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Composition dependence of band offsets for $(LaAlO_3)_{1-x}(Al_2O_3)_x$ gate dielectrics determined by photoelectron spectroscopy and x-ray absorption spectroscopy

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Introduction

$LaAlO_3$ is regarded as a promising candidate for high dielectric constant (high-*k*) gate dielectrics dielectrics in metal-oxide-semiconductor field-effect transistors (MOSFETs). In order to search for the optimum composition for the mixture of La_2O_3 and Al_2O_3, Fujitsuka *et al*. investigated the thermal stability and electrical properties for $(La_2O_3)_{1-x}(Al_2O_3)_x$ composite films (x=0 ~ 0.5)[1]. On the other hand, several results have been reported for Al_2O_3 rich side, namely $(LaAlO_3)_{1-x}(Al_2O_3)_x$[2, 3]. However, studies on band offsets have not been reported yet. In this study, we performed systematic investigation of band offsets in the composition range from $LaAlO_3$ to Al_2O_3 in order to propose the optimum composition for gate dielectrics.

Experimental

$(LaAlO_3)_{1-x}(Al_2O_3)_x$ composite films (x=0, 0.2, 0.33, 0.5, and 1) were prepared by the combinatorial pulsed laser deposition (PLD) method on clean *p*-type Si (100) substrates at the growth temperature of 300 °C using a KrF excimer laser. Ambient oxygen pressure during deposition was 10^{-6} Torr. The total thickness of each sample was set at 5 nm. Synchrotron radiation photoelectron spectroscopy and x-ray absorption spectroscopy (XAS) measurements were carried out at an undulator beamline BL-2C of the Photon Factory in High-Energy Accelerator Organization (KEK).

Results and discussion

Figure1 shows the values of bandgap (E_g), valence-band offset (VBO), and conduction-band offset (CBO) for $(LaAlO_3)_{1-x}(Al_2O_3)_x$ as a function of $LaAlO_3$-Al_2O_3 composition (x). The values of VBO and CBO for $LaAlO_3$ were determined to be 3.4 eV and 0.9 eV, respectively. These values are in good agreement with the previous ones theoretically obtained by Peacock *et al.*[4]. The values of VBO and CBO for Al_2O_3 are 4.7 eV and 2.0 eV, respectively. Although the value of VBO is in good agreement with the theoretical value, the value of CBO is rather smaller than the theoretical one. This is probably because the theoretical value of CBO was determined for Al_2O_3 bulk crystal. In fact, the reported experimental values of CBO for Al_2O_3 thin films were smaller than that of bulk Al_2O_3[5, 6]. The reduction of CBO in the Al_2O_3 thin film is consistent with our results. It should be noted that the CBO becomes smaller with increasing the ratio of Al_2O_3, while the VBO becomes larger. Since valence band of oxides mainly consists of O 2*p* states, the shift of the VBO can be related to the chemical shift of O 1*s* states. On the other hand, conduction bands of La_2O_3, $LaAlO_3$, and Al_2O_3 mainly consist of La 5*d** and Al 3*s**/3*p** states. Thus, the structure of the conduction band is significantly changed as a function of the ratio of La to Al. The shift of the CBO is, therefore, caused by both the effect of the chemical shift for core levels and the change in conduction-band structure. Since the values of VBO are sufficiently large in all compositions, the optimum composition of $(LaAlO_3)_{1-x}(Al_2O_3)_x$ in terms of band offsets are dominated by the value of CBO. Thus, $LaAlO_3$ (x=0) was determined to be the optimum composition in $(LaAlO_3)_{1-x}(Al_2O_3)_x$.

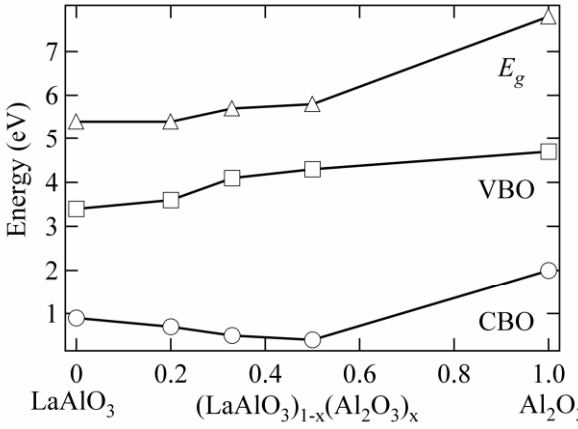

Fig. 1. Plot of bandgap (E_g), valence-band offset (VBO), and conduction-band offset (CBO) for $(LaAlO_3)_{1-x}(Al_2O_3)_x$ as a function of $LaAlO_3$-Al_2O_3 composition.

Reference

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