

Graphene as an imaging platform of charged molecules

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Raman images of graphene at different pH buffer solution

Raman images of graphene films in different pH buffer solution were taken using Raman microscopy (Raman-11, Nanophoton Corp.). A laser beam emitting at 532 nm is used for the excitation. The laser beam was focused by an objective lens (100 \times , NA 0.9, Nikon) onto the graphene films. Laser intensity and exposure time were optimized to take Raman spectra with sufficient signal to noise ratio. Figure S1a-c show Raman images of graphene films at pH =4, 7, 9, respectively. The images are merged images from the image constructed G-band intensity, I_G (red) and 2D-band intensity I_{2D} (green). The area of high I_G (red area) indicates that double-layer graphene were locally formed. In addition, there were no Raman peak areas (black area), indicating that some parts of graphene were broken during the sample preparation process. Excluding no graphene area for each pH condition, the average G-band peak position (G_{pos}) were calculated from the Raman images, yielding 1591.0, 1590.3, and 1587.7 cm^{-1} for pH = 4, 7, and 9, respectively.

(a) pH 4



(b) pH 7



(c) pH 9



■ 2 μm

Figure S1. (a-c) Raman images constructed by I_G (red) and I_{2D} (green) of graphene films at pH = 4 (a), pH = 7 (b), and pH = 9 (c). The scale bar is 2 μm .

Transient response of G-band peak position and 2D-band peak position against pH change

A buffer solution on the graphene films was replaced from pH = 4, 7, 9, 7, to 4, sequentially, and the Raman images were taken every 10 minutes. The result is shown in Figure S2. The Raman images are constructed by G_{pos} with the same color range. The result clearly shows that overall the G_{pos} changes responding to pH changes. The G_{pos} shifted toward higher frequency with increasing pH and vice versa. The average G-band peak position (G_{pos}) were calculated from the Raman images and the result is plotted in Figure 3. In the same manner, 2D-band peak position ($2D_{\text{pos}}$) as a function of pH is also plotted in Figure S3.

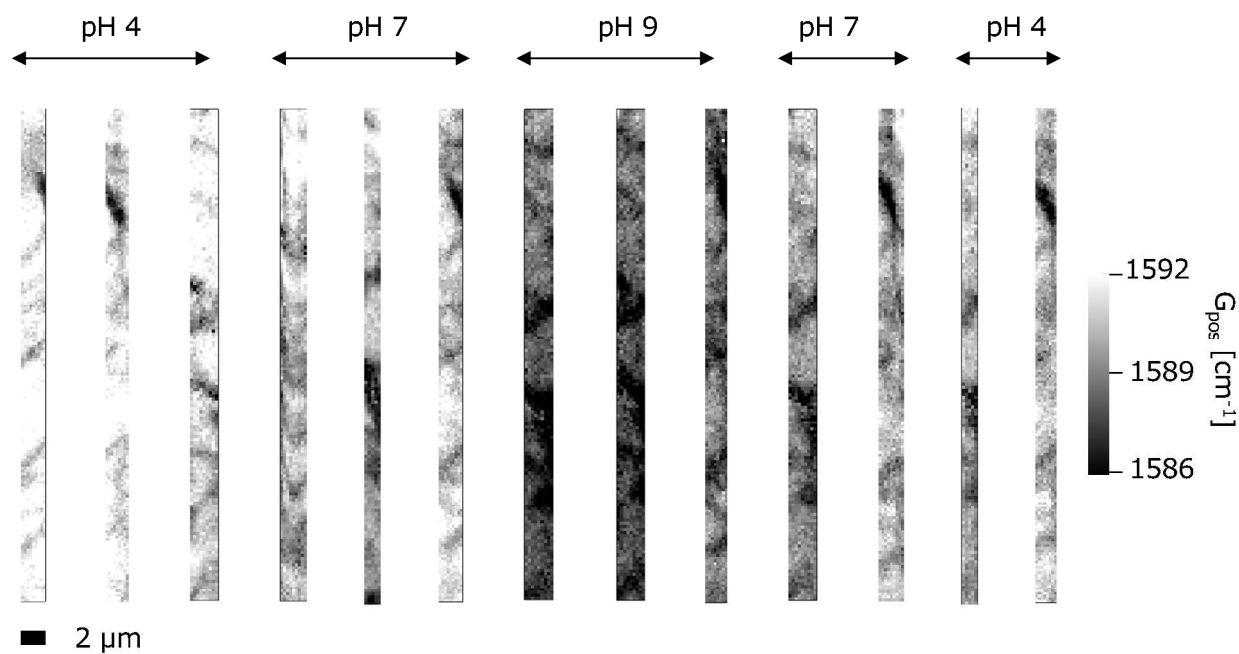


Figure S2. Raman images constructed by G_{pos} of graphene films at different pH from pH = 4 to pH = 9.

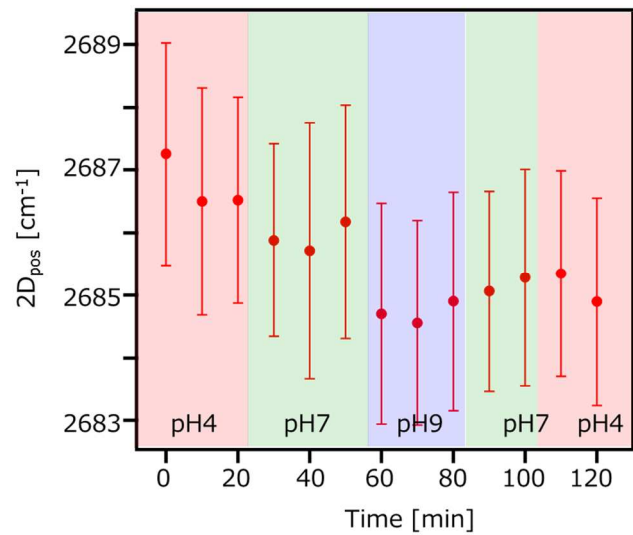


Figure S3. Transient response of $2D_{\text{pos}}$ against pH change.