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Short communication

Composition and optical properties of amorphous Al-$MoSe_xO_y$ thin films and electrical characteristics of Al-$MoSe_xO_y$ field effect transistors

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ABSTRACT

Al-$MoSe_xO_y$ (un-doped and Al-doped $MoSe_xO_y$) films are prepared by radio frequency (RF) magnetron sputtering, and influence of the Al doping contents on the structure, composition and optical properties of the Al-$MoSe_xO_y$ films has been studied. We find that the amount of the Al can modulate the Mo^{4+}/Mo^{5+}/Mo^{6+} composition ratios and tune the optical band gap of the Al-$MoSe_xO_y$ film. The concentrations of the higher molybdenum oxidation states (Mo^{5+}, Mo^{6+}) increase, and the value of the band gap increase from 1.86 to 2.90 eV with the increment of the Al doping contents. Back-gated field effect transistors (FETs) have been fabricated and investigated by utilizing the sputtered Al-$MoSe_xO_y$ film channel. The incorporation of the Al makes the ambipolar FET device based on un-doped $MoSe_xO_y$ channel into p-type FET. P-type conductive FET based on 5.2% Al-$MoSe_xO_y$ channel has the highest field-effect mobility (55.2 $cm^2\ V^{-1}\ s^{-1}$) and preferable I_{on}/I_{off} ratio (~10^4).

1. Introduction

The interest of two-dimensional (2D) materials has been revived since the discovery of graphene due to its superb physical properties [1–3]. Nevertheless, the lack of natural energy band gap limits its development in electron devices [3]. Transition metal dichalcogenides (TMDs), also characterized by layered structure, have not only high mobility but also suited band gap and have been investigated extensively due to the applications in field effect transistors (FETs) [4–10]. Molybdenum diselenide ($MoSe_2$), as a member of the TMDs, has been also made into FET devices and investigated the device characteristics [11–13]. The field-effect mobility (~50 $cm^2\ V^{-1}\ s^{-1}$) and the I_{on}/I_{off} ratio (>10^6) of the n-type FET based on ultra-thin $MoSe_2$ flakes were obtained by Larentis et al. [11]. According to the research of Wang et al. [12], a back-gated FET based on crystalline $MoSe_2$ shows n-type channel behavior with average mobility of 50 $cm^2\ V^{-1}\ s^{-1}$. Lu et al. [13] have fabricated ambipolar back-gated FET using large-area few layers $MoSe_2$, while the mobility is only calculated to be 0.02 $cm^2\ V^{-1}\ s^{-1}$ for electron and 0.01 $cm^2\ V^{-1}\ s^{-1}$ for hole. From above mentioned, however, few p-type FETs based on $MoSe_2$ films with good device performance have been studied in recent years, and p-type FET is essential to complementary metal oxide semiconductor digital logic devices. With regard to 2D layered semiconductor, molybdenum trioxide (MoO_3), formed by double layers of edge-sharing MoO_6 octahedra and held together by weak van der Waals forces [14], has been used for developing the application of FET devices [15,16]. Research shows that [16] MoO_3 with high dielectric constant (>500) could minimize the coulomb scattering effect, which provides a solution for obtaining high mobility (>1100 $cm^2\ V^{-1}\ s^{-1}$). However, the I_{on}/I_{off} ratio of the fabricated MoO_3 FET is too low to make the device completely switch off. In view of the above-mentioned research status, it is feasible to combine $MoSe_2$ and MoO_3 to prepare $MoSe_xO_y$ channel material, which would make up the shortage of MoO_3 FET in I_{on}/I_{off} ratio and utilize the MoO_3 phase in the channel material to enhance the mobility of the FET device.

Many methods have been used to obtain the $MoSe_2$ films, such as chemical vapor deposition, sputtering, electro-deposition, chemical bath deposition, etc [17–21]. Among these techniques, magnetron sputtering has the advantage of high deposition rate, large area deposition and good uniformity. In this paper, Al-$MoSe_xO_y$ thin films are prepared by RF magnetron sputtering and

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influences of Al doping contents on structure, composition and optical properties of the Al-$MoSe_xO_y$ thin films have been studied systematically. Simultaneously, the current–voltage (I–V) characteristics of the Al-$MoSe_xO_y$ back-gated FET based on Al-$MoSe_xO_y$ channel are also investigated.

2. Experimental

Al-$MoSe_xO_y$ films were prepared on quartz and Si/SiO_2 substrates by BMS 450 RF magnetron sputtering system using $MoSe_2$ (99.95%) and Al (99.99%) for co-sputtering at room temperature. Ar (99.999%) and O_2 (99.999%) were used as sputtering gas, and the Ar/O_2 flow ratio was 12:1 (SCCM). The distance between the target and the substrate was fixed at 7 cm, and the chamber pressure was pumped down to 9.5×10^{-5} Pa. The RF power, sputtering pressure were 70 W and 0.3 Pa, respectively. In situ annealing for the as-deposited films was carried out after deposition in argon atmosphere for 30 min at 423 K. The selection of the above experiment conditions (vacuum background, RF power, sputtering pressure, temperature, etc.) are based on the previously studies of our teams [22,23]. To fabricate Al-$MoSe_xO_y$ FET, the first step was sputtering 150 nm Al-$MoSe_xO_y$ film on SiO_2 (270 nm)/Si (100) substrates. After that, for making the source, drain and back gate contacts, 100 nm of Au was deposited on the top and 200 nm of Ag was deposited on the back via vacuum evaporation. Cross sectional view of the structure of the back-gated Al-$MoSe_xO_y$ FET are shown in Fig. 3(a) (Inset).

X-ray diffraction (XRD) was conducted for structure analysis of the films with monochromatic Cu Ka radiation (40 kV, 45 mA, and $\lambda = 1.54178$ Å) on X' Pert Pro MPD X-ray diffractometer. UVISEL ER spectroscopic phase modulated spectroscopic ellipsometry system was used to measure the thicknesses of the films. X-ray photoelectron spectroscopy (XPS) measurements were performed for the chemical composition and states analysis using a monochromatic Al Kα (1486.6 eV) X-ray source. Optical transmittance of the films in the wavelength range of 300–1600 nm and current–voltage (I–V) measurements of the Al-$MoSe_xO_y$ FET were measured by the UV-3150 Shimadzu ultraviolet–visible–near infrared scanning spectrophotometer and Agilent semiconductor analyzer B1500A, respectively.

3. Results and discussion

To study the composition and chemical states of the Al-$MoSe_xO_y$ films, XPS analysis were performed on the samples. Tables 1 and 2 show summary of XPS analysis of valence states, peak position, and relative content of Al-$MoSe_xO_y$ films. XPS spectra of the films with different Al doping contents for (a)–(d) Mo 3d, (e) O 1s, (f) Se 3d and (g) Al 2p are shown in Fig. 1. As seen, the core-level binding energies (BEs) of Mo 3d, O 1s, Se 3d and Al 2p increase with the increasing of the Al doping concentration, which is due to the charge transfer and environmental charge density [24]. As shown in Fig. 1(a), the Mo 3d spectra of un-doped $MoSe_xO_y$ films consist of three spin–orbit doublets corresponding to Mo^{4+}–Se, Mo^{4+}–O and Mo^{5+}–O. BEs of Mo $3d_{5/2}$ for Mo^{4+}–Se, Mo^{4+}–O and Mo^{5+}–O are 228.5 eV, 229.2 eV and 230.5 eV, which is consistent with the previously reported results of $MoSe_2$ [23,25], MoO_2 [26] and Mo_2O_5 [27], respectively. Further, the oxide in Mo (IV) state of amorphous $MoSe_xO_y$ films only require a single doublet, which is consistent with amorphous oxide in Mo (IV) state of Baltrusaitisa's investigation [28]. For O 1s and Se 3d features of un-doped $MoSe_xO_y$ films (shown in Fig. 1(e) and (f) and Table 2), O 1s peaks at 529.7 eV [29], 530.2 eV [30] correspond to O–Mo^{4+} and O–Mo^{5+}, and Se $3d_{5/2}$ peak at 54.1 eV [25] corresponds to Se–Mo^{4+}, which further illustrates the chemical states of Mo cations. When Al doping contents increases to 3.5%, the concentration of Mo cations in the lower oxidation state (Mo^{4+}) decreases, and Mo cations in the higher oxidation state (Mo^{6+}) appear (shown in Fig. 1(b)). So the spin–orbit doublets are fitted into three corresponding to Mo^{4+}, Mo^{5+} and Mo^{6+}, and Mo^{4+} represents two chemical states for Mo^{4+}–Se and Mo^{4+}–O based on Fig. 1(e) and (f). The broad Mo 3d peaks at 232.1 eV and 235.31 eV for Mo $^{6+}$ $3d_{5/2}$ and Mo $^{6+}$ $3d_{3/2}$ agree well with the reported literature about MoO_3 [27,30]. O 1s peaks at 530.1 eV, 530.6 eV, 530.7 eV and 531.8 eV correspond to O–Mo^{4+}, O–Mo^{5+}, O–Mo^{6+} and O–Al^{3+} [27,31–33]. According to Fig. 1(g), Al 2p peak at 73.8 eV are consistent with the previous

Table 1
Summary of XPS analysis of valence states, peak position, and relative content of metal cations in Al-$MoSe_xO_y$ films.

		Mo^{4+}		Mo^{5+}	Mo^{6+}	Al^{3+}
		$3d_{5/2}$		$3d_{5/2}$	$3d_{5/2}$	2p
Un-doped	BE (eV)	228.5	229.2	230.5		
	Content (at.%)	15.2	11.9	5.4		
3.5% Al	BE (eV)	229.1		230.6	232.1	73.8
	Content (at.%)	14.5		6.5	6.3	3.5
5.2% Al	BE (eV)	229.6		230.7	232.2	74.2
	Content (at.%)	7.9		9.8	7.4	5.2
7.1% Al	BE (eV)	230.0		231.1	232.7	74.5
	Content (at.%)	4.7		10.1	8.3	7.1

Table 2
Summary of XPS analysis of valence states, peak position, and relative content of anions in Al-$MoSe_xO_y$ films.

		O–Mo^{4+}	O–Mo^{5+}	O–Mo^{6+}	O–Al^{3+}	Se–Mo^{4+}
		1s	1s	1s	1s	$3d_{5/2}$
Un-doped	BE (eV)	529.7	530.2			54.1
	Content (at.%)	23.9	13.1			30.5
3.5% Al	BE (eV)	530.1	530.6	530.7	531.8	54.4
	Content (at.%)	4.9	16.2	18.8	5.2	24.1
5.2% Al	BE (eV)	530.6	530.9	531.2	532	54.7
	Content (at.%)	3.6	24.4	22.0	7.8	11.9
7.1% Al	BE (eV)	530.8	531.4	531.7	532.4	55.2
	Content (at.%)	5.5	25.1	24.8	10.5	3.9