Tuning the properties of graphene still presents a major challenge in graphene study. This challenge is evident in electronic applications that require high precision tasks.

In this study we present the laser-induced effects in single-layer graphene. Mechanically exfoliated monolayer graphene samples deposited on Si/SiO₂ substrates were irradiated with 532 nm green laser at 35 mW power for about 10 minutes under ambient conditions. The irradiated samples were characterized by time-resolved Raman spectroscopy, Raman mapping and Atomic Force Microscopy. Subsequently, the samples were transferred to Quantifoil TEM grids and the structure was investigated by aberration-corrected scanning transmission electron microscopy (STEM).

Our results show that the laser irradiation has locally modified the graphene surface and structure. The time-dependent Raman spectra of graphene undergo dramatic changes during the laser irradiation. However some of these changes disappear in time. Eventually we observe upshift in G mode position, slight increase in D mode intensity, decrease in 2D mode intensity and broadening of its line width. However Raman mapping images show significant D mode intensity.

There are no observable changes in the optical images after the laser treatment whereas in the AFM images certain structures are observed on modified regions.

We explain our results in terms of defect formation by breaking of the sp² C-C bonds, and formation of an additional layer of material on top of the graphene. This new layer is highly sensitive to electron irradiation and its nature remains to be clarified in more detail.

The results also show that the chemical reactivity of monolayer graphene is enhanced as a result of laser treatment.

The approach can further be utilized for local modification of properties of graphene as well as patterning it by laser-beam irradiation.
Fig. 1: AFM images after laser irradiation. a) Four modified spots on the monolayer graphene under the laser beam. b) Magnified image of (a). c) Height profile along the line shown in (b).

Fig. 2: STEM images of laser irradiated graphene. a) STEM image of the new layer on top of the graphene. b) and c) show the sensitivity of this new layer to electron beam.