Structures of Self-Assembled Au Nanodot Superlattice

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Metal nanodots are widely investigated due to the unique applications in several fields such as single electron transistors, unprecedented catalystic activity, nonlinear optical switching, and immunoassay labeling. Self-assembly is the autonomous organization of components into patterns or structures without artificial intervention. Recently, many self-assembled two-dimensional (2D) array of metal nanodots were formed successfully. However, less attention was paid on the multi-layered structures of various outer-shell metal nanodots. In the present work, various outer-shell Au nanodots were deposited on Si(001) substrates. Displacement reaction of outer-shell of the Au nanodots was investigated to examine the change of low-angle X-ray diffraction (LA-XRD) patterns in the multi-layered Au nanodots.

5-nm-sized Au nanodots were prepared by a two-phase method. The reaction reagents were hydrogen tetrachloroaurate, tetraoctylammonium bromide (TOAB), dodecanethiol (DT), and sodium borohydride. Various Au nanodots are named Au@TOAB, Au@DT, and Au@TOAB-DT, respectively. The symbol @ herein represents a boundary between the inner-core of Au atoms and the outer-shell of protective organic molecules (TOAB or DT). Au@TOAB- DT represents the use of the Au@TOAB nanodots as a precursor to react with DT.

The various outer-shell Au nanodots show good 2D array structures on the amorphous carbon and Si substrates. From electron diffraction patterns of the Au nanodots, 6-fold symmetry was apparent near the central transmitted spot. In addition, the diffraction patterns from the atomic lattices of the Au nanodots exhibited ring patterns. Using the displacement reaction, the outer-shell TAOB of Au nanodots can be substituted by DT. Size distribution of Au@TOAB-DT nanodots was found to be more uniform than that of Au@DT. On the other hand, the LA-XRD patterns of multi-layered structures of Au nanodots, which were composed of various outer-shell molecules, were rather different. The Au@TOAB nanodots show an unusual one-dimensional diffraction behavior so that the values of l*d is constant (=1.87 nm herein, l: the order of reflections, d: the spacing of reflections). The values of standard deviation and coefficient of variation (CV) of rational series of reflections in the Au@TOAB samples were 0.018 and 0.094%, respectively. In additions, the values of full width at half maximum (FWHM) of the diffraction patterns of Au@TOAB samples are constant (0.17 degree). The unusual diffraction behavior of X- ray was previously found in the illite mineral. However, no one- dimensional diffraction behavior is seen in the Au@TOAB-DT and Au@DT nanodots, even though the 2D array can be formed. As a result, self- assembled multi-layered metal nanodots with an unusual one-dimensional diffraction behavior shall provide a new route of processing in the nanoscience.

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