

# Applications of Heavy Ion Irradiation in Photonic Crystal Research

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Photonic crystals (PC) have received extensive attention for the photonic band gap (PBG). The polystyrene (PS) particles bottom-up approach is a productive method for photonic crystal manufacture, this kind of photonic crystals having an unique PBG that depends on the particle's shape, sizes and defects. Heavy ion irradiation is a very useful method to induce defects in PC and change the shapes of the particles to tune the PBG. MeV heavy ion irradiation leads to an anisotropic deformation of the particles from spherical to ellipsoidal, the aspect ratio of which can be precisely controlled by using the ion energy and flux. Sub-micrometer PS particles were deposited on a Cu substrate and were irradiated at 230 K by using heavy ion energy and fluence in the range from 2 to 10 MeV and  $1 \times 10^{14} \text{ cm}^{-2}$  to  $1 \times 10^{15} \text{ cm}^{-2}$ , respectively.

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## I. INTRODUCTION

Self-assembly is an important method for fabricating photonic crystals because it is cheap and productive [1–3]. Photonic crystals have unique properties in terms of light propagation and are expected to have applications in optical devices because of their photonic band gap [4, 5]. Actual applications of photonic crystals require the photonic crystals with complete bands gap and controllable defects.

Photonic crystals usually possess fcc or hcc close-packed structures through self-assembling of spherical colloid particles by sedimentation or convective capillary flow. Photonic crystals built from non-spherical particles can lead to breakthroughs in realizing of 3D photonic band gaps [6], which is impossible for a close-packing structure. Though production of non-spherical particles has been reported [7], it is difficult for these non-spherical particles to aggregate into a well-ordered crystal structure by self-assembling. Ion-irradiation-induced anisotropic deformation is a useful method and has been investigated recently [8–11]. The method can deform the lattice of entire assembled structures. A focused ion beam (FIB) is a useful method for fabricating controllable defects in photonic crystals [12], but as is well

known, FIBs are expensive and unproductive.

According to the thermal spike model [13], the high electronic energy deposition and low directional straggle in the ion trajectory cause a cylindrically shaped narrow liquefaction region around the ion track. Plastic relaxation of the local shear stress induced by thermal expansion of this hot cylindrical region can result in plastic deformation perpendicular to the cylindrical axis. The diameter perpendicular to the ion beam direction increases and the diameter parallel to the ion beam direction decreases causing the spheres to become oblates. However, these research efforts were limited to inorganic substances, such as  $\text{SiO}_2$  and  $\text{ZnO}$ , and the deformation of organic substances had not been reported until our work was published [14].

Organic materials have unique physical and chemical properties compared with inorganic materials. In the fabrication of inverse opal PCs, colloidal particle assembly was used as a template and should be removed after filling the voids with high-refractive-index inorganic materials (for example,  $\text{TiO}_2$  with a refractive index of 2.6 was the most extensively studied material) [15,16]. In such cases, a polymer template was usually employed because it can be easily removed by using an organic solvent or can be burned off in an oven without damage of the inorganic skeleton [17,18].

In this paper, MeV Au, Cu and Si ions were used to bombard monodisperse PS particles. We have studied

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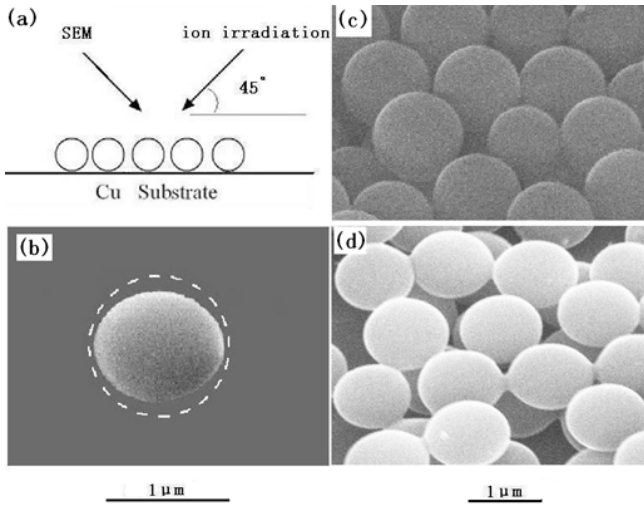


Fig. 1. (a) Derrection of ion beam and SEM observation, (b) shapes of PS particles before and after irradiation, in which the dashed sphere is the original perimeter of a PS particle, (c) and (d) SEM images of PS particles before and after being irradiated by 6 MeV Au ions at 230 K.

the relation of the anisotropic of deformation rate with the ion species, energy, and fluence.

## II. EXPERIMENTS AND DISCUSSION

PS particles of 1041 nm were dispersed in ethanol. Droplets of a dilute dispersion were dripped onto the clean surface of a Cu foil and were dried naturally in air for 24 hours. The samples were irradiated at 230 K by using the  $2 \times 1.7$  MV tandem accelerator at the Institute of Heavy Ion Physics, Peking University with a vacuum below  $10^{-4}$  Pa under an angle of  $45^\circ$  with respect to the surface of the Cu foil. The ion energy ranged from 2 to 10 MeV, with a fixed irradiation fluence of  $2 \times 10^{14}$   $\text{cm}^{-2}$ . At an energy of 6MeV the irradiation fluence changed from  $1 \times 10^{14}$   $\text{cm}^{-2}$  to  $1 \times 10^{15}$   $\text{cm}^{-2}$ .

The morphologies of the irradiated and non-irradiated particles were probed by scanning emission microscopy (Amary 1910FE SEM). These images were taken in the direction perpendicular to the irradiation beam to have an optimum observation of the deformation of spherical particles, as indicated in the schematic inset in Figure 1(a). Figure 1c and (d) show the SEM images of the 1041 nm diameter PS particles unirradiated and irradiated by 6 MeV Au ions with a fluence of  $2 \times 10^{14}$   $\text{cm}^{-2}$  at a temperature of 230 K. Figure 1(b) shows the shape changes of PS particles before and after irradiation, where the dashed sphere was the original perimeter of the PS particle. It clearly shows that anisotropic deformation of tested particles occurs after irradiation. The tested spheres were deformed into oblate ellipsoids. The extent of PS particle deformation could be qualitatively evaluated from the aspect ratio of the transverse

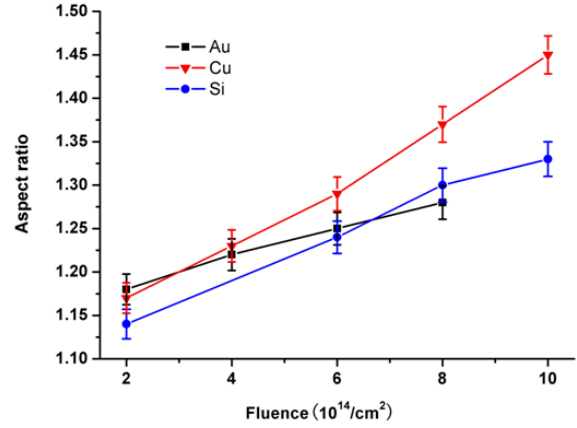


Fig. 2. Relation of the aspect ratio of 1041 nm PS particles with Au,Cu.and Si ions fluences.

diameter to the longitudinal diameter of the oblate ellipsoid.

In order to find the variation in the deformation rate with increasing ion fluence, 1041 nm PS particles were irradiated by 6 MeV Au, Cu, and Si ions at fluences from  $2 \times 10^{14}$  ion/ $\text{cm}^2$  to  $10 \times 10^{14}$  ion/ $\text{cm}^2$ . Because organic materials and inorganic materials have totally different physical and chemical properties, the deformation mechanism of PS particles under irradiation could not be invoked from that of  $\text{SiO}_2$  [13]. The aspect ratio changes of the 1041 nm PS particles as a function of fluence for 6 MeV Au, Cu, and Si ions are shown in Figure 2. During the ion irradiation, PS particles become smaller with increasing fluence while no volume changes was found for  $\text{SiO}_2$  particles as shown in Ref. 11. It should be noted that the shrinking process took place inhomogeneously, becoming larger at the initial stage and approaching a constant at the final stage. This may be caused by the gradual loss of gas atoms during the irradiation process and cross-linkage [16, 17]. In PS, the formation of hydroxyl (-OH), alkene (C=C) and alkyne (C-C) groups has been noticed with MeV heavy-ion bombardment [17], which means that besides the thermal spike model [13], a chemical change has to be considered.

Figure 3 shows the effect of ion electronic loss on the aspect ratio of PS particles irradiated by Au, Cu and Si ions with a fixed fluence of  $6 \times 10^{14}$  ions/ $\text{cm}^2$ . The aspect ratio of PS particles irradiated by Au ions is increasing fastest, and the influence of the lateral straggling on the deformation caused by ion irradiation has been proved [10] for destroying the anisotropism of the shear strain. The creep rate of the irradiated material depends on the orientation of the applied tension relative to the ion beam (anisotropic viscosity tensor); a small lateral straggling means that the shear stress caused by the electronic energy loss has better directivity.

Au ions have the smallest lateral straggling compared with Cu and Si ions; the lateral straggling of Au ions is about 70 nm under an electronic energy loss equal to

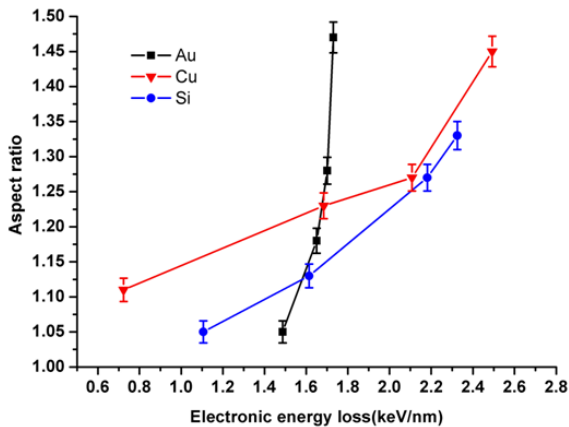


Fig. 3. Relation of aspect ratio of 1041 nm PS particles with ion electronic energy loss.

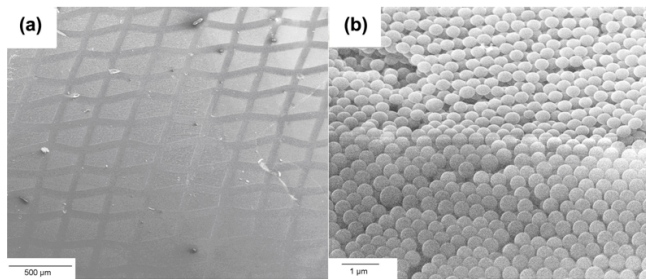


Fig. 4. SEM image of irradiated sample: (a) small magnification and (b) large magnification.

1.62 keV/nm, while the lateral stragglings of Cu and Si are 370 nm and 250 nm under the same electronic energy loss. Heavier ions have a smaller lateral straggling range compared with the light ions, which is beneficial for a more remarkable anisotropic deformation. Up to now the lightest ion was used to deform the particles is Ne ion [11].

A Cu grid with a hole size of  $200 \times 200 \mu\text{m}^2$  was used as a mask on the close-packed photonic crystal foil, as shown in Figure 4(a). The 6 MeV Au-ion irradiation was carried out with the sample cooled down to a temperature of 220 K and the fluence was up to  $4 \times 10^{14}$  ions/cm<sup>2</sup>. The SEM image showed an obvious boundary between the irradiated and the unirradiated areas; the area among the network is the irradiated area.

Figure 4(b) shows the details of the boundary between the irradiated and unirradiated areas, an obvious difference being observed. An anisotropic deformation was present for PS particles at the irradiated area, and the volume shrinkage of PS particles could be observed. It can be foreseen that some different patterns could be used as mask to make controllable defects in photonic crystals, which will possibly provide an optional choice for future optical devices.

### III. CONCLUSION

The fabrication of photonic crystals and the formation of controllable defects in PC are both important for applications. MeV heavy-ion irradiation leads to a shape change of PS particles from spherical to ellipsoidal, with an aspect ratio that can be precisely controlled by using the ion energy and fluence. A mask may possibly be used as means to position defects in photonic crystals.

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