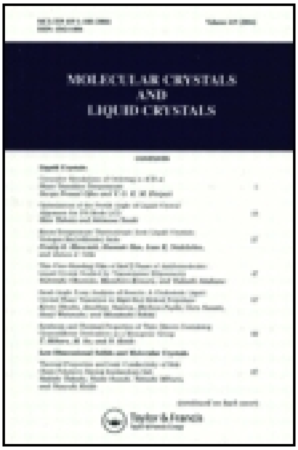


Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:
http://www.tandfonline.com/loi/gmcl19

Effect of 3-dimensional Stacking for Silver Nanoparticle Multilayers

Shuichiro Kuwajima [a], Yuji Okada [a], Yuji Yoshida [a], Koji Abe [a], Tomohiko Yamaguchi [a], Hiroshi Nagasawa [b], Nobutaka Tanigaki [a], Enji Sakurai [c] & Kiyoshi Yase [a]

[a] National Institute of Materials and Chemical Research, 1-1 Higashi, Tsukuba, Japan

[b] Osaka City University, 3-3-138, Sugimoto, Sumiyoshi-ku, Osaka, Japan

[c] National Research Institute of Metals, 1-2-1 Sengen, Tsukuba, Japan

Version of record first published: 24 Sep 2006

To cite this article: Shuichiro Kuwajima, Yuji Okada, Yuji Yoshida, Koji Abe, Tomohiko Yamaguchi, Hiroshi Nagasawa, Nobutaka Tanigaki, Enji Sakurai & Kiyoshi Yase (2001): Effect of 3-dimensional Stacking for Silver Nanoparticle Multilayers, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 370:1, 223-226

To link to this article: http://dx.doi.org/10.1080/10587250108030075

Mol. Cryst. and Liq. Cryst., 2001, Vol. 370, pp. 223-226

Effect of 3-dimensional Stacking for Silver Nanoparticle Multilayers

SHUICHIRO KUWAJIMA, YUJI OKADA, YUJI YOSHIDA, KOJI ABE, TOMOHIKO YAMAGUCHI, HIROSHI NAGASAWA[a], NOBUTAKA TANIGAKI, KENJI SAKURAI[b] and KIYOSHI YASE

National Institute of Materials and Chemical Research, 1-1 Higashi, Tsukuba, Japan, [a]Osaka City University, 3-3-138, Sugimoto, Sumiyoshi-ku, Osaka, Japan and [b]National Research Institute of Metals, 1-2-1 Sengen, Tsukuba, Japan

The multilayers structures of silver nanoparticles passivated with a fatty acid are investigated by means of X-ray reflectivity (XR) measurement. A distinct "quasi-Bragg reflection" peak appears at the position corresponding to the layer distance. From the calculation, it is found that the layer distance is slightly smaller than the diameter of the nanoparticle.

Keywords: X-ray reflectivity; nanoparticle; metal; multilayers

INTRODUCTION

X-ray reflectivity measurement, providing many informations (thickness, density and roughness), is a powerful method for structural analysis of thin films [1,2]. In spite of it potential, there are few reports for mono- and multi-layers of metal nanoparticles which are important roles for fundamental issues and practical applications [3]. The silver nanoparticles developed by Nagasawa *et al.* were found to form 2-dimensional hexagonally packing structure [4,5]. In this study, we investigate the layer structure of nanoparticle mono- and multilayers by XR measurement for the aim of controlling 3-dimensional nanoparticle arrangement.

EXPERIMENTAL

The silver nanoparticle passivated with tetradecanoate ($C_{13}H_{27}COO^-$) was prepared in the same way as described in previous papers [4,5]. As shown in Figure 1, to form a monolayer, the nanoparticles were dispersed in toluene and spread onto an air/water interface in a Langmuir-Blodgett trough NL-LB240S-MWC (Nippon Laser & Electronics Lab.). To accumulate the nanoparticle monolayer without collapse, the monolayer was compressed carefully with a slow compression speed. The monolayer was transferred onto a hydrophobized silicon (Si) wafer at room temperature by the horizontal lift method. The multilayers were prepared by repeating the transfer.

The reflectometer was connected to a X-ray generator of a rotating-anode type (target: Cu) and an Si 111 double-monochromator was installed on the tube wall. The special scintillation detector was used for the measurements, which were a wide dynamic range of 10^6 order for signal counting.

For the estimation of the layer structure parameters, calculations were carried out with MUREX program [6].

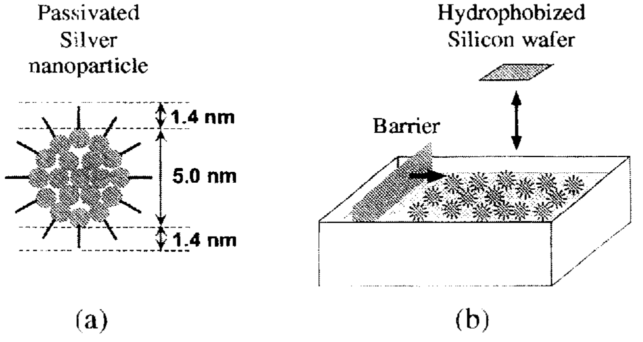
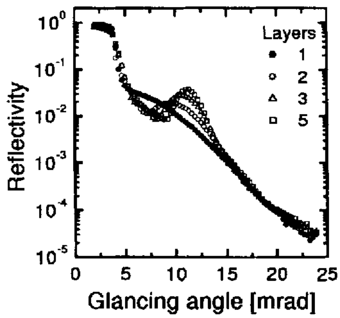
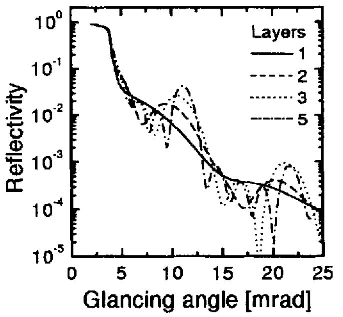


FIGURE 1 Schematic sketches of the passivated silver nanoparticle (a) and the procedure for level transfer (b).



(a)



(b)

FIGURE 2 Measured (a) and calculated (b) reflectivity profiles of the silver nanoparticle mono- and multi-layers.

RESULTS AND DISCUSSION

In Figure 2, measured and calculated X-ray reflectivity profiles of the silver nanoparticle 1, 2, 3 and 5 layers are shown. In multilayers, remarkable "Kiessig fringe" does not appear, however, a distinct peak is observed around 11.1 mrad. In the theory of X-ray reflectivity, it is known that quasi-Bragg peaks appear in the reflectivity profile of a periodic mutilayer.

In the calculation, we apply a simple layer model which is assumed to be three layers; the middle layer is composed of silver and hydrocarbonsons, the upper and lower layers are of only hydrocarbons, whose densities are 3.8 g/cm^3 and 0.29 g/cm^3, respectively. The density of middle layer is evaluated from the critical angle of XR profile, and that of hydrocarbon layer is assumed to be average density in the layer. In this model, the roughness of 30 % for layer thickness, which is due to the spherical shape of nanoparticle, is taken into account. In low angle region, both profiles are similar each other and a simple layer model seems to be adequate. This indicates that the nanoparticles form mono- and multi-layers structure without change its inherent shape. The periodic units corresponding to the distance between silver-core layers of 2, 3 and 5 layers are 7.5, 7.3 and 7.2 nm, respectively. In the multilayers, therefore, the periodic distance between upper and lower nanoparticle layers is slightly smaller than the diameter of the nanoparticle (7.8 nm) as shown in Figure 1 (a).

However, a remarkable decrease in reflectivity is observed in high angle region, because the effect of inhomogeneity of electron density in a layer for reflectivity seems to be not negligible at high angle. It is necessary to analyze the structure by more suitable model.

CONCLUSION

We have confirmed layer structures of the passivated silver nanoparticle multilayers by X-ray reflectivity measurement. A distinct quasi-Bragg reflection, indicating periodic layer structure in the nanoparticle multilayer, appeared. This is the first observation in the XR measurement of multilayers constructed only with nanoparticles. In the multilayers, the layer distance is slightly shorter than the diameter of nanoparticle.

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