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An experimental investigation on the effective electron mass in $InGaO_3(ZnO)_3$ by valence electron energy-loss spectroscopy

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ABSTRACT

$In_{2-x}Ga_xO_3(ZnO)_m$ are reported to exhibit an extremely high field-effect mobility, and therefore considered as the potential channel materials for transparent field-effect transistor. To gain a deep insight into the high mobility of $In_{2-x}Ga_xO_3(ZnO)_m$, the effective electron mass, which is inherently related to the mobility of charge carriers as well as the density of states, is investigated by valence electron energy-loss spectroscopy. The effective electron masses in $InGaO_3(ZnO)_3$ nanobelts with the incident electron beam along the c axis and <1120> are determined as 0.130 m_0 and 0.132 m_0, respectively. These results critically checked by a comparison with calculated values and values measured by others. It not only well validates the results acquired from theoretical investigations, but also shed a new light on the isotropy of the effective electron mass in $InGaO_3(ZnO)_m$.

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1. Introduction

High-mobility oxides are critical in many applications from transparent electronics to photovoltaics [1–3]. Especially for transparent electronics, high-mobility oxides are the potential channel materials for transparent field-effect transistor (TFET), and considered as the basis for new optoelectronic devices [4,5]. A new homologous compound series in the system of In_2O_3–Ga_2O_3–ZnO, $In_{2-x}Ga_xO_3(ZnO)_m$ (m is integer), has been reported to exhibit an obviously high field-effect mobility of ~80 $cm^2 V^{-1} s^{-1}$ at room temperature, which is considered to be exclusive for their crystallographic structure consisting of a 'natural superlattice' with alternate stacking of In–O planes and Ga/Zn–O slabs [6]. This exceptional physical property has made $In_{2-x}Ga_xO_3(ZnO)_m$ promising and generate broad interest in various applications, e.g., high-resolution displays, thermoelectric enhancement, energy generation [7]. A critical factor leading to such high electronic mobility is a conduction band with a low effective mass. Given that, first principles calculations of effective mass have been carried out on $In_{2-x}Ga_xO_3(ZnO)_m$ to get a deeper insight into their high mobility [8,9]. Medvedeva et al. calculated the electronic properties of $InGaZnO_4$ employing first-principles full-potential linearized augmented plane-wave method (FLAPW) within the local density approximation [8]. $InGaZnO_4$ presented nearly the same effective electron mass in all crystallographic direction. This effect is due to the hybrid nature of the conduction band formed from the s states of all cations and the p states of the oxygen neighbors. The effective electron masses calculated using the simplified GGA + U method by Wen et al. also does not exhibit an obvious anisotropy in $In_2O_3(ZnO)_m$ with m = 1–3 [9]. However, the conductivity and mobility, which is inherently related to the effective electron mass, have been experimentally reported to show an obvious anisotropy in $In_{2-x}Ga_xO_3(ZnO)_m$ [10,11]. Thus it seems reasonable to respect an anisotropic effective electron mass in $In_{2-x}Ga_xO_3(ZnO)_m$. In looking for an explanation for this discrepancy, the quasi-isotropy of the effective mass in $In_{2-x}Ga_xO_3(ZnO)_m$ calculated from first-principles no doubt needs an experimental verification.

Experimental measurements of effective electron mass have been achieved by using techniques such as infrared spectroscopic ellipsometry and micro-Raman scattering for hexagonal InN [12], cyclotron resonance for n-type ZnO [13], and synchrotron-excited ultraviolet photoemission spectroscopy for TiN_x [14]. These methods usually need a combination of transmission, reflectivity, Hall- and conductivity measurements or various approximations, which can greatly simplify the evaluation of the effective mass, however must be handled with care because of their restriction to either certain physical quantities or certain materials. While the direct determination of the effective mass using a Kramers–Kronig analysis (KKA) of valence electron energy loss spectroscopy (VEELS) would be a favorable method in the case that the usually made approximations do not hold or the measurements of other properties like the transmission, for instance, is difficult. In addition, VEELS provides an effective way to estimate effective electron mass even on a nanometer scale [15,16]. When operated inside a transmission electron microscopy (TEM) or a scanning transmission electron microscope (STEM), it not only enjoys an excellent spatial resolution, but

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also enables the measurement correlated with local electronic structures, microstructure and chemical composition. By employing VEELS, Zhang et al. have obtained the effective electron mass in ZnO and ZnSe nanowires [15]. The effective electron mass in III–V semiconductor materials has also been determined by the same method by Gass and coauthors. The local variation of effective electron mass, which could prove to be useful for materials where the percentage composition of an element is too low to measure by current techniques such as core-loss EELS or energy dispersive X-ray analysis but has a large effect on its electronic properties (e.g. the dilute nitrides), is shown as a map with nanometer scale resolution [16,17]. These results give excellent agreement with the literature values, confirming the validity of VEELS in evaluating the effective mass. Although it is an appropriate method, no applications of the VEELS to $In_{2-x}Ga_xO_3(ZnO)_m$ have been reported so far. In our previous work, the high quality $In_{2-x}Ga_xO_3(ZnO)_3$ nanobelts were synthesized by a thermal evaporation process [18,19]. In this work, we use the KKA of VEELS and subsequent calculation to estimate the effective electron mass in $InGaO_3(ZnO)_3$ nanobelts and in particular to examine its (an)isotropy. Consequently certain orientation dependent features in the loss function and complex dielectric functions are successfully identified. Nevertheless, the effective electron masses along the c axis and in the ab plane of $InGaO_3(ZnO)_3$ nanobelts do not exhibit noticeable differences. These experimental results well validate the quasi-isotropy of the effective mass in $In_{2-x}Ga_xO_3(ZnO)_m$ calculated from first-principles. Furthermore, they are supposed to provide a comprehensive understanding of the high mobility in $In_{2-x}Ga_xO_3(ZnO)_m$.

2. Experiments

The $InGaO_3(ZnO)_3$ nanobelts are synthesized by a thermal evaporation process. Growth procedures have been reported in Ref [18,19]. The synthesized products were collected on 300-mesh copper grids, and then characterized by high-resolution TEM (HRTEM, Philips Tecnai F20). The EELS measurements are conducted in the diffraction mode with a beam voltage of 200 keV. The energy resolution of 0.8 eV is determined by the full width as half-maximum (FWHM) of the zero loss peak. The collection semi-angle is about 6 mrad. After spectrum acquisition, the zero-loss peak was subtracted from the raw spectrum and the plural scattering was removed by Fourier-log deconvolution. The KKA is used to derive the dielectric function [20].

3. Results and discussion

Fig. 1(a) shows a representative TEM image of an individual $In_{2-x}Ga_xO_3(ZnO)_m$ nanobelt. The nanobelt has a uniform width of ~100 nm. The EDX spectrum of that nanobelt is shown in Fig. 1(b). It consists of In, Ga, Zn, and O, except the Cu and C signals coming from the TEM sample grid and carbon adhesion film. According to the quantitative EDX analyses made on tens of the $In_{2-x}Ga_xO_3(ZnO)_m$ nanobelts, the atomic ratio of In/Ga/Zn is about 1:1:3, suggesting that the specimens under examination are $InGaO_3(ZnO)_3$ nanobelts. Fig. 1(c) shows an HRTEM image taken perpendicular to the wide surface of the nanobelt. The clearly resolved d-spacing between the two adjacent lattice fringes perpendicular to the nanobelt growth direction is close to 0.29 nm, corresponding to that of the {1010} plane. The corresponding selected area electron diffraction (SAED) pattern is shown in Fig. 1(d). It is along the [0001] zone axis with the intensity of the {1120} spots obviously stronger than that of the {0110} spots. It is an inherent feature of the layer structure of $In_{2-x}Ga_xO_3(ZnO)_m$ reported in previous studies [18,21]. To observe the exclusive layered structure of $In_{2-x}Ga_xO_3(ZnO)$, the nanobelt was tilted to make its lateral surface perpendicular to the incident electron beam. At this point, the HRTEM image observed is along the <1120> zone axis. As shown in Fig. 1(e), the superlattice periodicity is evidently along the [0001] direction, and the distance between the two adjacent InO_2^- layers is about 1.4 nm. Fig. 1(f) shows the corresponding SAED pattern. The weak satellite

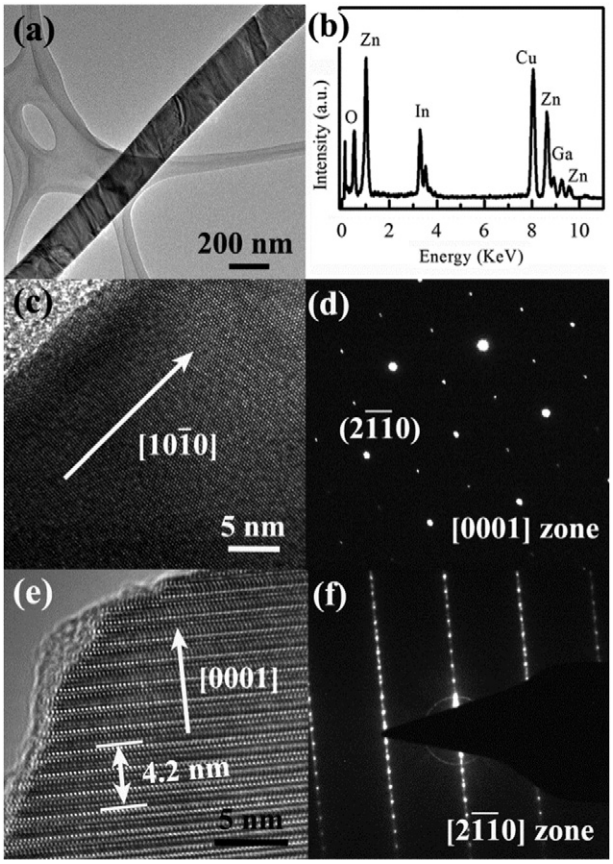

Fig. 1. (a) Low-magnification TEM image; (b) EDX spectrum; (c) HRTEM image projected along the c axis; (d) SAED pattern projected along the c axis; (e) HRTEM image projected along the <1120> axis; (d) SAED pattern projected along the <1120> axis of $InGaO_3(ZnO)_3$ nanobelt.

spots along the c-axis reconfirm the formation of $InGaO_3(ZnO)_3$ structure.

VEELS measurements were performed on the $InGaO_3(ZnO)_3$ nanobelt. Fig. 2(a) shows a comparison of the single-scattering distributions with the incident electron beam along the c axis (across the layers) and <1120> (within the layers), respectively. The spectra are quite similar with a dominant peak located at 18.6 eV. It represents the collective oscillation of the valence electrons in $InGaO_3(ZnO)_3$ nanobelt excited by the incident fast electrons. From the KKA, loss functions are extracted from the single-scattering distributions. Then, the second-order

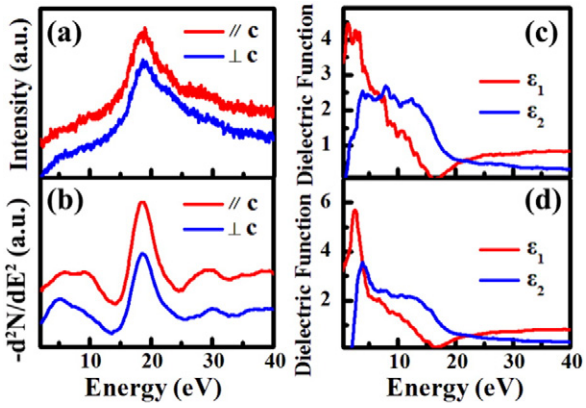

Fig. 2. (a) Low-loss energy spectra after being processed by the Fourier-log deconvolution for removing multiple scattering; (b) The corresponding second derivatives of the loss function; (c) and (d) derived dielectric functions respectively taken along the <0001> and <1120> zone axis of $InGaO_3(ZnO)_3$ nanobelt.

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