Gamma Radiation Studies on Optical Materials

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Abstract—Results for the effects of γ’s on materials for a new laser-driven accelerator are presented. Various optical and laser materials are compared. Si and fused c-SiO₂ appear ideal for sub-bandgap laser wavelengths.

Index Terms—lasers; damage; silicon; a,c-silica; impedances; transmissive/reflective; parallel/serial.

I. INTRODUCTION

A NY number of studies, in various contexts, on the effects of radiation on electronics exist but there are relatively few on the optical properties of interest here where both surface and bulk properties are important. An example is laser damage thresholds as a function of pulse width, polarization, frequency, power and number of pulses as well as its interplay with other sources such as displacement damage. Further, the range of materials studied is typically limited to the obvious ones for space applications e.g. glasses[1], [2] that can be quite sensitive and hard to fully remediate.

Our first accelerator cells[3], [4] were modular and based on high-reflectivity, coated silica. Our ultimate goal is an “accelerator-on-chip” with reflective and transmissive optics in a micromachined, highly repetitive, parallel architecture[5]. Thus, we are pursuing a program whose goal is to understand the sources and types of radiation, their magnitudes, effects and potential mitigations as well as the underlying science. Here, we describe the basic problem and then give our first results and conclusions based on using a single, well-defined source - the Lockheed Martin Co₁₀⁰₀ γ-source.

II. PROBLEMS AND POTENTIAL PAYOFFS

High energy colliders are arguably the most complex scientific instruments ever built using every form of technology. As for many modern systems, they have grown exponentially[6]. However, in most cases, the cost and reliability have improved due to reduced size as well as increased production and integration[5]. Although the next generation collider will use conventional RF, there is a clear need for new techniques such as laser acceleration, assuming adequate materials are available. These systems and their tensor electron beams suggest a number of important applications including fast electro-optical switches, high density magnetic storage and direct-write, e-beam lithography[5]. Similarly, due to the size and types of materials, one also expects many space applications that would be impractical with current accelerator technology.

However, because future colliders require higher energies and luminosities, beams with unprecedented energy densities are required. While such beams provide many possibilities for frontier physics, they also imply unprecedented containment and damage problems.

Fig. 1 demonstrates the problems and opportunities. Most glasses have complex, compound compositions that are quite susceptible to damage. Varieties tested with 152 MeV protons all showed damage at 10 kRad(Si)[2]. In our first run, a thick, plate-glass sample was irradiated with 42 kGy of gammas that visually blackened it and lowered its transmission spectrum as shown by the lowest curve in Fig. 1. We tried to restore this using both UV and thermal anneals typical of those we use for quartz vacuum windows. The best we could achieve after many attempts is shown by the middle curve. The material is a better UV/visable filter or detector than it is a window for our uses.

III. BASIC IDEAS AND CHARACTERISTICS

One can understand vacuum laser acceleration consistently for both particles and fields as well as conventional RF even though conducting waveguides have much higher permittivity. The limiting factor on the acceleration gradient is the electric field damage threshold of a material. Structures made from dielectric materials are typically expected to provide an order of magnitude improvement over what has been achieved with conventional RF. Phase space volumes, phase and group velocities, emittance as well as electrical and optical impedances are shared concepts as are their scaling and combinatorics.

![Fig. 1. Transmissivity spectra through 1.1 cm thick plate glass after Co₁₀⁰₀ γ-irradiation. Spectra are stacked according to their order in the insert.](image-url)
A. Figures-of-Merit (FoM)

While there are many FoMs such as the invariant 6D brightness useful for applications such as e-beam lithography, we will discuss only those useful for damage considerations. For example, in the detector region, showers from radiative Bhabhas can damage permanent magnets and detector components/monitors and this rate is directly proportional to the luminosity. Likewise, impedances relate directly to aging and damage effects such as heating.

1) Luminosity: The most important figure-of-merit for colliders is the usable luminosity. The generalized luminosity[7], [8] was based on the observation that all colliding beam machines as well as all incident channels in any particular collider can be expected to have a luminosity proportional to the square of the primary, incident bunch ‘charge’ $N_B^2$ or some equivalent thereof that can be brought into collision per unit time within an effective area that contains the effective number $\bar{N}_B$ based on conversion efficiencies and detector constraints[7], [8], [9].

For a laser driven accelerator, when the laser and e-beam normalized emittances are matched, one can write, in terms of the standard expression,

$$\mathcal{L} = \frac{f \times n B N_B^2 H_{\lambda}}{4 \pi \sigma_x \sigma_y} \zeta = \frac{f \times n B N_B^2 \gamma H_{\lambda}}{4 \pi \varepsilon \beta^*} \zeta \approx \frac{P_L}{\lambda \varepsilon R} H_{\varepsilon} \zeta \hspace{1cm} (1)$$

where the parameters are the usual ones[7], [8], [9] e.g. $\sigma_{x,y}$ is the undisrupted, rms spot size at the interaction point (IP) and $\beta^*$ is the magnetoptical ‘depth-of-field’ at the IP. The arrow simplifies to round beams and $P_L(f n N_B)$ is the incident, primary electron beam power. $H_{\varepsilon}$ is a pinch enhancement factor of order unity. $\beta^*$ is equivalent to the Rayleigh range $Z_R>\lambda$, the laser wavelength. In this scenario, luminosity increases with decreasing $\lambda^2$.

2) Impedances: One sees that $\mathcal{L}$ is proportional to the power available to accelerate the primary beam. The on-axis, unloaded gradient of an accelerator cell can be defined as

$$G_U = \frac{\sqrt{P_Z \varepsilon}}{\lambda} \frac{1}{\sqrt{2} \varepsilon_{\mu m}} G_U [MeV/m] = \sqrt{P[Z]} \hspace{1cm} (2)$$

assuming $\varepsilon_c \equiv 2.25 \Omega$. This defines the characteristic impedance of a structure’s accelerating mode that can vary greatly but is reasonable. The peak power is determined by the structure’s damage threshold. Dielectrics like fused silica typically allow 1-2 J/cm²[10] in the near IR below 10 ps pulse widths. Commercial sources[11] are available that can provide 50 MHz rep-rates as compared to typical RF rep rates of 120 Hz.

This expression implies shorter wavelengths are preferred over power or impedance but if power or efficiency was not a problem we could get any $\mathcal{L}$ we could use. Any choice of $\lambda$ assumes that one has the power source and the means to fabricate structures to the required tolerances that can withstand the power at that wavelength. With the rapid development of high power lasers and micromachining techniques we have a good justification for LEAP[4] with good materials science.

If we assume that a fixed power is available, we can enhance $\varepsilon_c$ by series addition of cells to improve energy gain per unit power by $\sqrt{n_c}$.

$$E_b = \sum_{n_c} \frac{l_c}{\sqrt{n_c}} Z_c = l_c \sqrt{n_c} G_U . \hspace{1cm} (3)$$

$l_c$ is the acceptable slippage distance between bunchlet and wavelet $l_c \leq \lambda/2(1-\beta)$. This increases the length and it is clear that adding cells in parallel[8] can reduce it. The impedance of this parallel path $Z_T$ for a wavelet determines the so-called shunt impedance as well as the allowable number of parallel bunchlets. It is determined by $Z_c$ and the transmittance $T(\lambda)$ of the structure between cells. As will be shown, $T$ can be large for silicon (or silica) for laser wavelengths $\lambda>1.2\mu$m when we couple the wavelets into these materials properly.

The beam excites higher modes that dissipate energy and this provides another impedance $Z_h$ that leads to a loaded gradient $G_L$ that is not less than half $G_U$ but depends on the charge per bunchlet. This is a justification for tensor beams that can reduce this charge. Clearly, we want to increase $Z_c$, reduce $Z_h$ and maximize $T$.

B. Dielectric loaded waveguide/accelerator

Whether one has a disc or dielectric loaded guide structure, the modes can be classified as transverse magnetic TM$_{klm}$ with $E_z \neq 0$ and $B_z = 0$ and p polarized wavelets for cylindrical symmetry. For waves of finite extent, the group velocity in any direction is $v_g < c$. From the waveguide dispersion equation or geometric arguments one then has, above cutoff,

$$\beta_g = \frac{v_g}{c} = \sqrt{1 - \left(\frac{v_g}{v_p}\right)^2} \quad v_g \sqrt{v_p} = \frac{c^2}{n^2} = \frac{1}{\varepsilon \mu} . \hspace{1cm} (4)$$

The index $n$ is generally complex and the cutoff frequency $v_c$ results from the waveguide’s cross section e.g.

$$v_c = c \sqrt{\left(\frac{k}{2a}\right)^2 + \left(\frac{l}{2b}\right)^2} \hspace{1cm} (5)$$

for a rectangular guide. Note that the mode’s unloaded, lossless wave impedance $Z_T = (E/H)_{xy} = \beta_g Z_0$. We want $v_p$ close to $c$ i.e. $v \approx v_c$ because this increases the slippage distance or guide length. For high enough energies:

$$v_p/c = \lambda w_g / \lambda = \sqrt{1 - \left(\frac{1}{\lambda / \lambda_c}\right)^2} \leq \gamma^2 . \hspace{1cm} (6)$$

This ratio is determined by varying the cell length for a given laser $\lambda$, aperture and crossing angle to optimize gain. One concludes that we must either increase $\lambda_c$ i.e. the guide size or decrease $\lambda$ - neither of which is very agreeable except for the large changes achievable with lasers.

An old idea is to load the waveguide with dielectric. For small apertures:

$$v_p/c = \lambda w_g / \lambda = \sqrt{\frac{1}{\varepsilon \mu r - (\lambda / \lambda_c)^2}} \leq \gamma^2 . \hspace{1cm} (7)$$

This expression suggests a graded index and shows why a laser accelerator is possible. Increasing $\varepsilon$, or $\mu_r$ from vacuum values provides knobs that allow smaller waveguides and higher accelerating fields. Problems include variable capacitance through charge buildup from lost electrons, dielectric breakdown and aging from poor heat dissipation or radiation damage.
IV. Importance of Reflectance and Transmittance

In contrast to RF approaches, the reflectance, transmittance and absorbance, with \( R + T + A = 1 \), are important to every part of our problem. Thus, the expressions above are valid and, in a geometric picture, they can be thought of as defining the mean angle of propagation of the fundamental mode down the guide i.e. \( \beta_n = \cos \theta_c \). For conventional metallic guides, the characteristic angle \( \theta_c = 90^\circ \) defines the cutoff frequency where there is no propagation down the guide. For consistency with optical conventions, we will rotate our angle by \( 90^\circ \) because the guide surface is more relevant than its axis. Thus, normal incidence, defining \( k_\perp = k \), now occurs at \( 0^\circ \).

Brewster’s angle \( \theta_B \sim 52^\circ \) for the wavelengths and dielectrics or semiconductors of interest here so this angle defines the onset of the parallel, transmission mode while \( R \) and the number of reflections per unit length determines the extinction rate of the serial, reflective mode for smaller angles and thereby the serial shunt impedance \( Z_R \) as opposed to \( Z_T \) mentioned earlier.

Dielectrics have nearly zero absorbance while metals always have greater than zero. Polished metals thicker than \( \approx 50 \text{ nm} \) can have high reflectances up to 98% at normal incidence while uncoated dielectrics such as glass typically have 4-8% but the addition of a single, quarter-wave layer of intermediate index material can reduce this to <1%. This is an example of impedance matching where \( R \) can be thought of as resulting from some mismatch. It depends on the materials, the angle of incidence, wavelet polarization, surface structure and incident intensity.

V. Damage Mechanisms

In our damage studies, we scan light of variable frequency \( \omega \) at normal incidence on flat polished surfaces and measure \( R \) and \( T \) for materials subjected to increasing intensities \( I_k \) or doses \( D_i \) of \( \gamma \)-rays or neutrons. \( R \) and \( T \) depend on many characteristics besides the surface finish and thickness \( l \) i.e. \( R \equiv \mathcal{R}(\omega,l,\theta,P,\epsilon_j,\mu_j,I_k,E_{BG},D_i,t) \). We can write:

\[
R = \frac{(Z_0 - Z_l)(Z_0 - Z_l)^*}{(Z_0 + Z_l)(Z_0 + Z_l)^*} \frac{\alpha^*}{\alpha} \left[ \frac{[(n_2 - n_1)^2]}{[(n_2 + n_1)^2]} \right]^2
\]

(8)

with \( P \) the wavelet polarization, \( E_{BG} \) the bandgap energy, \( Z_j(\omega) = (\mu_j/\epsilon_j)^{1/2} \) the complex wave impedance. \( Z_2 \) can be taken as a terminating impedance and \( Z_1 \) as the characteristic impedance of a lossy line. We write the transmissivity similarly and \( n_j \), the complex “index”, as:

\[
n_j(\omega) = \sqrt{\mu_j/\epsilon_j} = n_{0j}(\omega) + i k_{0j}(\omega) + n_{2j}(\omega) I(\omega) + \cdots
\]

(9)

where \( n_{0j}(\omega) \) is the index of refraction that determines the phase (and group) velocities (4) and \( k_{0j}(\omega) \) is the extinction index that determines the absorption coefficient \( \alpha_j(\omega) \):

\[
\frac{T(\theta)}{T(0)} = e^{-\alpha t} \quad \text{with} \quad \alpha_j = 4\pi \nu k_{0j}.
\]

Frequency is in wavenumbers. \( k_0 \) relates to conductivity so it is negligible for dielectrics in the visible and infrared even around strong vibrational bands. Similarly, for semiconductors below bandgap e.g. standard grade Si [12], one has \( k_0 \ll 10^{-5} \) until multi-phonon absorption picks up below 1200 cm\(^{-1}\).

The Sellmeier model describes resonant, nonlinear variations of \( n_{0j}(\lambda) \) with \( \lambda \) over this region as well as the dispersion \( d n_{0j}/d\lambda \) discussed later. The sign and magnitude of the leading nonlinear term with intensity, \( n_{2j} \), can lead to a variable lensing action. Such terms produce laser damage through multiphoton excitation and ionization processes \( q \omega > E_G \) or self focusing effects[13] that increase the effective intensity \( I \). Some typical results and measurements are shown in Table I. As expected, no correlation in the damage results have been observed.

### Table I

<table>
<thead>
<tr>
<th>Material</th>
<th>Laser wavelength (( \lambda = 800 \text{ nm} )) with ( 150 \text{ fs pulse duration} ) and ( 13 \mu \text{m} ) spot size.</th>
<th>( \gamma )-Dose [kGy]</th>
</tr>
</thead>
<tbody>
<tr>
<td>a-SiO(_2)</td>
<td>2.03 1.80</td>
<td>50 62 67 134 60</td>
</tr>
<tr>
<td>Al(_2)O(_3)</td>
<td>1.57 1.15</td>
<td>45 63 61 139</td>
</tr>
<tr>
<td>HR(800)</td>
<td>0.77 0.77</td>
<td>49 72 59 134 64</td>
</tr>
<tr>
<td>LiNbO(_3)</td>
<td>0.50 0.19</td>
<td>46 67 63 141</td>
</tr>
</tbody>
</table>

Radiation damage is specific to the type of radiation and the integrated dose. Materials age, damage, anneal, relax, absorb or adsorb other materials in ways that can seriously change their properties in time dependent ways. Such changes can show a strong frequency dependence as when hydrogen diffuses into or out of silica or the growth of color centers from point defects.

Such defects can be vacancies, substitutional dopants or impurities, substututional interstitials or impurities that migrate. These are important because they behave differently in amorphous and crystalline materials as well as metals and oxides. In oxides, they are often charged which can lead to significant changes in light transmission. In some semiconductors, we have also seen these effects in the bulk transmission due to intentional doping that can have significant concentrations and can be both charged and neutral. Undoped, thick Si may have observably better transmission than typical Si wafers.

VI. Choice of Radiation Source

The types of dose and their magnitudes in a collider depend on the subsystem and the location within the subsystem. Typically, in descending order come the beam dumps, the positron target, the damping rings, the accelerator, the detectors and the electron source. Of all of these, the source is the lowest energy and of most interest for practical applications.

Because most source implementations (including ours) involve only low energy electrons, x-rays and gammas (\( E < 10 \text{ MeV} \)) we chose the Lockheed Martin Co\(^{60} \) \( \gamma \)-source for two reasons. First, it is well calibrated with a well-defined isoradation grid that allows irradiating many samples simultaneously. More importantly, it provides an average \( \gamma \) energy of 1.25 MeV whose stopping power is equivalent to \( 600 \text{ keV} \) electrons[14]. In fact, the LET is rather flat for electrons from 200 keV-20 MeV. Thus, it is an ideal source to study materials for this important system that will necessarily use many of the same materials as the accelerator and detectors[5].
VII. EXPERIMENTAL SETUP AND PROCEDURES

We used an Hitachi U-4001 spectrophotometer to obtain spectra for various conditions over a wavelength range from 200≤λ(nm)≤3200. Due to water absorption bands near 1400, 1900 and 2700 nm, we purged the system with dry air and maintained a positive flow during measurements. One still sees occasional fluctuations in these regions (see Fig. 1) that can be due to changes in surface moisture or when running for extended periods. Baseline scans are necessary for this and to match different sources and detectors over the range that spans the near-UV, visible and near-IR.

Widely different shapes and sizes of samples complicated the situation but they all had polished, parallel entrance and exit surfaces. We used perpendicular entrance angles that were good to a degree or better between the different irradiations for which the relative calibrations were on the order of 0.5% based on using calibration samples and running sample-free scans against our baseline before and after measuring each batch of irradiations. All irradiations and their subsequent measurements were done at room temperature on samples that were “encapsulated” when not undergoing measurements.

The scan rate, range, resolution and step size were all varied for effect as well as for the material being scanned e.g. the high reflector coatings and laser rods were run at both low and high resolutions down to 1 nm steps. The lowest wavelength accuracy of the instrument was ≈1 nm at the longest wavelengths. The shortest wavelength was set to include the 4th harmonic of Nd:YAG at 266 nm although we would have liked to include the VUV down to 100 nm and the mid-IR through 14000 nm because longer wavelength, higher-power, stable lasers would simplify our particular application. Having the higher ranges available also simplifies the assignment of contaminant lines.

After our last irradiations, we obtained access to an IFS 66v/S Bruker FTIR spectrophotometer that allowed overlapping measurements up to 10 μm. Due to strong water absorption above 4-5 μm, this system operates under vacuum which is an important improvement for nearby, weak absorption lines.

VIII. MEASUREMENT RESULTS

A. Amorphous materials

Amorphous, fused silica (a-SiO₂) is an ideal example whose properties support many applications. It was used for our first accelerator cells due to its good thermal stability that provides stable, high-reflector coatings under high power beams. Because it has a high bandpass for our nominal wavelength range, it has many uses that make its transmission characteristics and their stability important. Fig. 2 shows the transmission spectra, in 1 nm steps, for an uncoated, polished accelerator cell from CVI Laser[15] as a function of total γ-dose in Si equivalents.

The maximum transmission here is T ≈ 95 % at 1600 nm. There are four prominent absorption regions near 210, 1385, 2210 and 2760 nm or in frequency: 47,620, 7,220, 4,524 and 3,622 cm⁻¹. The most interesting of these is the activation dip at 212 nm (5.84 eV) where significant damage appears but it is well below any laser wavelength λ₂ of interest here except for the 4th harmonic of Nd:YAG where it may be useful for our electron source[5].

Because of its high energy and the fact that the band gap for pure SiO₂ (defect-free quartz or amorphous) is E_g>8 eV (155 nm), this results from electronic excitation of impurities or lattice defects. The ionization potential (IP) of free Al is 5.98 eV or 207 nm but this can’t explain the result without creation of additional stable defects of comparable energy because of the strength of the effect. Because the silica[15] is of Type-III, its largest metallic component is expected to be Al (10-20 PPM) which which we see is too small. Although it has the potential for far more Cl, the ionization potential (∼13 eV) is too high to be relevant and HCl vibrations are too low. We know of no other nearby electronic excitations except Na at 5.14 eV (241 nm). It is present in the few PPM range and helps explain the nearby transmission shoulder.

Similarly, we note that free Si has an IP≈8.2 eV but has a 3p-3d transition at 221 nm. E' defects[1] related to oxygen vacancies with an electron trapped in the 3p orbital of a Si-Si bond provides a stable defect(hole). Since this can be produced with 1 MeV photons from secondary Comptons this can explain the effect. The observed damage is linear with dose:

\[ T(212 \text{ nm}) = 0.823 - 1.36 \cdot \text{Dose(MGy)} \]  

although it is barely visible at low doses for neutrons or γ’s. Increasing the dose would allow us to determine whether the effect saturates as observed for the fused quartz wafer.

The dominant excitation near 2760 nm is often observed. We interpret it as a combination of water and a two-dimensional stretching mode associated with OH that is typically[15] at 1000 PPM. The line at 1385 nm reinforces this assignment by its interpretation as the first harmonic. The small modulation near 900 nm increases with total dose and is the second harmonic with evidence for higher harmonics in the subsequent falloff near 670 nm. This is a good indication of broken SiO₂ bonds that are subsequently passivated by H to form Si-O-H consistent with the fact that OH is energetically more stable than water: H₂O + Si-O-Si- → 2(-SiOH) and molecular hydrogen[17]. While we have seen evidence for growth of Non Bridging Oxygen Hole defects (NBOH) near 670 nm[18] in both SiO₂ and Al₂O₃, that has no water lines, we saw no obvious growth at 1385nm. This may be due to migration loss of the cracked hydrogen or surface related.
The remaining region around 2200 nm is two or more lines, as is the strong excitation at 2760 nm with a weak water band evident at 2600 nm (Fig. 1 also). It is important to note that water has symmetric and antisymmetric stretching modes near 3650 and 3760 cm\(^{-1}\) with the latter (2660 nm) the stronger. The difficulty in the 2200 nm region is that it could be related to harmonics of unobserved, lower lying levels. The simplest interpretation is that it results from H\(_2\) vibrations because of its energy. The near degeneracy could relate to passivation of O vacancies or substitution impurities in the form of Si-H H-Si or Si-H-Si. Alternatively, one knows that CH\(_4\) and CO (also H\(_2\)) are hard to remove from processes even under UHV pumping at 10\(^{-10}\) Torr. While it is difficult to deal with harmonics or coupled bands, one might assign these lines to harmonics related to vibrations of CO and a triple bond of CO. The spectral transmission of CO\(_2\) matches the profile from 2-3 \(\mu\)m very nicely but there is no evidence for carbon in silica. We will discuss this in more detail by comparison to crystalline SiO\(_2\), Si and Lithosil\([16]\] that has extremely small metallic impurities.

A discussion\([19]\) of hydrogen defects in a-SiO\(_2\) shows the difficulty arising in understanding amorphous materials and helps to explain the lack of systematic characterizations that provide more direct predictive power for glasses.

The results for a-SiO\(_2\) apply to many substrates insofar as transmission is concerned as shown for a high reflector coating (HR) in Fig. 3. This is intended for use in the Ti:Sa region but is not the same silica material used for Fig. 2 because of the cell’s shape. The sharpness of the primary reflection band and the surrounding oscillations result from multi-layer coatings with quarter wave optical thickness \(l = \lambda/4n_\text{HR}\) on a substrate with \(n_\text{Si} > n_\text{HR}\) based on phase changes \(\delta = 2\pi l n_\text{HR}/\lambda\) for incident light with wavelength \(\lambda\) at normal incidence with a \(\pi\) phase change on reflection at the \(n_\text{Si}, n_\text{HR}\) interface.

Previous lines, observed at longer wavelengths in Fig. 2, are interspersed with different orders about the primary near \(\lambda = 800\) nm. Because the light was incident on the substrate side in these two runs, the peak transmission (modulated) is essentially the same as for Fig. 2 and shows that \(k_\text{Si}\) is negligible except near the absorption bands that act as bandpass filters. The dispersion increases dramatically at the shorter \(\lambda\)’s (Fig. 10). No damage was observed in the HR coating up to 378 kGy.

### B. Crystalline materials

Examples for crystalline data are shown in Fig. 4 for comparable thickness wafers of quartz (c-SiO\(_2\)) and silicon. There is no evidence for damage in any Si sample up to 9 mm for \(\gamma\)-doses up to 370 kGy. Similarly, there is virtually no damage to the quartz except in the near-UV below 300 nm. Contrary to the amorphous silica (a-SiO\(_2\)) in Fig. 2, the damage in quartz appears to saturate. Up to \(\approx 300\) kGy, it grew more slowly at a rate:

\[
T(212) = 0.908 - 0.539 \cdot \text{Dose (MGy)}
\]

or \(< 40\%\). There is less water in the quartz which indicates the possibility of fewer O vacancies that can be passivated. Using the Lambert-Beer law we can estimate vacancy concentration:

\[
C(\text{PPM}) = \frac{100}{t(\text{cm})} \log_{10}\left(\frac{T}{T_0}\right) \approx 150
\]

where \(t\) is the thickness of the sample. This is consistent with the expected water content in the a-silica in Fig. 2.

Based on these results, it appears that both Si(\(\lambda_L > 1.2\mu\)m) and fused c-SiO\(_2\) (\(\lambda_L > 0.3\mu\)m) are adequate with certain caveats for the quartz but that quartz is clearly preferable to fused a-SiO\(_2\) for longer wavelengths.

![Fig. 4. Transmission spectra through 500 \(\mu\)m wafers of Quartz(upper group of curves) and Silicon(lower group) as a function of integrated dose (Si).](image)

It is useful to study other groups in the Periodic Table as well as hybrid combinations within a group. SiGe and SiC are examples of substitution elements from the same group. SiC has better mobility, bandgap (3.26 eV), breakdown voltage and thermal conductivity than Si. We also explored both synthetic and natural diamond. The two samples that we studied had such poor transmission over the whole range that we could say nothing about the radiation damage. The natural diamond had many defects, presumably from natural radiation damage, that make it useful as a radiation detector but not for most of our applications.

We were able to get good III-V GaAs and have just obtained several II-VI ZnSe samples. With proper doping, these materials provide good semiconductor lasers with GaAs being the first known example. Fig. 5 shows a thick sample of GaAs that is compared to an undoped, 320 \(\mu\)m (110)-wafer. This wafer is especially interesting for micromachining tests. GaAs(1.43 eV)
is easier to do laser damage tests on than Si(1.12 eV) because of its higher bandgap energy that allows us to use the fundamental of Nd:YAG but not Ti:Sa as used for Table I. While the refractive index is somewhat lower so that it is more reflective than Si, it is still comparable and has a much higher extinction index. While both GaAs samples were undoped, they were still too dissimilar to analyze for the complex, nonlinear index (9). Because there was no evidence of damage, we began controlled neutron doses on similar samples.

Because most metallic and non-metallic materials absorb strongly in this region while water is quite transparent above 8 μm, Fig. 6 argues against using CO₂ lasers for our application even though longer wavelengths above 2 μm are needed as discussed later in a section on dispersion.

However, beginning around 4.5 μm, water absorbs strongly up to 7 μm. As a result, the transmission for the a-SiO₂ cell went essentially to zero and stayed there above 4400 nm with lines near 3800, 4150 and 4400 nm observable. These same lines are seen in the quartz with a 4450 line strongest. This is consistent with the existence of ionic molecules of H₂O. Finally, there were observable lines in both SiO₂ samples near 3100 and 3200 nm that were not seen with the Hitachi instrument.

D. Laser and electro-optic materials

Important laser crystals of particular concern were Ti:Sa and Nd:YAG because they are the most widely used for high-power applications in the near IR e.g. Nd:YAG has a fundamental at 1.06 μm. Sapphire is a synthetic, hexagonal crystal of aluminum oxide and yttrium aluminum garnet (YAG) is a cubic crystal that can be grown with high quality so there was some hope that these could withstand radiation better than other host replacement materials such as glasses that are suspect for many reasons. Both materials have a broader transmission bandwidth than a-SiO₂ with similar or better radiation resistance up to nearly 6 μm.

We have used Ce doped YAG for our fluorescent screens that see full electron beam with no apparent degradation over the course of many runs remaining a brilliant yellow. We obtained a variant of the Nd:YAG crystal having 1 % Nd and 0.5 % Cr that still appears green (≈ 530 nm). Fig. 7 compares spectra before irradiation.

The Cr³⁺ rod is 2.5 cm long by 0.635 cm diameter and the pure Nd³⁺ is 10 cm long with polished sides. Although the primary transmission is somewhat different, the main differences show up best in the short wavelength, electronic region. After only a minor irradiation comparable to the glass in Fig. 1, there were obvious visual changes that grew worse as shown in Fig. 8. This was one reason for obtaining the Cr variant. These...
results extend and compliment previous work[20] and are consistent with it where the increased Cr doping clearly improves the radiation hardness.

E. Dispersive effects and comparisons between materials

It is interesting to consider the dispersion curves for the different materials because of the especially short laser pulses that are important here as well as the potentially long path lengths through the materials that may be required in the parallel, transmission mode. Fig. 9 shows some calculated curves over our typical wavelength range. These could be extended but were not because there was considerable dispersion in different results for certain materials. Examples are YAG and diamond that have impressive properties with important technical possibilities. Diamond (and Si) is an important covalent semiconductor so its dielectric properties are important but were only generally available in the visible e.g. an important, recent reference[21] cites a single 1923 work that covers a narrow wavelength range. A similar and related statement can be made for dn/dλ for a number of materials considered here.

In Fig. 9, characteristic bands of materials are evident as distinguished by a common crystal structure e.g. diamond and its equivalent for binary, compound semiconductors zinc blende.

The order within these two groups is correlated with their respective bandgap energies i.e. GaAs(1.43) and ZnSe(2.58) and for diamond (5.46), Si(1.12) and Ge(0.63). We did not plot diamond as discussed above. An analytic relationship between n, dn/dλ and E_G would provide predictive possibilities. Above 2.5 μm, Si is again the preferred material - especially considering our earlier discussions and silicon’s many practical advantages which indicates a need for lasers near or above 2.5 μm.

IX. SUMMARY AND CONCLUSIONS

Although radiation damage is an important subject for high energy physics, our concerns are quite different from the usual ones in our field. These tend to be highly focused and rather piecemeal even though the field provides an array of interesting scientific problems.

Because we were interested in laser and optical materials, we began to study some common glasses, including fused silica, since it was expected that these materials would be well understood because of their importance to the modern telecommunications industry for optical wave guides, nonlinear optics and laser glass. Our main interests were to find what was available, their general properties and to understand how different damage mechanisms changed or influenced these properties. The subject is more complex and interesting than anticipated – perhaps because distinguishing general properties from specific characteristics is complicated by the large variations in chemical composition and purities as well as the often large changes in response e.g. many of these materials can serve as detectors just as easily as windows depending solely on wavelength. To understand the importance of such characteristics our study has grown considerably.

We described laser acceleration in conventional terms and indicated some of the potential applications beyond high energy physics. While we were driven to this approach by the incredible advances in microelectronics and telecommunications that have been occurring, the need to integrate these is clear and the potential fallout is large[5]. Perhaps the only essential difference between what we are trying to do and industry is based on the need for extremely radiation resistant materials. However, if these can be developed in a natural way at a reasonable cost then it should benefit everyone.
We obtained results for a broad array of materials including a range of characteristics for some of them. Examples include differing thicknesses of quartz, Si, α-SiO₂, GaAs, Al₂O₃, Ti:Sa, YAG, diamond, LiNO₃, CaF₂, MgF₂ and a number of different glasses and dopings of Nd:YAG. The variations in response are remarkable. Fortunately, the quartz and silicon appear quite acceptable at current levels if we do not go above their long wavelength cutoffs as are the fluorides.

MgF₂ is a better window material than Sapphire or YAG and CaF₂, a cubic, is even better reaching up to nearly 10 μm but ZnSe runs up to 20 μm and for related reasons is a potential laser candidate[22]. CaF₂ is especially noteworthy having the highest laser damage threshold (> 2.2 J/cm²) with little to no difference between air and vacuum. Its transmission spectrum is quite flat and > 90% from 0.8-3.2 μm with no water evident.

We have seen no radiation damage in any fluoride sample but measurements are ongoing. Their dispersions are also lower than all materials except ZnSe.

The diffusion of water and hydrogen into silica and the development of silanol or hydride groups leads to the lowest absorption bands but generally restricts the use of silica to fused silica and quartz. It is interesting that diamond and Al₂O₃ show quite flat transmission but both show an effect at 670 nm. The most important result may be that Si appears to be an ideal material except possibly for CO₂ lasers. Although we have used it under full electron beams, we have yet to do the laser damage tests because of its unfavorable bandgap at typical laser wavelengths that are available (Ti:Sa and Nd:YAG). These will be done and we are also beginning tests on ZnSe (and ZnTe).

Clearly, neutron displacement studies are of interest both for Si but also for differences between a and c-SiO₂. Doping and impurities add a very large complication - presumably because they begin to lower the Fermi level to that of the defects that may be mobile and can take many forms. However, presently, it seems clear that pure amorphous materials are preferred over typical glasses and that pure crystalline materials are preferred over the pure amorphous because of their greater number of states and structural characteristics such as greater susceptibility to permutation. It will be interesting to see whether these conclusions remain after neutron displacement studies that are now being pursued with a “pure” C₁₂D₁₄ source.

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REFERENCES

[11] Power sources are presently available that could provide higher gradients than RF with 50 MHz rep rates e.g. see the IMRA America, Inc. website.
[12] The standard means for producing ingots of monocrystalline Si is the Czochralski (Cz) technique for which there appears to be no correlation between transmissivity and conductivity.
[15] UV grade, fused silica from CVI Laser Corp. was made by flame hydrolysis of silicon tetrachloride (SiCl₄) (Part# RAP-50-UV).
[16] Lithosil is a trademark of Schott Lithotec AG with ≤0.05 PPM of both Al and Na impurities. We have been unable to obtain this material. Infrasil 301 is a fused quartz product of CVI of interest with < 10 PPM of OH. A “pure” silica is also of interest.