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AUTHOR(S) SIDNEY SINGER

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Los Alamos National Laboratory
Los Alamos, New Mexico 87545

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Diamond: A high power optical material

Sidney Singer

Defense Laser Technology, MS F617
Los Alamos National Laboratory
Los Alamos, New Mexico 87545

ABSTRACT Single-crystal chemically/isotopically pure synthetic diamond operated at low temperature offers an unusual opportunity for a small transparent optic that may transmit as much as tens to hundreds of megawatts of light, even in the presence of high peak power in the light beam. This property stems from the probable very low absorption of light in such material and the nature of the phonon energy transport at low temperatures.

1. Introduction

Although diamond has been used for a remarkable variety of purposes, the rarity and expense of suitable material has in the past limited its use in practical optical applications. Now that relatively usable and affordable material is available, and rapid progress is being made in the low pressure deposition of diamond and diamond-like films, important advances in solid optics, optical coatings, semiconductor electronics, and thermal engineering seem possible. In this paper, we will review the relevant properties of diamond, show how they might be utilized to permit the damage-free transmission of very powerful optical beams, and indicate how low temperature operation and the use of isotopically pure materials can enhance the power handling capability of diamond. Finally, we will address the issues associated with the propagation of high peak power optical pulses.

2. Properties of diamond

Table I shows some relevant properties of (mostly natural) diamond.

TABLE I. PROPERTIES OF DIAMOND

Density	3.51 g/cm ³
Refractive index	2.417 (@ 589 nm)
Expansion coefficient	8×10^{-7} deg ⁻¹ (300° K)
	~ 0 (80° K)
Dielectric constant	5.7
Thermal conductivity	
300° K	20 watt/(cm-deg)
80° K	100 - 150 watt/(cm-deg)
80° K synthetic (extrapolated)	1000 watt/(cm-deg) [chemically, isotopically pure]
Transparency	225 - 2500 nm; >6000 nm
Absorption	Type IIA: ~0.01 - 0.001 per cm Synthetic: < 0.0001 per cm (est)
Typical impurities	N, B, Al, Ni, Fe

Several important properties merit further discussion: the thermal conductivity and the transparency.

2.1 Thermal conductivity^{1,2} In a nearly perfect crystal of synthetic diamond in thermal equilibrium, energy is transported isotropically by propagating lattice vibrations (phonons) whose spectrum is Planckian. When energy is deposited because of the absorption of light (linear processes), phonons carry the energy away from the deposition site. The phonon distribution becomes skewed and the limit to the distribution asymmetry permitted by the crystal determines the resistance to the transport of heat. Two processes affect the shape of the disturbed phonon distribution: "normal" processes (N-processes) and Umklapp processes (U-processes). In N-processes, momentum is conserved on a microscopic basis. Such interactions do not restore a skewed distribution to its Planckian form. If only N-processes could occur, there would be no resistance to the flow of heat and the thermal conductivity would be infinite. In U-processes, momentum can be transferred to the lattice (equivalent to a Bragg reflection of the phonon). U-processes do tend to restore the distribution to its equilibrium mode and thus provide a limit to the thermal conductivity. Other conditions, such as imperfections in the crystal, structural defects, impurities, and mass defects (isotopes) also contribute to the U-process rate.

The importance of reducing U-process rates can be appreciated by seeing how they affect the thermal conductivity. For a Debye-type solid, the relaxation-time method¹ gives for the thermal conductivity K ,

$$K = \frac{4\pi h^2}{v k_B T^2} \int \nu^4 \exp(h\nu/k_B T) (\exp[h\nu/k_B T] - 1)^{-2} R^{-1}(\nu, T) d\nu$$

$R(\nu, T)$ is the rate of occurrence of the various kinds of U-processes. Some of the contributions to $R (= \sum R_i)$ are:

Natural effects:	$R_n \sim (\nu T)^2 \exp(-270/T)$
Isotope effect:	$R_I \sim \nu^4$
Point defects:	$R_{pd} \sim \nu^4$

If K is to be as large as possible, then R must be small. It is therefore necessary to use diamond at low temperatures, where the natural U-process rates are very small, and to use material of the highest quality. In the extreme, the use of diamond made from a single isotope of carbon — presumably C^{12} — would be justified. In his analysis of the thermal conductivity of silicon at low temperature, Klemens³ noted the importance of single isotope material to attain the highest value of K . He also pointed out that if the absorption of light results in the production of optical frequency phonons, a multi-isotope material suffers a major degradation in K owing to the ν^4 dependence in the expression for R_I .

Observation is consistent with these ideas. Figure 1 shows the thermal conductivity of several materials as a function of temperature. In figure 1a, the thermal conductivity of LiF is shown for varying concentrations of the Li^6 isotope; the conductivity is greatest for

the least concentration of Li^6 . In figure 1b, the behavior of several materials is shown including the behavior of C^{12} diamond as projected by Seitz⁴. The rapid increase in K as the temperature decreases from 300°K is due mainly to the decrease in the natural U-process rate; the decrease on the low temperature side of the maximum is presumably due to phonon scattering at the boundary and the classical T^3 dependence. The limitation on thermal energy transport imposed by reflection at the boundary may be reduced by acoustic matching at that surface, a point to be addressed below.

2.2 Transparency Even the best natural diamond is not very transparent by optical standards. Type IIa diamond is most transparent, with reported¹ absorption coefficients of $0.01 - 0.001 \text{ cm}^{-1}$. Many samples of natural diamond show strong absorption at about $0.7 - 0.8 \mu\text{m}$ and in the vicinity of $2 \mu\text{m}$ and are thought to be due to the presence of nitrogen impurities. Such material would be unsuitable for a very high power window or lens because of the large energy deposition that would be realized in the optic.

In a perfect crystal, there is no basis for absorption (via linear processes) of light other than from the reststrahlen process, which is expected to be negligible. Consequently, nearly-perfect diamond should have a very low absorption coefficient from its ultra-violet cutoff wavelength at about $0.25 \mu\text{m}$ into the infrared. The evidence to support this notion is shown in figure 2. In figure 2a, the absorption coefficient of sapphire that is nearly free from its major contaminant, Cr, is less than 10^{-4} cm^{-1} . In figure 2b, the absorption coefficient of alkali halides is also shown to be of the same level when the chemical impurity level has been reduced sufficiently. Yoder⁵ attributes almost all absorption in diamond to impurities (figure 3) and argues that pure material will have a very low absorption coefficient. Material with an absorption coefficient less than about 10^{-4} cm^{-1} would make an excellent high power optical material as we show below.

We can find no measured data for the absorption coefficient of synthetic type IIa diamond. Most of the available material is made for heat sinks and has enough nitrogen contamination to give it a yellow color. Because of the importance of this material property, Los Alamos is presently engaged in the process of measuring the absorption of various samples of synthetic diamond.

3. Application to high power optics¹⁰

3.1 High power CW beams The power that can be transmitted by an optic is limited by the optical phase front distortion produced by the material temperature difference across the light beam. Insight into such a limitation can be obtained by considering the effect of absorption due to a light beam of intensity I_0 incident on a round window of thickness l_0 and radius R whose edge is held at a fixed coolant temperature T_0 (figure 4). Assuming symmetry about the axis of propagation, the equation that describes the window temperature is

$$KV^2 T = a I_0$$

where K is the thermal conductivity, a is the optical absorption coefficient in the

material, and the optical signal I_0 is assumed to have a square transverse profile with radius R_0 . For this simple case, the temperature difference between the center and edge of the beam is

$$\Delta T = -(1/4) \frac{\alpha P_0}{\pi K}$$

Expressed in terms of fractions of a wavelength, the change in optical path length between the center and edge of the light beam is

$$\frac{\Delta(nL)}{\lambda} = (\Delta T n_0 L_0 / \lambda) \left(\frac{1}{L_0} \frac{dL}{dT} + \frac{1}{n_0} \frac{dn}{dT} \right)$$

where the terms in the bracket are the physical and "optical" expansion coefficients, ΔT is the temperature difference, n_0 and L_0 are the index of refraction and the optic thickness, and λ is the wavelength. For a good quality optical beam, the maximum allowable path difference is about 1/10 of a wavelength. Combining these two equations, one obtains for the maximum incident power-thickness product

$$P_0 L_0 = 0.4 \pi \lambda K \left[\alpha n_0 \left(\frac{1}{L_0} \frac{dL}{dT} + \frac{1}{n_0} \frac{dn}{dT} \right) \right]^{-1}$$

Figure 5 shows $P_0 L_0$ plotted ($\lambda = 1 \mu\text{m}$) against the right hand side of the equation with the regions for room temperature operation of glass and diamond and cryogenic operation of diamond noted. It is apparent that diamond has an advantage of almost 1000 at room temperature and about 25,000 at 80°K , and that beam powers of tens to hundreds of megawatts should be able to be transmitted.

3.2 High peak power beams When high peak power optical beams, such as those produced by pulsed lasers or Free Electron Lasers (FELs) are used, additional failure modes may be important. For FELs, high average and peak powers can co-exist. If the average power is high, the peak power may be so large that non-linear processes can grow and adversely affect the diamond's optical power handling capacity. There are two types of FELs: those that use an induction linac to produce the electron beam and those that use an RF accelerator. Here, we confine our discussion to the RF FEL.

The pulse format from an RF FEL consists of a train of short pulses (micropulses) that are emitted continuously (the CW case) or in clusters (macropulses). The duration τ of a micropulse is determined by the frequency ν of the RF power used in the electron beam accelerator because of the need to keep the accelerating field constant during the transit of the electron beam bunch across an accelerator cavity. For example, for a frequency of 500 MHz, τ would normally be less than about 50 ps. The spacing between micropulses is usually stated as n , the number of RF cycles. The relation between the peak power P_p and the average power P is

$$P_p = \frac{n P}{\nu \tau}$$

Further, the diameter of the light beam produced in the wiggler of an FEL must be small to

make the laser gain usefully large and is usually about 1 mm. Using these as typical values and $n = 10$, the peak optical intensity is 40,000 times larger than the average intensity. For example, for a 100 kW "welding" laser in a 1 mm spot, the peak intensity is about 4 GW/cm^2 . Where the light emerges in macropulses, the peak intensity is related to the macropulse duty cycle and will be higher than in the CW case. With such intensities, many non-linear processes are possible.

3.2.1 Optical breakdown. Optical breakdown occurs when the large electric field of the light beam amplifies the free electron density in the lattice to the point where a microplasma is produced. It is an important issue for pulsed lasers where the pulse duration exceeds 1 ns. For the ultrashort FEL pulses, the threshold for optical breakdown is very high and is not expected to be a problem. However, the presence of ultraviolet light in the FEL beam (section 3.2.4) suggests that this conclusion needs to be re-examined. It is worth noting that for nanosecond pulsed optical beams, the damage threshold of diamond is 6-12 Joules/cm², significantly larger than for most materials⁵.

3.2.2 Multi-photon ionization. If the optical intensity is high enough, an atom can "absorb" simultaneously n photons whose combined energy exceeds the band gap in diamond. For a $1 \mu\text{m}$ light beam, this means that 5 photons must be incident on an atom in a time period of 2×10^{-16} sec. This corresponds to an intensity of the order of 1 TW/cm^2 and the probability of absorption scales as I^5 . At the GW/cm^2 level, the multiphoton absorption is probably less than the absorption owing to chemical impurities.

3.2.3 Stimulated Raman scattering (SRS) SRS is a concern because the SRS cross section in diamond is large and the process causes about 13% of the energy of each photon to be converted to lattice energy. If much more than about 10^{-4} of the beam is Raman-scattered, the energy load in a small optic may be unacceptable. To show how the effect of SRS can be mitigated, we consider the case where the diamond optic is used as a reflector, window, or diverging lens near the wiggler output of the FEL resonator. We assume an axisymmetric gaussian beam pulse traveling in the z direction,

$$I(r,t) = [2P/(\pi w^2)] \exp(-2r^2/w^2) \exp(-t^2/\tau^2)$$

where I is the peak intensity, P the peak power, τ the pulse width, and w is the beam diameter defined by

$$w = w_0 [1 + (z/Z_R)^2]^{1/2},$$

Z_R is the Rayleigh range $\pi w_0^2/\lambda$, and λ is the optical wavelength. As the primary light travels through the diamond, it "pumps" the medium and enables SRS to occur parallel and transverse to the beam direction. The differential gain due to SRS is

$$dG = I(r,z,t) \sigma dx,$$

where dx is the direction of integration and the SRS gain σ is given by

$$\sigma = \sigma_0 [\tau/(2T)] = 7 \times 10^{-9} [\tau/(2T)] \text{ (cm/watt)}.$$

T is the relaxation time (17 ps). The value for σ_0 is taken from McQuillan⁸; the term $[\tau/(2T)]$ is a correction for the short pulse duration⁹. Because the Raman and laser light

are co-located as they move through the optic, there is a relationship between the radius r and the local time t of the pulse, namely, $r = c' t$. c' is the speed of light in the diamond.

If the diamond optic is placed a distance Z from the wiggler, the beam will have expanded sufficiently that the SRS gain is too small to produce an excessive energy load in the optic. Z is defined by

$$\begin{aligned} Z/Z_R &= (c' \tau / w_0)^2 [(1 + Q^2)^{1/2} - 1] \quad (\text{Parallel}) \\ Q &= (\pi G^2)^{-1/2} (n \sigma_0 P) / (\nu \tau c' T) . \\ Z/Z_R &= [P (n/\nu) (\sigma_0/T) L / (\pi G w_0^2) - 1]^{1/2} \quad (\text{Perpendicular}) \end{aligned}$$

If G is less than about 25 (generations), SRS should not be a problem. Then for an average power of, say, 1 MW, the distance Z is several meters and the diameter of the diamond optic (which must be several times larger than the beam diameter) will be less than about 1 cm. Thus, by the simple expedient of moving the optic a short distance from the wiggler, the effects of SRS can be largely eliminated.

The cross section for other stimulated phenomena such as Stimulated Brillouin Scattering and filamentation is lower than that for SRS. Hence, if the SRS process can be avoided (as described above, for example), so can the consequences of these other processes.

3.2.4 UV damage effects. Because of the way electron beam energy is converted to light in an FEL, some optical energy is produced at harmonics of the fundamental optical wavelength. For example, for a fundamental wavelength of 1 μm , powerful FELs may produce significant amounts of light above the 5th harmonic where the diamond is opaque. The photon energy there is high enough to induce reactions that could lead to strong interactions with the powerful primary light beam, such as photochemistry of surface contaminants, production of free electrons near the surface, and UV damage to 1 μm AR coatings. The production of a large number of free electrons could seed the optical breakdown process described in section 3.2.1. This effect needs evaluation.

3.2.5 Hard radiation damage. FELs usually require beams with electron energies of about 50 - 100 MeV. The stopping of the electrons generates energetic photons and some neutrons, each of which can produce dislocations and color centers in the diamond lattice. However, in modern FELs, we believe that the use of low-emittance electron beams and efficient electron beam energy recovery techniques, coupled with appropriate shielding of the optics and the electron beam dump, can eliminate hard radiation damage.

3.3 Acoustic matching. In a diamond optic, energy absorbed from the light beam appears as phonons which propagate through the lattice until U-processes convert the phonon energy into lattice energy, i. e., heat. At low temperatures where U-process rates are nearly negligible, the time for conversion into heat is long compared to the phonon transit time across the optic. Phonons will make many reflections from the optic surfaces, and their inability to exit the surface efficiently limits the transport of energy out of the optic. Berman¹ indicates that the constraint may be a factor of 2 - 3. If this constraint could be removed, the energy burden in the optic could be significantly reduced. For example, assume that the optic is a window with very smooth planar surfaces. At such surfaces, most phonons reflect (specularly) because of the large acoustic mismatch between the diamond and the external low density medium. The specular reflections move the phonons towards the edge of the window. If the region near the edge of the window is acoustically matched to the external medium

(presumably a coolant), it may be possible to transport much of the phonon energy out of the optic *before* it becomes heat.

Little⁶ has analyzed the transfer of heat through two joined isotropic solids. With some simplifying assumptions (e. g., perfect contact at the interface), he finds that the power flow through the interface is

$$P = 20 \Gamma T_1^3 \Delta T c_1^{-2} \text{ GW/cm}^2$$

T_1 is the temperature ($^{\circ}\text{K}$) in the reference solid; c_1 is its velocity of sound (cm/sec); and ΔT is the temperature difference between the two media. Γ is a complicated function of the sound velocities and densities of the two materials and has a maximum value of 0.5 when the media are acoustically matched, i. e., $c_1 \rho_1 = c_2 \rho_2$. At low temperatures where the phonon mean free path is large compared to surface irregularities, imperfect contact has only a small effect on P . In their measurements of thermal transport across boundaries, Swartz and Pohl⁷ found good agreement with Little's acoustic matching model for $T < 30^{\circ}\text{K}$ and attribute the disagreement above that temperature to phonon scattering in thin disordered layers at the interface.

If we use Little's expression for P to estimate the power flow across the interface of an optic of radius 0.5 cm and thickness 0.2 cm at 100°K , and using $c_1 = 10^6$ cm/sec, the power flow is about $40 \Gamma \text{ kW}$. If acoustic matching is not employed, Γ can be less than 0.01 - 0.001 instead of 0.5 (perfect match).

We suggest that it may be possible to approximate acoustic matching by inserting multiple layers of suitably chosen materials as the interface between the two materials. Then the product ρv changes from $\rho_1 c_1$ to $\rho_2 c_2$ on a scale long compared to the acoustic wavelength. The concept is similar to that of a graded optical index variation for AR interfaces.

4. Discussion

The principles discussed in this paper are illustrated by the concept of a diamond window shown in figure 6. The 1 cm window is edge cooled with liquid nitrogen, and is made from chemically pure synthetic diamond that has very few lattice defects. If the window thickness is no more than a few mm, the total optical absorption can be less than about 10^{-5} of the incident power. To facilitate the transport of phonons out of the window and into the substrate/coolant, the outer edge interfaces utilize layered films to provide acoustic matching. Even if the acoustic matching is only 20% effective ($\Gamma = 0.1$), the power flow through the interface could be as large as 4 Kw. If this represented the total power deposition in the optic, the incident power would have been 400 MW.

Alternately, one might replace the acoustic matching layers with low-pressure deposited diamond to bridge the gap between the window and the supporting fixture. In this approach, phonon transport in the diamond film is probably poorer, as is the transport into the fixture. However, the contact area between the diamond film and the fixture can be made much larger and the fabrication process may be simpler.

AR coatings for such a window presents some interesting challenges. Normal multi-layer films will have an optical absorption that is much larger than that of the window. However, the thinness of the films and the enormous thermal conductivity of the underlying diamond may prevent heating and temperature gradients in the layers. As an alternative to films, it may be possible to use reactive ion etching on the surfaces to produce a graded optical index similar to the process used for glass optics.

Finally, we note that two important effects have not been reviewed here. These are the absorption at the surface of the optic, and the effect of the photoelastic properties of diamond. The former results in an absorption at the optic surface that may be as large as its bulk absorption, and the latter causes a phase distortion in the beam that does not depend on the material temperature change across the beam.

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6. Footnotes

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Figure captions

Figure 1. The isotope effect in thermal conductivity. (a) a comparison of measured thermal conductivity with the theory of Calloway for LiF for varying concentrations of the Li^6 isotope. The Li^6 concentrations are (per cent): . Δ 0.02; Δ 0.01; x 4.6; 0 90.4; 0 25.4; + 50.1. The explanation for the higher thermal conductivity of the 0.02% samples is based on the presence of other impurities (Berman¹). (b) The temperature dependence of the thermal conductivity of several natural and single isotope materials. Courtesy, Russell Seitz.

Figure 2. The effect of chemical purity and processing on the optical absorption of transparent materials. Shown is the optical absorption in sapphire vs photon energy for a highly purified sample and the identified residual absorption due to Cr^{3+} . Also shown is the absorption coefficient vs photon energy of alkali halide indicating the effect of doping and forging (vs single crystal properties).

Figure 3. The wavelength dependence of the optical absorption of several materials and natural diamond. The absorption in diamond near $10\text{ }\mu\text{m}$ is expected to diminish greatly in very pure synthetic material. (Yoder⁵)

Figure 4. Optical power throughput capacity of a window. The figure depicts a flat-profile optical beam of radius R_0 and power P_0 incident on a flat diamond window of thickness L_0 , radius R , thermal conductivity K , and optical absorption coefficient α . The edges of the window are held fixed at a reference temperature.

Figure 5. A plot of the power-thickness product of an optical material vs the figure of merit developed in the text. The data for the parameters in the figure of merit are taken from Singer¹⁰.

Figure 6. Concept for a very high power diamond window based on the low temperature thermal conductivity and low absorption of very pure cold diamond and the use of acoustic matching to transport energy out of the window.

Fig. 4

Fig 5

Fig. 6