Confined femtrosecond laser modification of Si: a new pathway to induce phase transformation

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Femtosecond regime laser induced modification of materials is a matter of fundamental interest in material science. With femtosecond laser pulses, absorption of the energy by the electronic system occurs entirely before thermalisation begins, that is before energy is transferred to the lattice. This can result in extremely high energy concentration and non-equilibrium responses by the material that cannot occur under longer time scale laser irradiation. Buy using high-NA optics and focusing deep below the surface it is possible to confine the high energy density by selectively absorbing the energy only subsurface. In insulators, focussing the laser allows otherwise weakly absorbed wavelengths to be absorbed by intensity dependant process only at focus. Semiconductors, such as Si, have smaller bandgaps than insulators and so achieving subsurface femtosecond laser modification in such materials requires unconventional methodology to achieve.

In this work, femtosecond laser modifications are induced deep subsurface in Si using two methods. The first is to induce the modification at the interface between Si, and an SiO_2 capping layer using a laser wavelength (below 1.1 m) that is only strongly absorbed by the Si. In this way, the capping layer provides confinement of the materials response. The second approach is to use a weakly absorbed wavelength (above 1.2 m) and a spherical sample such that the curvature of the surface matches the convergence of the laser beam when focussed at the centre of the sphere (fig. 1b), avoiding refraction induced defocussing (fig. 1a). In this way, the convergence angle of light within the sphere occur without spherical aberrations and remains large enough that intensity dependant absorption is restricted to focus, just as for insulators.

Characterisation of the modifications consists principally of Raman microspectroscopy and transmission electron microscopy (TEM). This too requires unconventional methodology as the modifications must be exposed from under 10s of microns to millimetres of material. This is achieved by mechanical polishing and focussed ion beam (FIB) milling guided by techniques including infrared microscopy. This reveals variation in density including, in the SiO_2 capped sample, a void surrounded by a high-density region (fig. 2). Within the high-density region are crystalline phases not previously observed by conventional high-pressure techniques, including st12-Si and bt8-Si. This clearly illustrates that confined subsurface laser modifications provides an avenue to investigate phase transformations that are not otherwise accessible.

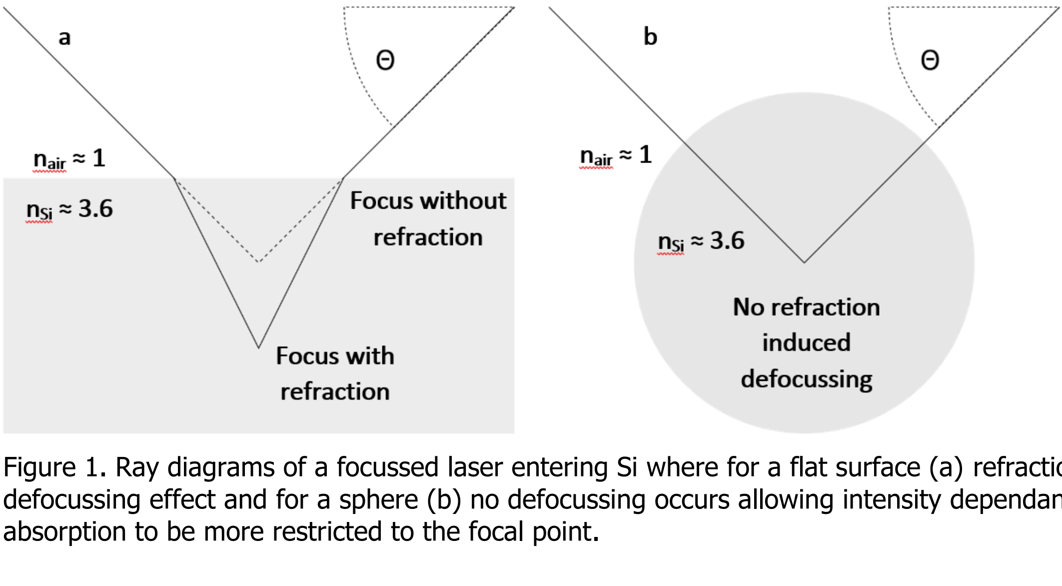

Figure 1. Ray diagrams of a focussed laser entering Si where for a flat surface (a) refraction causes a defocussing effect and for a sphere (b) no defocussing occurs allowing intensity dependant absorption to be more restricted to the focal point.

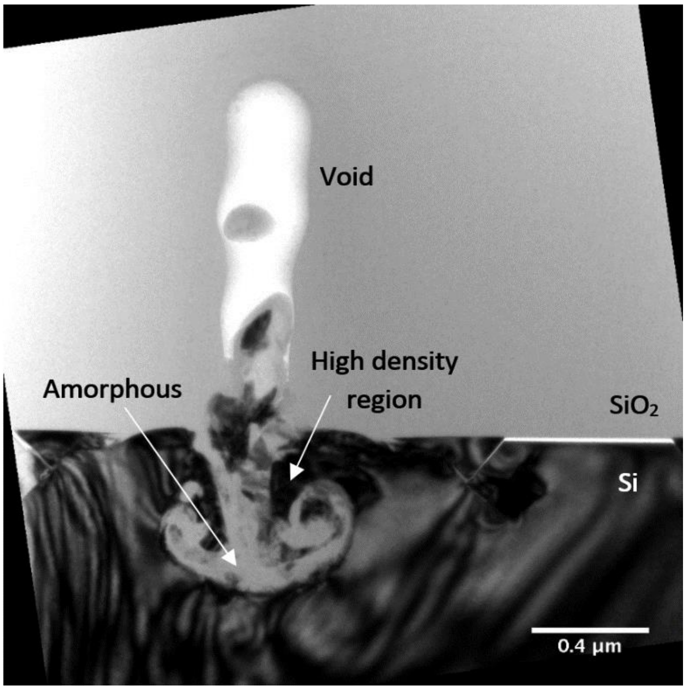

Figure 2. BF TEM of a microexplosion induced at the interface between Si and SiO_2. A void is visible extending from the interface up into the SiO_2. To either side within the Si is a highly compressed region containing high density crystalline phases.