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Annealing Effects on the Phase and Electronic Structure Evolutions of SiO Film by Electron Energy Loss Spectroscopy

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Composite films consisting of Si nanoclusters embedded in silicon oxide (also known as silicon rich oxide (SRO)) have attracted much attention due to their potential technological importance in the Si-based optoelectronic industry. The annealing process significantly affects the luminescence of the SRO films, causing major changes in the integrated luminescence intensity and the peak wavelength. The effect of annealing has been generally summarized as the generation of Si nanocrystals hosted in the SiO_2 matrix, followed by a nanocrystal growth process as the annealing temperature increases [1]. The luminescence has always been ascribed to the radiative recombination process in the Si nanocrystals or at the interface [2]. However, it has been found that even the low temperature (well below what would be expected to generate the Si nanocrystals) annealed SRO films give luminescence peak(s) in the visible range [3]. As the optical properties of the films are determined by the microstructure/electronic structure of the material, it is important to understand the films' structure change as a function of the annealing temperature.

In the present work [4], we have carried out a systematic study on the microstructure and electronic structure evolution of the as-deposited SRO film and the films annealed at temperatures ranging from 300-1100^{o}C, using combined transmission electron microscopy and electron energy loss (EEL) spectroscopy related techniques. We have found that the as-deposited SRO film is basically a single phase $SiO_{1.0}$, as suggested by its electronic structure characteristics disclosed by valence electron energy loss spectrum. Such single phase undergoes a continuous but incomplete phase decomposition to Si and SiO_2, indicating that the matrix of the Si clusters is not phase pure SiO_2, but a mixture of SiO_2 and SiO (i.e., SiO_x). The resulted Si phase first appears as small amorphous clusters, which continue to grow to larger sizes at higher annealing temperatures, but only crystallize at a critical temperature of ~800-900^{o}C. Such cluster/matrix configuration of the annealed SiO films is also consistent with the appearance of the interfacial plasmon in the EEL spectrum and its oscillation strength change with the annealing temperature. An interesting correlation between the films' phase/electronic structure evolution and the trend of their photoluminescence property change is identified.

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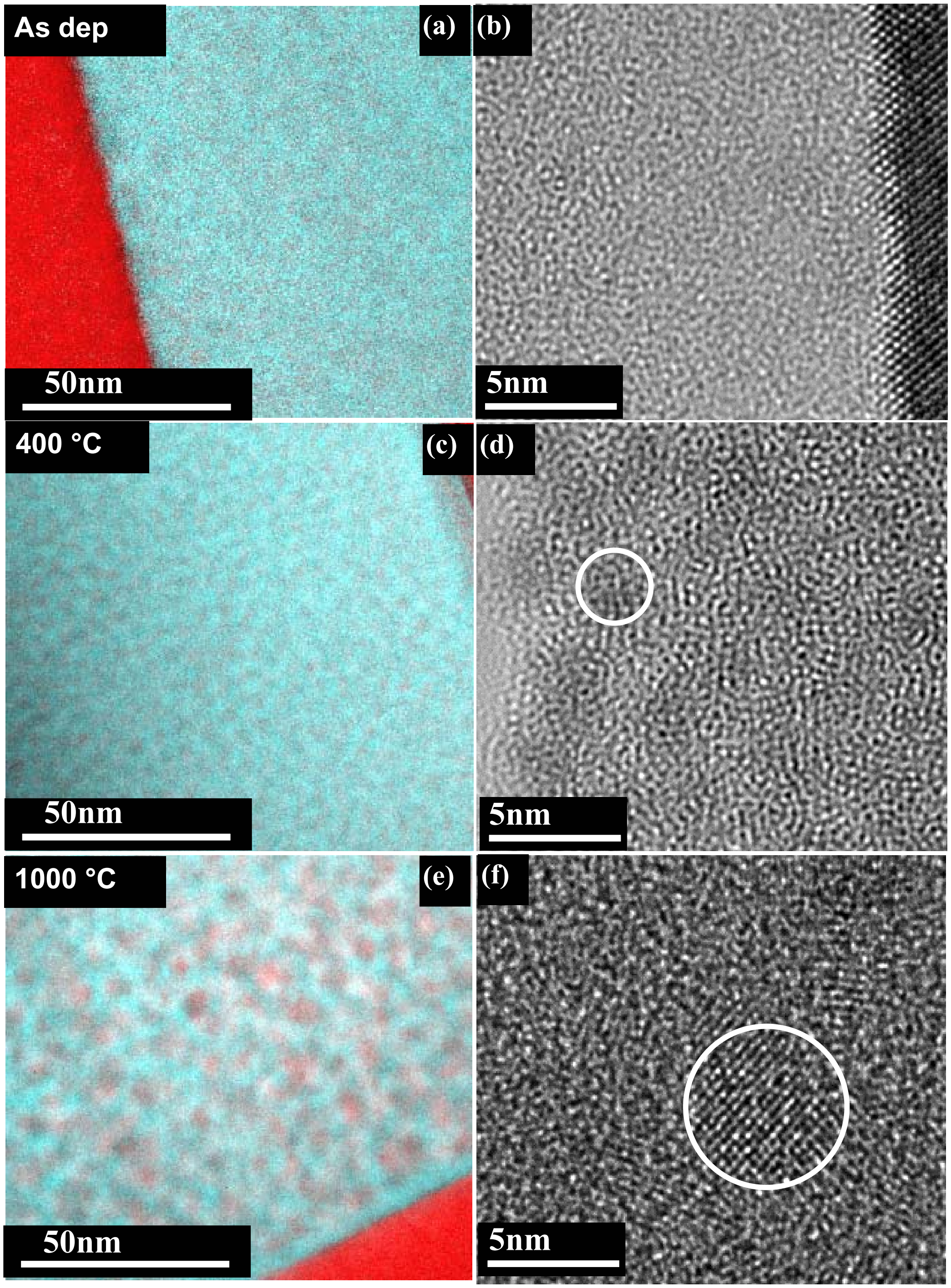

FIG. 1. Selected elemental maps and high resolution images of the SRO films annealed in different temperature. (a), (c), (e): The overlapped elemental maps of the as-deposited film and those annealed at 400 and 1000 ^{o}C, with Si in red and O in blue; (b), (d), (f): The corresponding high resolution images.

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