (E_{BD}) at the damage threshold of diamond obeys a pattern best described as follows: $E_{BD} \simeq A/\sqrt{2\omega}$, where $A = 30.7$ and 38.7 MV$\mu^{1/2}$/cm at 532 and 1064 nm, respectively. The case of CVD diamond demonstrates that if problems arising from localized high absorption at the deposition surface can be avoided, this material should be of much promise for contemplated high-power free-electron laser window applications.

Diamond has optical and thermal properties that are highly attractive from the point of view of designing windows for high-power lasers such as free-electron lasers operating in the visible or the near infrared (IR).[1] For this reason, it is essential to perform laser-damage experiments on diamond because a proper understanding of the damage mechanism will allow the designer to set upper bounds for the tolerable peak power as well as the tolerable peak irradiance. In this regard, it is interesting that, 15 years ago, Liu, Yen, and Bloembergen[2] concluded that pulse-induced damage in diamond, at wavelengths of 532 and 1064 nm, originates from an "intrinsic" breakdown process at threshold field strengths comparable to those of other wide-band-gap materials, but that the critical power for self-focusing is relative low, which makes diamond "unsuitable" as a high-power laser-window material. More recently, it has been argued[3] that since diamond has an exceptional figure of merit for resistance to thermal stress [$R' = \sigma_f(1-\nu)k/(\alpha E)$], this material should be a "good choice" as an optical window material that can withstand laser damage. The results of damage experiments,[3] however, which were performed on chemically vapor-deposited (CVD) free-standing diamond films at the same wavelengths as in Ref. 2 but much larger spot sizes, turned out inconclusive in the sense that damage was not the result of dielectric breakdown but of surface ablation caused by high linear absorption associated with significant defect or disorder in the films. In this letter, we report on laser-induced breakdown experiments that were carried out under strictly identical conditions on a single-crystal type-IIa natural diamond and a thick specimen of high-quality CVD diamond; we will also reexamine the data of Liu *et al.*[3] and attempt to relate their work to our measurements through spot-size scaling, thus providing an initial database for bulk damage thresholds of diamond in the highly transparent regime.

Our experiments were performed on a modified Z-scan apparatus[4] that uses a Q-switched, mode-locked Nd:YAG oscillator to create picosecond-duration light pulses at the 1064 nm fundamental wavelength. The $\lambda = 532$ nm harmonic was generated by passing the beam through a 2-cm-thick KD^*P angle-tuned crystal. Spatial profiles in the focal interaction region were recorded by pinhole beam scanning while pulse widths were measured by means of a second-order autocorrelation technique. Focal intensity "packets" had Gaussian characteristics, i.e.,

$$I(r,t) = (P/A)\exp(-r^2/\omega_0^2)\exp(-t^2/\tau^2), \qquad (1)$$

where P is the incident peak power on axis and A is the nominal beam-waist area ($A = \pi\omega_0^2$); note that throughout this letter (see Table I), spot sizes represent the full width at the $1/e$ maximum in irradiance (FW1/eM$=2\omega_0$), whereas pulse durations are quoted as full width at half-maximum (FWHM), or $t_p = \tau\sqrt{\pi}$. Two diamond specimens were investigated: a single-crystal sample of type-IIa diamond obtained from a commercial supplier[5] and a CVD diamond sample of outstanding optical quality[6] made and polished at Raytheon Company. Regarding CVD diamond, it should be kept in mind that this material is polycrystalline and contains grain boundaries that are more absorbing than perfect diamond. Furthermore, it is now well established that the grain size strongly increases as the deposition progresses, which implies that, in a massive CVD diamond, the layers closest to the deposition surface do not match the crystalline quality of bulk material or the growth surface. To assess the resistance to pulsed laser radiation, each specimen was positioned at the beam waist, and its on-axis transmittance was recorded as a function of the incident peak power, for the two wavelengths of interest. The onset of irreversible laser damage induces beam scatter, which manifests itself as a decrease of the in-line transmittance, that is, through small axial apertures; as illustrated in Fig. 1, the criterion we used to define the damage threshold corresponds to a 5% reduction in transmittance. These thresholds are as listed in Table I, which immediately tells us that the laser hardness of high-quality CVD diamond almost matches that of natural diamond, considering that the inherent uncertainty of our measurements is of the order of 20%. Damage first occurs at the exit surface and exhibits morphological features that are indicative of a dielectric breakdown mechanism. With

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TABLE I. Key data relating to picosecond laser-damage experiments performed on natural type-IIa diamond single crystals and a specimen of chemically vapor deposited (CVD) polycrystalline diamond.

Specimen identification	DD-IIa[a]	DD-IIa[a]	CVD[a]	CVD[a]	DK-IIa[b]	DK-IIa[b]
Laser wavelength (nm)	1064	532	1064	532	1064	532
Pulse duration (ps)	32	23	32	23	30	30
Spot size in air (μm)	52	34	52	34	7.84	4.32
Specimen thickness (mm)	0.75	0.75	0.76	0.76	0.94	0.94
Rayleigh range (mm)	9.54	8.26	9.54	8.26	0.22	0.13
Incident peak power (kW)[c]	1810	310	1600	260	548	174
Enhancement factor (1)[d]	4	4	4	4	1.29	2.22
Internal peak irradiance (GW/cm^2)[c]	280	110	250	95	1220	2170
Effective spot size (μm)	52	34	52	34	6.97	3.00

[a]Experiments conducted at CREOL/University of Central Florida (this work).
[b]Experiments conducted at Gordon McKay Laboratory/Harvard University (Ref. 2).
[c]At the damage threshold; estimated uncertainty ~25% in irradiance.
[d]Caused by back-face reflection (CREOL) or beam self-focusing (Harvard).

CVD diamond, however, it was observed that in a configuration where the laser beam impacts the deposition surface, the onset of damage occurs at a substantially lower power level and is caused by front surface graphitization/ablation presumably due to highly localized absorption; this "thermal" failure mode is of no concern in the context of the present investigation.

The fractured appearance of the damage pit, at the exit surface, points to a subsurface field-induced breakdown mechanism[7] of the same nature as frequently observed when a high-power laser pulse passes at normal incidence through a transparent dielectric medium. The damage asymmetry between entrance and exit surfaces can be explained by considering the electric field amplitudes in the vicinity of the two surfaces, taking Fresnel reflections into account. Specifically, Boling[8] explains that as the pulse-induced plasma density increases, a standing wave is formed right at the back interface, which gives rise to internal electric fields that can become twice as large as normally anticipated. It is this large antinodal field that causes the electron-avalanche triggered subsurface "explosion,"

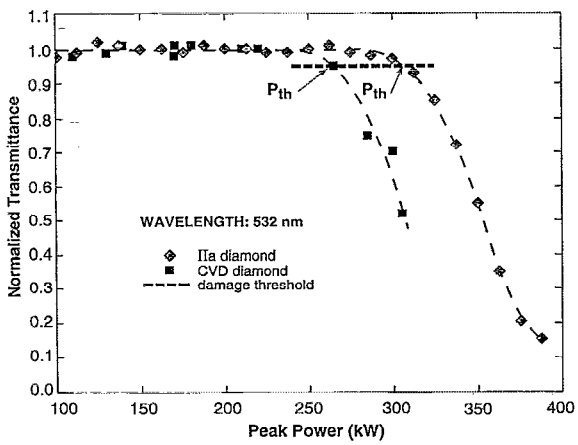

FIG. 1. Normalized in-line transmittance of two laser-irradiated diamond samples as a function of the incident pulse peak power, at the frequency-doubled Nd:YAG wavelength. Critical laser-pulse parameters are as given in Table I. It is postulated that irreversible damage occurs at an input power P_{th} that degrades the transmittance by 5%.

thus suggesting that the peak irradiance at the threshold for dielectric breakdown should be

$$(I_p)_{th}=\gamma(1-\mathscr{R})P_{th}/A, \tag{2}$$

if γ designates the irradiance enhancement factor, and $\mathscr{R}=(n-1)^2/(n+1)^2$ is the entrance surface reflectivity. With $n=2.39$ and 2.42 for $\lambda=1064$ and 532 nm, respectively,[9] we estimate that in our experiments the threshold peak irradiances reached levels as given in Table I; that is, in the 0.1–0.3 TW/cm^2 range, CVD diamond exhibiting perhaps 10% or 15% less resistance than single crystals. At this point, it should be emphasized (see Table I) that both our specimens were much thinner than the Rayleigh range ($Z_R=k\omega_0^2$, where k is the propagation constant), and hence that our experiments were conducted in an "optically thin" geometry, which rules out any self-focusing effect.

This, however, is not the case for the experiments described in Ref. 2, which made use of pulses of duration comparable to our own but much more tightly focused, thus creating nearly "optically thick" conditions in terms of target thickness vs Rayleigh range (see Table I, specimen DK-IIa). Since the nonlinear refractive index of diamond is known to be positive in the wavelength range of interest ($n_2\simeq 2.3\times10^{-13}$ esu at 1064 nm and $n_2\simeq 4.0\times10^{-13}$ esu at 532 nm, in the picosecond time frame[10]), it follows that self-focusing must be taken into consideration if the task on hand requires a correct evaluation of the internal irradiance distribution. In this context, and based on Marburger's theory of self-focusing,[11] it has recently been shown[12] that, if I_0 and z_0 refer to peak intensity and focal position in the absence of self-focusing, the axial intensity variation as a function of the distance z from the entrance plane can be expressed in the following manner:

$$\frac{I(z)}{I_0}=\left[\left(1-\frac{z}{R}\right)^2\left(\frac{\omega(0)}{\omega_0}\right)^2+\left(\frac{z}{Z_R}\right)^2\left(1-\frac{(1-\mathscr{R})P}{P_2}\right)\left(\frac{\omega_0}{\omega(0)}\right)^2\right]^{-1}, \tag{3}$$

where $R=z_0[(Z_R/z_0)^2+1]$ specifies the position of the geometric prefocus, $\omega(0)=\omega_0\sqrt{(z_0/Z_R)^2+1}$ is the beam ra-

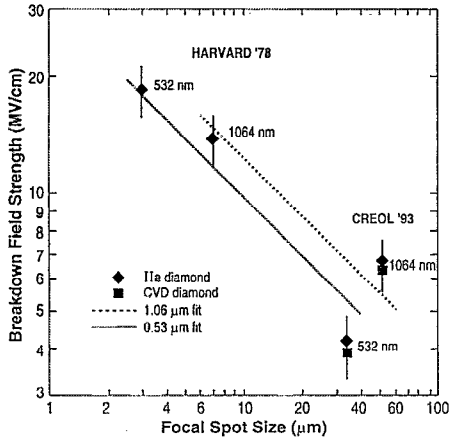

FIG. 2. Breakdown rms field strength of diamond at laser wavelengths of 532 and 1064 nm, based on experiments performed at Harvard (Ref. 2) and CREOL (this work). Critical laser-pulse parameters, including peak power and peak irradiances at the damage threshold, are as given in Table I. Note the fairly strong dependence on focal spot sizes, which, for the sake of clarity, is emphasized here through fitting to a Bettis-type scaling law.

dius at the entrance, and $P_2=3.77\ P_{cr}$ represents the second critical power in the sense that the first critical power, or minimum power required for catastrophic self-focusing, is given by the expression

$$P_{cr}=c\lambda^2/(32\pi^2 n_2). \tag{4}$$

Similarly, the positional dependence of the root-mean-square (rms) power radius of an initially Gaussian beam distorted by self-focusing can be obtained from

$$\left(\frac{\omega_{rms}(z)}{\omega(0)}\right)^2=\left(1-\frac{z}{R}\right)^2+\frac{z^2}{k^2[\omega(0)]^4}\left(1-\frac{P(1-\mathscr{R})}{P_3}\right), \tag{5}$$

if P_3 is set equal to $4P_{cr}$. Since, according to Ref. 2, the "center" of the diamond sample was placed at the beam waist, which means $z_0\simeq 470\ \mu$m, these equations yield self-focusing enhancement factors and effective spot sizes as listed in Table I. At 1064 nm, Eq. (2) then indicates a peak irradiance of 1.22 TW/cm^2 at the damage threshold, which is substantially less than reported by Liu *et al.*[2] [$(I_p)_{th}=2.93$ TW/cm^2], primarily because these authors assumed a nonlinear index ($n_2=7.2\times10^{-13}$ esu) derived from three-wave mixing experiments that may not be applicable for assessing the effect of picosecond duration pulses.

Laser-induced damage in normally transparent materials reflects a dielectric breakdown process that is best described in terms of the rms electric field strength, E_{BD}, which relates to the peak internal irradiance at the damage threshold, $(I_p)_{th}$, through the Poynting relation[13]:

$$E_{BD}=[(I_p)_{th}/(cn\epsilon_0)]^{1/2}=[(I_p)_{th}z_0/n]^{1/2}, \tag{6}$$

where c is the speed of light, ϵ_0 is the free-space permittivity, and Z_0 is the free-space impedance. Figure 2 displays our results in the form of a breakdown field strength vs focal spot size plot, which assumes that pulsewidth variations (the pulse duration ranges from 23 to 32 ps) are of little consequence considering that the Bettis scaling law (Ref. 14) $E_{BD}\propto t_p^{-1/4}$, probably holds since picosecond pulses satisfy the applicable thermal diffusion time constraint. Furthermore, if we accept the premise[14] that breakdown field strengths vary inversely as the square root of the focal spot size at a fixed laser frequency and a constant pulse duration, it is seen that in a first approximation, the diamond data generated at Harvard and CREOL combine to yield the dependencies exhibited in Fig. 2, i.e., $E_{BD}\simeq 30.7\times 1/\sqrt{2\omega}$ at 532 nm and $E_{BD}\simeq 38.7\times 1/\sqrt{2\omega}$ at 1064 nm. Consequently, for spot sizes of 4.5 μm, the breakdown field of diamond at laser wavelengths in the green and pulse widths in the picosecond range should be about 14.5 MV/cm, which turns out to be surprisingly close to the breakdown thresholds of KH_2PO_4, SiO_2, CaF_2, and LiF under similar conditions.[15] In the near IR, the breakdown field of diamond is approximately 25% higher, which again appears to be compatible with observations reported in the literature[13,15] and points to the availability of across-the-gap three-photon absorptions to assist the avalanche ionization process, if and when the laser frequency obeys the condition $h\nu > \mathscr{E}_G/3$. Since multiphoton absorptions of order $m>3$ are very unlikely, in other words, since intrinsic multiphoton absorption cannot provide a credible "channel" for initiating laser-induced breakdowns at 1064 nm, we conclude that the case of diamond substantiates the notion[15] that a theory of laser damage based on electron avalanche ionization must postulate the availability of "seed" electrons originating from extrinsic sources, i.e., nitrogen impurities or sp^2 carbon in the case of natural or CVD diamond, respectively. Experiments on single-crystal diamonds other than type-IIa and/or synthetic diamonds of smaller grain size may provide valuable information in this regard.

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