It is well known that an autocatalytic process is necessary for the appearance of chemical oscillation. According to recent theoretical work on electrochemical oscillation, the autocatalytic process becomes to appear by coupling of the NDR in the $j$ vs. $U$ curve and the ohmic drop in the electrolyte. Therefore, it is very important to clarify the origin of the NDR for understanding the mechanism of the electrochemical oscillation. As shown in Fig. 1(a), the NDR in the present case appears only when the surfactant is added to the solution. On the basis of the result, the mechanism of the NDR can be explained as follows. The codeposition of Cu and Sn, in which the Cu deposition is dominant, should result in a negatively polarized surface owing to a difference in the electronegativity (or work function) between Cu and Sn as well as the principle of the equal Fermi level of electrons within a conductor. The negatively polarized surface induces the adsorption of the surfactant that is protonated and positively charged in acidic solution, which in turn suppresses the diffusion of metal ions and causes a decrease in $j$. Such a decrease in $j$ becomes stronger with negative potential shift, thus causing the appearance of the NDR. We are going to report more detailed mechanism of the self-organized formation of the layered nanostructures as well as the other experimental results.

**Fig. 1** (a) $j$ vs. $U$ curves in the presence (solid curve) and absence (dashed curve) of Amiet-320. (b) A Time course of current oscillation at $U = -432$ mV. Electrolyte: 0.15 M CuSO$_4$ + 0.15 M SnSO$_4$, and 0.6 M H$_2$SO$_4$ + 0.5 M citric acid + 0.5 mM Amiet-320.

**Fig. 2** Auger depth profiles of composition in growth direction of the film obtained under oscillatory condition.

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