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## Color generation via electron beam irradiating organic ice

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Natural colors are usually produced by light scattering or partial absorption of materials. Inspired by butterfly wings, a fashionable method for color generation, also called structural color, has been widely investigated and produced through various nanostructures, such as photonic crystals and plasmonic structures [1]. More commonly, multilayer planar thin-film coatings can be designed as Fabry–Perot cavities to modulate the reflectivity or transmissivity at visible frequencies, leading to colorful structures.

Here, we demonstrate a layered nanostructure for color generation by organic ice irradiated to focused electron beam. Our previous results have shown that organic molecules, e.g. anisole, condensed to form an ice film at low temperature can be utilized as a negative resist for electron beam lithography [2]. As shown in Fig. 1, a dedicated instrument consisting of a scanning electron microscope, a gas injection system, liquid nitrogen cooled cryostages, and a load-lock chamber is necessary to realize condensation, exposure and sublimation processes for organic ice [3]. During the electron beam irradiating organic ice (Fig. 2), the reaction probably begins with a first breakdown of a suitable bond, then molecule fragments react and build a network, solidifying fractions of the exposed volume. As the exposure continues, lighter elements from the reacted ice will be removed, thus rearranging the structure of exposed area. The finally deposited products of the exposed area are carbonaceous structures, which are non-volatile, chemically inert and very stable at ambient conditions [4].

Considering the optical property of anisole, its refractive index is usually 1.52 at visible region. In figure 3, the acceleration voltage of electron beam is 5 kV and area dose is 5 mC/cm<sup>2</sup>. The refractive index of anisole ice turns larger after electron beam exposure, such as 1.58 at 500 nm. This is consistent with the exposure process mentioned above, where lighter element, e.g. hydrogen, is released and the products contain a higher proportion of carbon. As refractive index of amorphous carbon is  $n_c = 2.55 + i0.02$  at 546.1 nm, it is reasonable to predict that the refractive index of anisole ice could be increased furthermore after larger dose exposure. That would pave a new way for color generation, since general structural color is modulated only by changing geometry.

Optical images of exposed anisole thin-films on silicon substrates under various doses have been shown in Fig. 4. The thickness is measured by AFM. It should be noted that the critical area dose at 5 kV is around 5 mC/cm<sup>2</sup>, so exposure using 15 mC/cm<sup>2</sup> is located at the saturation area in the contrast curve for anisole ice. That means the pattern thickness should not be changed within this area. Thickening of exposed film here is mainly attributed to bubble formation inside the film, which is evidenced by the blueshift of reflection spectrum (color).

Eager to investigate further the color generation and modulation by electron beam irradiating organic ice, we are now aiming at controlling bubble formation during electron beam exposure and achieving a wide range of refractive index tunability for organic ice.

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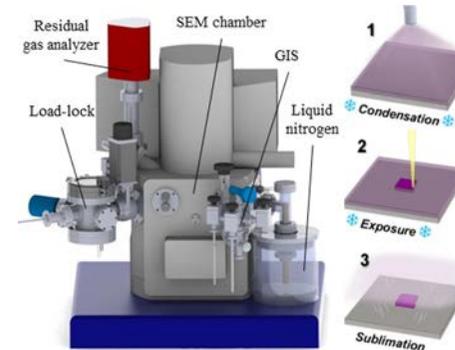


Figure 1. A dedicated instrument for organic ice resist processing.

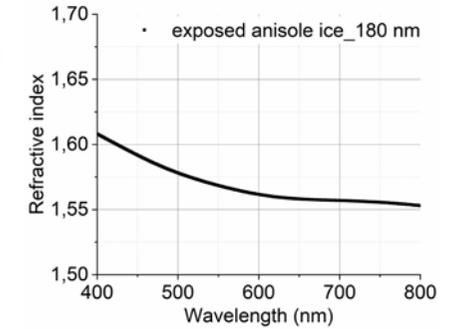


Figure 3. Refractive index of anisole ice.

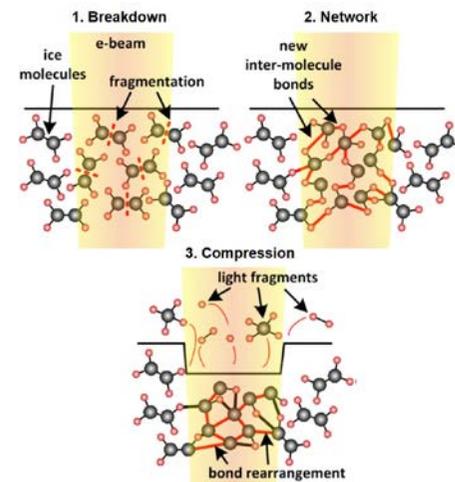


Figure 2. Multi-step model for electron beam irradiating organic ice.

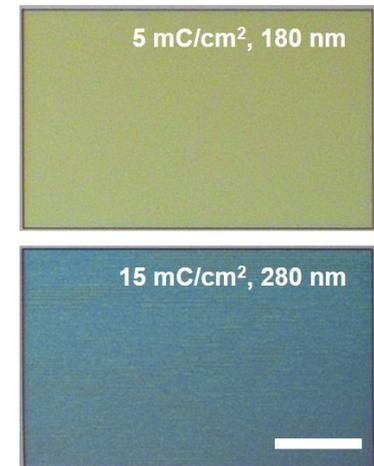


Figure 4. Optical images of exposed anisole thin-films on silicon substrates. Scale bar is 100  $\mu$ m.