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Refractive index of r-cut sapphire under shock pressure range 5 to 65 GPa

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High-pressure refractive index of optical window materials not only can provide information on electronic polarizability and band-gap structure, but also is important for velocity correction in particle-velocity measurement with laser interferometers. In this work, the refractive index of r-cut sapphire window at 1550 nm wavelength was measured under shock pressures of 5–65 GPa. The refractive index (n) decreases linearly with increasing shock density (ρ) for shock stress above the Hugoniot elastic limit (HEL): n = 2.0485 (± 0.0197) – 0.0729 (± 0.0043)ρ, while n remains nearly a constant for elastic shocks. This behavior is attributed to the transition from elastic (below HEL) to heterogeneous plastic deformation (above HEL). Based on the obtained refractive index-density relationship, polarizability of the shocked sapphire was also obtained. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4894854]

I. INTRODUCTION

The study of the variation of refractive index with pressure is of practical importance in science and technology. On one hand, the measurement of high-pressure refractive index has been used as a useful way to probe the electronic polarizability and band-gap structure.1,2 On the other hand, knowledge of the high-pressure refractive index of transparent optical windows is very important for velocity correction to particle-velocity measurements performed using laser interferometer techniques.3–5

To match the shock impedances of materials to be studied, many solid transparent materials, such as PMMA, fused silica, LiF, and sapphire, have been chosen as optical windows.6–9 Among these window materials, sapphire has the highest shock impedance, high strength, and good transparency, and therefore, its refractive index is of great interest in dynamic high pressure experiments.10–14 Setchell10,11 studied the refractive index of shocked c-cut sapphire at 532 nm and 633 nm wavelengths. The refractive index obtained at 532 nm wavelength shows a simple linear decrease dependence on density, in contrast to a more complicated dependence at the wavelength of 633 nm. Jones et al.12,13 measured the refractive index of sapphire subjected to uniaxial tension along the c-axis, as well as elastic shock compression along the a-axis. They found that the change of refractive index with density was quite different under compression and tension, and the a-cut sapphire also showed a linear decrease of refractive index with density. Jensen et al.14 recently reported the refractive index data at 1550 nm wavelength when shock compression was along the c-axis. However, these studies mainly focused on refractive index properties of a-cut and c-cut sapphire under elastic shock response, no data were published for shocks above Hugoniot elastic limit (HEL). Since sapphire is an important window material for high pressure shocks, we measured the refractive index of sapphire single crystals subjected to the shock pressure up to 65 GPa, and examined its electronic polarizability. Experimental method is shown in Sec. II. Results and discussion are shown in Sec. III, and the conclusion is presented in Sec. IV.

II. EXPERIMENTAL METHOD

Sapphire single crystal samples used for shock-wave experiments were grown via the Czochralski method, with purity determined to be better than 99.99%, provided by Anhui Institute of Optics and Fine Mechanics, Chinese Academy of Science. Its density at ambient conditions is measured to be 3.988 g/cm³ with the Archimedean method. Specimens were cut into cylindrical disks with their top and bottom surfaces parallel to the (1102) planes (r-cut), and polished to an optical finish.

In previous studies,11–14 symmetric plate-impact experiments were usually performed to measure the refractive index of transparent window materials, because they can simplify data analysis significantly with minimum uncertainties. However, this method is not suitable for high shock pressures owing to the brittle nature of sapphire. Therefore, two different experimental configurations, symmetric and asymmetric impact experiments, were designed to measure the refractive index of sapphire shocked below and above the HEL, respectively, as shown in Fig. 1. Related materials parameters are listed in Table I.

A. Shock pressures below HEL

The schematic of the symmetric plate-impact configuration is shown in Fig. 1(a). Impact loading was performed on a one-stage light gas gun. An aluminum film ~600 nm-thick was vapor deposited directly on the impact surface of both
sapphire flyer and sapphire target to reflect the probe light. A two-channel Doppler pin system (DPS) with picosecond resolution was used to measure the movement of the flyer and target. DPS was operated at 1550 nm laser wavelength and is similar to photon Doppler velocimetry (PDV). All fringe signals were recorded by a high bandwidth oscilloscope. Typical velocity profiles are shown in Fig. 2 for shot SA01. The velocity measured before time $t_0$ (black line) is the flyer velocity ($u_f$) before impact. After impact at $t_0$, shock wave propagates into the sapphire target as well as the sapphire flyer. However, due to an additional optical effect caused by the change in refractive index of the shocked layer, the velocity profile measured after $t_0$ is not the true particle velocity ($u$), but an apparent one ($u_{APP}$). When the shock reaches the free surface at time $t_1$, a free-surface velocity ($u_{FS}$) profile is observed. The fluctuations in $u_{APP}$ in Fig. 2 should be attributed to the noise in our DPS detection system rather than birefringence, because such fluctuations vary significantly from shot to shot even under similar shock pressure (shots SA02 and SA03).

![FIG. 1. Schematic of experimental setups for shocks (a) below HEL and (b) above HEL.](Image)

B. Shock pressures above HEL

For shock pressures above HEL, the experimental configuration was schematically shown in Fig. 1(b), and the impact loading was achieved with explosive-driven flyer plates. Copper or 304 stainless steel (304SS) disks were used as flyers to impact the sapphire targets. Flyer diameters were 95 mm or 200 mm, and their thicknesses were in the range 2.799 ± 0.001 mm to 4.040 ± 0.001 mm. To block out the light flash from trapped air, 304SS plates or permalloy foils were used as drivers. A 304SS film ∼1 μm thick was deposited on sapphire sample to reflect the probe laser light. A single-channel DPS was used to measure the interface and free-surface velocity at the front and rear surfaces of the sapphire window. In addition, the space between the sapphire free surface and the optical fiber holder was evacuated to below ∼10 Pa.

Typical results from experiment SA08 are shown in Fig. 3.

In the elastic regime, a single elastic shock wave propagates in the sapphire sample as shock pressure is below its HEL. According to Setchell, the refractive index can be related to the apparent velocity by an expression

$$n = \frac{u_{APP} - Dn_0}{u - D},$$

where $n$ and $n_0$ are the refractive index value behind and ahead of the shock front, respectively. $D$ is the shock wave velocity in the target frame. In our work, $u_{APP}$ is measured directly by DPS. $n_0$ is 1.7462 at 1550 nm. $D$ is determined from the sapphire target thickness $h$ and shock transit time $\Delta t$. Due to the symmetric impact configuration, the true particle velocity ($u$) is exactly one half of the impact velocity, namely, $u = w_f/2$. After determination of parameters $u$, $u_{APP}$, $D$, and $n_0$, the refractive index in compression, $n$, can be calculated using Eq. (1), which depends on both shock pressure and the probe laser wavelength of the DPS.

![FIG. 2. Velocity profiles measured from an elastic shock.](Image)

TABLE I. Related materials parameters for shock experiments.

<table>
<thead>
<tr>
<th>Shot No.</th>
<th>Flyer/Driver materials</th>
<th>Flyer/Driver thickness (mm)</th>
<th>Sapphire sample diameter/thickness (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>SA01</td>
<td>Sapphire/none</td>
<td>3.000</td>
<td>12.00/3.000</td>
</tr>
<tr>
<td>SA02</td>
<td>Sapphire/none</td>
<td>3.044</td>
<td>12.00/3.020</td>
</tr>
<tr>
<td>SA03</td>
<td>Sapphire/none</td>
<td>1.519</td>
<td>9.38/1.520</td>
</tr>
<tr>
<td>SA04</td>
<td>Sapphire/none</td>
<td>1.517</td>
<td>9.38/1.519</td>
</tr>
<tr>
<td>SA05</td>
<td>304SS/304SS</td>
<td>3.528/0.740</td>
<td>25.00/4.989</td>
</tr>
<tr>
<td>SA06</td>
<td>304SS/304SS</td>
<td>2.799/0.725</td>
<td>25.00/4.985</td>
</tr>
<tr>
<td>SA07</td>
<td>Cu/304SS</td>
<td>3.000/4.817</td>
<td>25.00/4.895</td>
</tr>
<tr>
<td>SA08</td>
<td>304SS/304SS</td>
<td>4.040/0.798</td>
<td>25.00/5.050</td>
</tr>
<tr>
<td>SA09</td>
<td>Cu/permalloy</td>
<td>3.000/0.070</td>
<td>25.00/5.035</td>
</tr>
<tr>
<td>SA10</td>
<td>304SS/304SS</td>
<td>3.554/0.650</td>
<td>25.00/4.980</td>
</tr>
</tbody>
</table>
Subscripts E and P refer to the elastic and plastic waves, respectively. According to Eq. (2), we should know the values of parameters $n_E$, $u_P$, $D_E$, and $D_P$, in order to obtain the value of $n_P$.

From the velocity profiles shown in Fig. 3, $D_E$ can be obtained as the sample thickness $h$ divided by the shock transit time of the elastic wave, $\Delta t_E$. Considering the interaction of plastic wave with the release of elastic wave, we calculate $D_P$ following Ahrens et al.\textsuperscript{18}

$$D_P = \frac{(\Delta t_P - \Delta t_E)(2D_E u_E - 2u_E^2 - D_{PE} D_E + D_{PE} u_E) + h(D_E - u_E + D_{PE})}{\Delta t_P (D_{PE} - 2u_E + D_E)}.$$  \hspace{1cm} (3)

Here, $\Delta t_P$ is the observed shock transit time of the plastic wave, $D_{PE} = D_E + u_{FSE}$, where $u_{FSE}$ denotes the particle velocity of the elastic wave on the free surface of sapphire, $u_E$ is true particle velocity of the elastic wave, which equals $u_{FSE}/2$.

For Al$_2$O$_3$ crystal shocked to below 100 GPa, the particle velocity of the plastic wave obtained from free surface velocity approximation method is comparable to that from impedance-match method, which results in a difference less than 1%.\textsuperscript{18} Thereby, $u_P$ is approximated as one half of the plastic wave velocity at the free surface $u_{FSP}$.\textsuperscript{19,20}

As shown in Sec. III, the refractive index of sapphire under elastic shock wave compression changes linearly with the density

$$n_E = a + b \rho_E.$$ \hspace{1cm} (4)

Here, the density $\rho_E$ satisfies the expression

$$\rho_E = \rho_0 \frac{D_E}{D_E - u_E}.$$ \hspace{1cm} (5)

Equation (4) is determined separately through pure elastic shock compression experiments. Hence, for inelastic shots with the two-wave structure measured, $n_E$ can be determined by the combination of Eqs. (4) and (5).

### III. RESULTS AND DISCUSSIONS

Experimental results are summarized in Table II. For each shot, shock pressure and density were calculated from the Rankine-Hugoniot equation based on the measured shock wave velocity ($D$) and particle velocity ($u$). Figure 4 shows the measured $D$-$u$ data. Since no $D$-$u$ relationship data of the r-cut sapphire is reported in the literature, we compare our result with those by Graham and Brooks\textsuperscript{19} for sapphire samples showed along an orientation close to the r axis (only 3° deviations), and an agreement is found (Fig. 4). For the elastic wave ($0 < u < 0.4 \text{km/s}$), the shock velocity increases slightly with increasing particle velocity. The best linear fit gives

$$D = 11.08(\pm 0.06) + 0.83(\pm 0.28)u.$$ \hspace{1cm} (6)

For the plastic wave ($u > 0.4 \text{km/s}$), the $D$-$u$ Hugoniot data can also be described by a linear relation

$$D = 6.89(\pm 0.03) + 2.26(\pm 0.24)u.$$ \hspace{1cm} (7)

The slope value of this linear relationship (2.26) is slightly higher than that of a-cut sapphire (1.95).\textsuperscript{20} This is an indication of anisotropy in shock response of single crystal sapphire.\textsuperscript{21}

The measured apparent and true particle velocities of r-cut sapphire under shock compression are illustrated in Fig. 5. Overall, the $u$-$u_{APP}$ relation obtained both below and above HEL can be well fitted by a power law

$$u = au_{APP}^b.$$ \hspace{1cm} (8)

Here, $a = 0.5287 (\pm 0.0047)$ and $b = 0.9609 (\pm 0.0081)$, respectively. Both $u$ and $u_{APP}$ are in km/s. Equation (8) can serve as the velocity correction function for the shocked r-cut sapphire, when it is used as windows in particle-velocity measurements.

We have examined the optical transparency of r-cut sapphire disks at shock pressures up to 90 GPa via optical extinction spectrum measurement using an in situ, shock-generated light source. It is found that optical extinction coefficient of this material is wavelength dependent and is sufficiently small at longer wavelengths. Our DPS system works at the 1550 nm wavelength. The r-cut sapphire at the 90 GPa shock pressure is transparent to the 1550 nm light. Therefore, it is suggested that this material can be used as a dynamic window of DPS (PDV) at shock pressures at least up to 90 GPa.

Figure 6 represents the measured refractive index as a function of density in the shocked state, together with the previous results for c-cut sapphire at 1550 nm.\textsuperscript{14} When shock pressure is below HEL, the refractive index of both r-cut and c-cut sapphire changes linearly with density. But different from the c-cut sapphire, a slight decrease was observed for

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**FIG. 3.** Velocity profiles measured from shot SA08.
TABLE II. Experimental results for sapphire.

<table>
<thead>
<tr>
<th>Shot No.</th>
<th>Shock wave</th>
<th>Shock velocity (km/s)</th>
<th>Apparent particle velocity (km/s)</th>
<th>True particle velocity (km/s)</th>
<th>Density (g/cc)</th>
<th>Pressure (GPa)</th>
<th>Refractive index</th>
</tr>
</thead>
<tbody>
<tr>
<td>SA01</td>
<td>Elastic</td>
<td>11.16 ± 0.53</td>
<td>0.220 ± 0.004</td>
<td>0.1248 ± 0.0004</td>
<td>4.034 ± 0.022</td>
<td>5.56 ± 0.27</td>
<td>1.7460 ± 0.0004</td>
</tr>
<tr>
<td>SA02</td>
<td>Elastic</td>
<td>11.23 ± 0.54</td>
<td>0.318 ± 0.066</td>
<td>0.1790 ± 0.0002</td>
<td>4.053 ± 0.003</td>
<td>8.02 ± 0.39</td>
<td>1.7457 ± 0.0006</td>
</tr>
<tr>
<td>SA03</td>
<td>Elastic</td>
<td>11.21 ± 0.26</td>
<td>0.335 ± 0.001</td>
<td>0.1841 ± 0.0001</td>
<td>4.056 ± 0.001</td>
<td>8.23 ± 0.19</td>
<td>1.7450 ± 0.0001</td>
</tr>
<tr>
<td>SA04</td>
<td>Elastic</td>
<td>11.49 ± 0.18</td>
<td>0.389 ± 0.002</td>
<td>0.2110 ± 0.0005</td>
<td>4.063 ± 0.001</td>
<td>9.67 ± 0.18</td>
<td>1.7445 ± 0.0003</td>
</tr>
<tr>
<td>SA05</td>
<td>Elastic</td>
<td>11.30 ± 0.17</td>
<td>...</td>
<td>0.325 ± 0.007</td>
<td>4.106 ± 0.004</td>
<td>14.66 ± 0.53</td>
<td>1.7442</td>
</tr>
<tr>
<td>Plastic</td>
<td>9.27 ± 0.55</td>
<td>1.767 ± 0.044</td>
<td>0.922 ± 0.011</td>
<td>4.399 ± 0.033</td>
<td>36.59 ± 1.61</td>
<td>1.7279 ± 0.0043</td>
<td></td>
</tr>
<tr>
<td>SA06</td>
<td>Elastic</td>
<td>11.29 ± 0.17</td>
<td>...</td>
<td>0.356 ± 0.004</td>
<td>4.118 ± 0.003</td>
<td>16.03 ± 0.41</td>
<td>1.7440</td>
</tr>
<tr>
<td>Plastic</td>
<td>9.40 ± 0.68</td>
<td>2.134 ± 0.052</td>
<td>1.102 ± 0.019</td>
<td>4.488 ± 0.046</td>
<td>43.81 ± 2.38</td>
<td>1.7214 ± 0.0061</td>
<td></td>
</tr>
<tr>
<td>SA07</td>
<td>Elastic</td>
<td>11.25 ± 0.17</td>
<td>...</td>
<td>0.274 ± 0.008</td>
<td>4.087 ± 0.005</td>
<td>12.27 ± 0.54</td>
<td>1.7446</td>
</tr>
<tr>
<td>Plastic</td>
<td>10.027 ± 0.56</td>
<td>2.461 ± 0.100</td>
<td>1.261 ± 0.018</td>
<td>4.549 ± 0.064</td>
<td>51.62 ± 3.09</td>
<td>1.7168 ± 0.0085</td>
<td></td>
</tr>
<tr>
<td>SA08</td>
<td>Elastic</td>
<td>11.44 ± 0.16</td>
<td>...</td>
<td>0.305 ± 0.002</td>
<td>4.097 ± 0.002</td>
<td>13.89 ± 0.29</td>
<td>1.7444</td>
</tr>
<tr>
<td>Plastic</td>
<td>10.07 ± 0.71</td>
<td>2.711 ± 0.038</td>
<td>1.374 ± 0.022</td>
<td>4.601 ± 0.056</td>
<td>56.69 ± 3.32</td>
<td>1.7106 ± 0.0057</td>
<td></td>
</tr>
<tr>
<td>SA09</td>
<td>Elastic</td>
<td>11.57 ± 0.15</td>
<td>...</td>
<td>0.271 ± 0.004</td>
<td>4.083 ± 0.003</td>
<td>12.49 ± 0.33</td>
<td>1.7446</td>
</tr>
<tr>
<td>Plastic</td>
<td>10.47 ± 0.88</td>
<td>2.897 ± 0.079</td>
<td>1.476 ± 0.005</td>
<td>4.603 ± 0.061</td>
<td>62.67 ± 4.54</td>
<td>1.7111 ± 0.0065</td>
<td></td>
</tr>
<tr>
<td>SA10</td>
<td>Elastic</td>
<td>11.29 ± 0.17</td>
<td>...</td>
<td>0.294 ± 0.001</td>
<td>4.095 ± 0.002</td>
<td>13.22 ± 0.23</td>
<td>1.7444</td>
</tr>
<tr>
<td>Plastic</td>
<td>10.36 ± 0.72</td>
<td>3.077 ± 0.034</td>
<td>1.566 ± 0.008</td>
<td>4.687 ± 0.055</td>
<td>65.64 ± 3.95</td>
<td>1.7074 ± 0.0042</td>
<td></td>
</tr>
</tbody>
</table>

**FIG. 4.** Shock velocity is shown as a function of the true particle velocity.

**FIG. 5.** True particle velocity ($u$) vs. apparent particle velocity ($u_{app}$).

**FIG. 6.** Refractive index is shown as a function of the shock-state density.

the r-cut sapphire. A linear fit to the refractive index data of r-cut sapphire in this pressure region will give

\[ n = 1.8207(\pm 0.0352) - 0.0186(\pm 0.0087)\rho. \]  

(9)

However, this refractive index-density relationship cannot be extrapolated to pressure above HEL. The refractive index obtained above HEL shows much more pronounced decrease linearly with increasing density

\[ n = 2.0485(\pm 0.0197) - 0.0729(\pm 0.0043)\rho. \]  

(10)

In principle, the refractive index depends on density and temperature. When shocked below HEL, sapphire deforms elastically, and shows a state of homogeneous temperature distribution. However, once the shock pressure is above HEL, sapphire will undergo a heterogeneous plastic deformation, and local hot spots may occur. The temperature of these hot spots could be several thousands of Kelvin higher than that of the surrounding bulk materials. The difference in deformation and temperature might be a cause for that in the refractive index below and above HEL.

It should be noted that, the overall behavior of the refractive index obtained in this work is similar to that of shocked MgO single crystal, but quite different from shocked LiF, which shows a linear increase of refractive index with increasing density. Previously, Waxler and Weir performed experiments to study the effect of hydrostatic pressure on the refractive index of solids materials, including NaCl, quartz, LiF, diamond, MgO, Al₂O₃, and
glasses. While most materials display an increase of refractive index with increasing pressure, diamond, MgO, and sapphire show the opposite, possibly due to their strong interatomic bonding. Our shock experiments are consistent with the hydrostatic experiments.

Refractive index can be used to examine polarizability behavior of materials via the Lorentz-Lorenz equation:

\[
\frac{n^2 - 1}{n^2 + 2} = K \rho P,
\]

where \(K\) is a constant and \(P\) denotes polarizability. Differentiating this equation results in:

\[
\frac{dn}{d\rho} = \frac{(n^2 - 1)(n^2 + 2)}{6\rho} \left[ (1 - \Lambda_0) + \rho \frac{dT}{d\rho} \right].
\]

Here, \(\Lambda_0 = -(\rho/P)(\partial P/\partial \rho)_T\) is a strain-polarizability coefficient, and \(\tau_0 = (1/P)(\partial P/\partial T)_\rho\) is a temperature-polarizability coefficient. In the case of shock compression, Eq. (12) can be written as:

\[
\frac{dn}{d\rho} = \frac{(n^2 - 1)(n^2 + 2)}{6\rho} [(1 - \Lambda')],
\]

where the shock-polarizability coefficient \(\Lambda'\) depends on density and temperature:

\[
\Lambda' = \Lambda_0 - \rho \tau_0 (dT/d\rho).
\]

Based on Eq. (13), the refractive index-density data measured in current experiments were used to evaluate the shock-state polarizability coefficient \(\Lambda'\). Figure 7 shows the variation of \(\Lambda'\) with shock pressure. \(\Lambda'\) above HEL shows a considerable linear increase with increasing pressure, in contrast to the nearly constant value below HEL. The polarizability of a material is related to strain. \(^{27}\) When shocked above HEL, sapphire undergoes a loss of shear strength and its strain behavior is different from the uniaxial strain below HEL, \(^{19}\) which results in the different shock polarizability response to increasing pressure. On the other hand, the strain-dependence of polarizability may help explain the observed shock-strength dependence of the refractive index.

**FIG. 7.** Polarizability coefficient vs. shock pressure. Dashed lines shown are guide to the eyes.

**IV. CONCLUSION**

The refractive index of r-cut sapphire under shock pressure up to 65 GPa was obtained. Although the change in the refractive index is extremely small under elastic shock compression, considerable linear decrease of refractive index with increasing density (shock pressure) is demonstrated for shocks above HEL. Such observations are similar to those on shock-compressed MgO, which is also a strong dielectric crystal. The sapphire polarizability is quite different for shocks below and above HEL. Our results suggest that dynamic deformation and interatomic interactions play an important role in the change of refractive index with pressure.

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