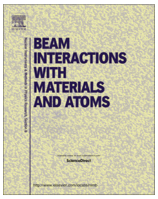
Nuclear Instruments and Methods in Physics Research B 325 (2014) 11–14

Contents lists available at ScienceDirect

Nuclear Instruments and Methods in Physics Research B

journal homepage: www.elsevier.com/locate/nimb



Measurement of thermal conductivity in proton irradiated silicon

Marat Khafizov [a,*], Clarissa Yablinsky [b], Todd R. Allen [a,b], David H. Hurley [a]

[a] *Idaho National Laboratory, P.O. Box 1625, Idaho Falls, ID 83415, USA*
[b] *Department of Engineering Physics, University of Wisconsin, Madison, WI 53706, USA*

ARTICLE INFO

Article history:
Received 10 October 2013
Received in revised form 31 January 2014
Available online 3 March 2014

Keywords:
Thermal conductivity
Ion irradiation damage
Extended defects
Thermal wave imaging

ABSTRACT

We investigate the influence of proton irradiation on thermal conductivity in single crystal silicon. We apply a laser based modulated thermoreflectance technique to measure the change in conductivity of the thin layer damaged by proton irradiation. Unlike time domain thermoreflectance techniques that require application of a metal film, we perform our spatial domain measurement on uncoated samples. This provides greater sensitivity to the change in conductivity of the thin damaged layer. Using sample temperature as a parameter provides a means to deduce the primary defect structures that limit thermal transport. We find that under high temperature irradiation the degradation of thermal conductivity is caused primarily by extended defects.

1. Introduction

Understanding the response of materials to irradiation damage is germane to an array of technologies ranging from electronic materials in deep space probes to structural materials in nuclear reactors. Irradiation by energetic particles (neutrons, ions and photons) changes the arrangement of atoms resulting in a significant change in material performance. For semiconductors, the thermal and electronic properties are most strongly influenced by irradiation damage. Examples include the use of ion irradiation to tailor electronic properties of semiconductors, and the degradation of thermal transport properties of oxide nuclear fuel caused by neutron bombardment [1,2].

Our emphasis here is to develop a technique that can measure degradation of thermal conductivity of nuclear fuel surrogates caused by charged particle irradiation. Proton and heavy ion irradiation can be used as an effective alternative to reactor neutron irradiation [3]. However, the irradiation damage caused by charged particles in the few MeV range is highly non-uniform with the damage zone extending only a few microns into the sample. This fact necessitates the use of a measurement technique that can provide micron order depth resolution. Here we present a thermoreflectance technique to measure degradation of thermal conductivity of a semiconducting material caused by charged particle irradiation.

The utility of modulated thermoreflectance techniques comes from the fact that the modulation frequency of the excitation source can be conveniently adjusted to confine the thermal wave to a thin layer at the surface of material [4]. The widely used time domain thermoreflectance (TDTR) technique has good potential for such applications [5,6]. TDTR requires deposition of a thin metallic layer. The primary role of this layer is that it introduces the thermal effusivity of the substrate into the boundary condition between film and substrate. The film also ensures strong optical absorption and for semiconductors issues associated with plasma waves are conveniently circumvented. However the film introduces a number of unknown parameters, such as film thickness and interface thermal resistance adding difficulty to the signal analysis required to extract the thermal properties of the substrate. To avoid this complication we follow a different approach that involves thermal wave imaging in the spatial domain on uncoated semiconducting substrates.

Silicon (Si) was chosen as a surrogate material for our laser-based measurement because it has optical properties similar to uranium dioxide, a standard oxide nuclear fuel used in light water reactors. Both uranium oxide and silicon are semiconductors with a bandgap in the range of 1–2 eV. While the thermal conductivities of these materials are different by an order of magnitude, it is the optical properties that determine how our laser-based measurement is implemented. It has been previously demonstrated that pump–probe thermoreflectance techniques can be applied directly to mechanically polished, bare silicon samples [7]. Surface defects from polishing promote fast carrier recombination (<1 ns). Thus for probe delays that are large in comparison to the carrier

---

* Corresponding author. Tel.: +1 2085261120.
E-mail address: marat.khafizov@inl.gov (M. Khafizov).

http://dx.doi.org/10.1016/j.nimb.2014.02.003
0168-583X/© 2014 Elsevier B.V. All rights reserved.

recombination time, the thermal and plasma waves can be completely decoupled.

2. Experimental

Schematic representation of our experimental approach is depicted in Fig. 1. It shows a thermal wave confined to the surface of the sample that is excited by an amplitude modulated pump beam. The frequency of the pump is tuned to confine the thermal wave to the damage region created by ion irradiation. Spatially resolved amplitude and phase profiles of the thermal wave are measured by recording reflectivity changes of the probe beam which is laterally scanned on the surface of the sample. In the absence of the electronic plasma wave the reflectivity change is directly proportional to the change in the surface temperature.

Our experimental setup has been previously described [7]. Briefly it uses a Ti:sapphire laser oscillator with a 100 fs pulse width. The 400 nm pump is obtained by doubling the laser output and the residual at 800 nm is used as the probe. Amplitude modulation of the pump pulse train is achieved using an acousto-optical modulator with modulation frequencies centered around 100 kHz. Both pump and probe are focused on the sample using a single 50× microscope objective resulting in a spot size of ~1 μm for both beams. Lateral scanning is achieved through an optical two lens lever described in detail in a previous paper [7]. The temporal delay between pump and probe is changed by varying the path length of the pump pulse using a mechanical delay line. The maximum transient temperature rise caused by a pump pulse is ~5 K, and the steady-state temperature rise is ~10 K. Small changes in reflectivity of the probe beam $\Delta R/R$ ranging from 10^{-5} to 10^{-3} are measured using lock-in amplification of the probe photodiode signal. Temperature dependent measurements were performed on a sample mounted inside liquid nitrogen cooled cryostat.

A boron-doped (p-type) (100) single crystal silicon wafer that has doping concentration of $2 \times 10^{16}\,\mathrm{cm}^{-3}$ is considered in this investigation. This wafer has been taken from the same batch used in previous work [7]. The wafer has already been polished by the vendor. One section of this wafer was subjected to additional mechanical polishing using a (3 μm) diamond slurry and then (0.05 μm) colloidal silica. This process significantly damages the crystal lattice in the near-surface region (~1 μm). This sample is then annealed at 300 °C to relieve the residual stress caused by polishing. A second section of this wafer was irradiated using NEC Pelletron® tandem accelerator by 1.6 MeV H^+ ions to a dose of 2.8×10^{15} ions/cm^2 at 700 °C. The damage profile was estimated using SRIM software calculations and is shown on Fig. 2 [8]. Two regions can be identified in the damage profile. The plateau region where the ions mostly undergo electronic interaction extends up to 30 μm has an estimated displacement damage of 0.2 dpa. The peak damage region resulting from nuclear stopping occurs at ~33 μm.

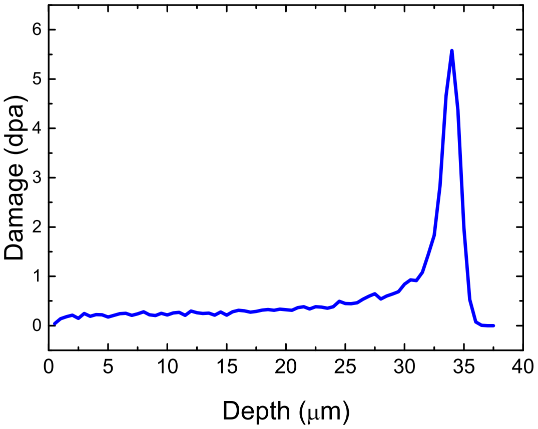


Fig. 2. Damage profile corresponding to 1.6 MeV proton irradiated silicon calculated using SRIM software.

3. Results

To test if irradiation damage enables a decoupling of the plasma and thermal wave components we conduct thermal wave scans at two different delays times. The thermal wave phase profile at $\Delta t = 14$ ps is shown in the top pane of Fig. 3. This profile can be decomposed into a plasma wave component that depends strongly on delay time [7], and a thermal wave component that is almost entirely independent of delay time [9]. The flat phase profile of the transient component has a spatial extent approximately equal to the pump spot size. Outside the excitation region the phase profile of the steady state component steadily decreases with increasing distance. Here the steady state component is to be associated with the thermal wave. The corresponding thermal wave amplitude at $\Delta t = 14$ ps is shown in the bottom pane of Fig. 3. The dip in the amplitude profile at $r \sim 1.5$ μm is due to a competition between the transient and steady state components that are approximately 180° out of phase. At large delays it is found that the transient component due to the plasma wave vanishes. This point is illustrated by the amplitude and phase response at $\Delta t = 13$ ns (realized using a small −100 ps delay) shown in Fig. 3. Based on these results and a previous study involving the mechanically polished sample we conclude that proton irradiation enables a decoupling of the plasma and thermal waves for large pump–probe delay times.

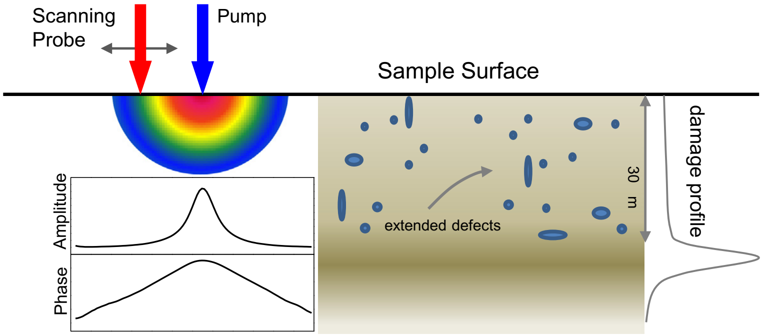


Fig. 1. Schematic representation of modulated thermoreflectance approach to measure thermal conductivity applied to ion irradiated samples.