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Phase formations and electronic excitation effects in alloy or compound nanoparticles

Hidehiro Yasuda[1*]

[1]Kobe University

The structural properties which are different from those in the corresponding bulk materials appear in nanoparticles which are the small manybody system, since the free energy changes extremely by the solute concentrations or atomic arrangements. The chemical free energy by the interaction between the different atoms, the strain energy due to the different atomic sizes and the interface energy play important roles in the structural properties of multicomponent nanoparticles, and their artificial control will make it possible to form specific structures. The spontaneous alloying, phase equilibrium and electronic- excitation- induced phase transitions in nanoparticles are reviewed based on the results obtained by in situ TEM.
Recently, it was found by that when GaSb particles were excited by 75 keV electrons, the compound transforms to a two-phase consisting of an antimony core and a gallium shell or an amorphous phase, or remains the original crystalline phase, depending on particle size and/or temperature. In the present paper, we studied by situ transmission electron microscopy the influence of defects induced by lower energy electronic excitation on the phase separation in GaSb particles.
Preparation of size-controlled GaSb particles was carried out with the use of a double-source evaporator installed in the specimen chamber of a TEM. Electronic excitation experiments and observations were carried out using the same microscope. The TEM used was Hitachi H-7000 operating at an accelerating voltage of 25 kV. The values of electron flux used for excitations were approximately $1.0x10^{20}$- $1.5x10^{21}m10^{-2}s10^{-1}$. The temperatures of the supporting films were kept at 293- 423 K. Structural changes associated with excitations were observed in situ.
When GaSb particles kept at 430 K were excited by 25 keV electrons, primary defects such as vacancies and gallium interstitials which are mobile at this temperature are formed in the particles. In the first stage, the vacancies form a void in the individual particles and in the second stage gallium interstitials segregate near the surface. From these results, it has been evident that when GaSb particles kept at 430 K are excited by 25keV electrons, two-phase separation takes place via void formation. In this case, it is considered that gallium atoms on the lattice points are displaced by the electronic excitation to form vacancies and gallium interstitials in the crystal. The vacancy concentration in the particle core is higher than that in the surface layer, but interstitial concentration increases toward the surface. Consequently, under the condition of vacancy supersaturation in the particle core the vacancy clusters will grow to form a void, and the subsequent surface segregation of interstitial clusters will bring about the separation to the two-phase structure.
Structural stabilities under excited states in nanometer-sized systems have been studied in GaSb compound nanoparticles. Electronic-excitation-induced phase transformations in nanoparticles have been studied by transmission electron microscopy. When GaSb particles were excited by electrons, the compound transforms to a two-phase consisting of an antimony core and a gallium shell or an amorphous phase, or remains the original crystalline phase, depending on particle size and/or temperature. It is suggested that such nonlinear responses of the phase transformations may arise from synergistic effects of bond instability, localized excitations, enhanced diffusivity, or

thermal equilibrium in reactions.

Keywords: nanoparticle, alloy phase formation, crystal growth, electronic excitation