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Electron transport in the two-dimensional channel material - zinc oxide nanoflake

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ABSTRACT

ZnO nanoflakes of 3–5 µm in lateral size and 15–20 nm in thickness are synthesized. The nanoflakes are used to make back-gated transistor devices. Electron transport in the ZnO nanoflake channel between source and drain electrodes are investigated. In the beginning, we argue and determine that electrons are in a two-dimensional system. We then apply Mott's two-dimensional variable range hopping model to analyze temperature and electric field dependences of resistivity. The disorder parameter, localization length, hopping distance, and hopping energy of the electron system in ZnO nanoflakes are obtained and, additionally, their temperature behaviors and dependences on room-temperature resistivity are presented. On the other hand, the basic transfer characteristics of the channel material are carried out, as well, and the carrier concentration, the mobility, and the Fermi wavelength of two-dimensional ZnO nanoflakes are estimated.

1. Introduction

ZnO started to receive attention since 1930s for the determination of its crystalline structure [1,2]. ZnO is typically of hexagonal wurtzite structure and it is easily grown using versatile methods [3]. After grown, ZnO usually reveals natively *n*-type doped feature due to either hydrogens [4], zinc interstitials [5], or oxygen vacancies. It is a wide and direct bandgap semiconductor with an energy gap of ~3.37 eV at room temperature. It is photoactive and somewhat transparent, and it can be doped to be conductive. More particularly, it has a high exciton binding energy of ~60 meV thus it has a room-temperature lasing capability. ZnO is a multifunctional material [3] for many possible application fields such as transistor channel materials, optoelectronics, photodetectors, transparent conductors, and ultraviolet light-emitting diodes. It can be doped by magnetic ions to form diluted magnetic semiconductors for spintronic applications [6]. Moreover, it can be easily converted to nanostructures like quantum dots, nanowires, and nanosheets [7], and it can be used to make hybrid solar cells as well [8].

It is just the transition period from a three-dimensional to a two-dimensional channel for the design of transistors. It is that why recently two-dimensional (2D) semiconductors attract much attention [9]. Unlike conventional films having polycrystalline and granular surface features, current 2D semiconductor channel materials shall present crystalline structure with several or several tens of nanometers in thickness and atomically flat surface. ZnO does show those features owing to advances in synthesis methods in recent years [10–12]. It is commonly accepted that electron transport is in strong disorder regime in conducting channels with a 2D resistance $\rho_\square$ higher than 10 kΩ [13]. In this strong disordered regime, electron transport behaviors are adequately expressed by the model of Mott's 2D variable range hopping (VRH) [14]. There are several reports about Mott's three-dimensional VRH in either semiconductor or oxide thin films [15,16]. It is very difficult to observe Mott's 2D VRH in thin films because the thickness of the film is not as thin as the localization length which is typically several to several tens of nanometers.

In this study, we will investigate electron transport in 2D ZnO nanoflakes which have an average thickness of ~17 nm. The 2D transport nature will be discussed and the model of Mott's 2D VRH will be adopted to analyze ten different samples. The disorder parameter T_0 will be evaluated and given as a function of room-temperature (RT) resistivity. The localization length, hopping distance, and hopping energy of electrons in 2D ZnO nanoflakes will be presented as well.

2. Experimental

ZnO nanoflakes were synthesized using a sequential hydrothermal

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process on Si wafers. The details about the synthesis method and the characterizations of fundamental physical properties, such as the average thickness of ~17 nm, were published elsewhere [17]. The morphology and dimensions of ZnO nanoflakes were examined by field-emission scanning electron microscope (SEM, JEOL JSM-7001). The as-grown ZnO nanoflakes on Si wafers were transferred and dispersed on a heavily doped Si substrate, capped with 300-nm thick SiO_2 and patterned with wide current leads and large contact pads which can be contacted with probe pins in the cryostat. The standard electron beam lithography and thermal evaporation were adopted to make source and drain Ti/Au (20/100 nm) electrodes on the specified ZnO nanoflake in the working area connecting with pre-patterned current leads. ZnO nanoflakes were usually several micrometers in lateral size thus only two contact electrodes were deposited on their surface. The channel length between the source and drain electrodes was maintained to be 1.5–2 μm. The heavily doped Si substrate was employed as a gating electrode. The as-fabricated ZnO nanoflake devices were annealed in a high vacuum of $\sim 3\times 10^{-6}$ Torr at 550 °C for 24 h, thus the contact was improved and the RT resistance of the devices were decreased to one hundredth. The nanoflake devices were then placed on the sample stage of the cryostat (Variable Temperature Inert Cryostat, CRYO Industries of America Inc.) in 1-atm helium (99.99%). The temperature was controlled between 80 and 300 K by the LakeShore 340 and the resistance was measured in the voltage range from −0.2 to +0.2 V by the Keithley 6430 electrometer.

3. Results and discussion

Fig. 1(a) exhibits a typical SEM image of ZnO nanoflakes, showing a large aspect ratio between the lateral size and the thickness. It also implies the atomically flat feature of the surface of ZnO nanoflakes. The lateral size is about 3–5 μm and the thickness is about 15–20 nm with an average of ~17 nm [17]. The aspect ratio between the lateral size and the thickness is about 200. As-synthesized ZnO nanoflakes are randomly distributed on the Si wafer. The Si wafer with ZnO nanoflakes are pressed against our device substrate. The ZnO nanoflakes are then transferred on the heavily doped Si substrate. The nanoflakes are thereafter deposited with at least two current leads as source and drain electrodes. The top view SEM image of the ZnO nanoflake devices is displayed in the inset to Fig. 1(a). The resistance estimated from the current-voltage curves near zero voltage is analyzed to study electron transport in the channel material of the ZnO nanoflake. The RT resistivity ($\rho_{\square,\ \mathrm{RT}}$) of our ZnO nanoflake devices is higher than 10 MΩ that indicates a strong disorder regime. The Z10 device of $\rho_{\square,\ \mathrm{RT}} \sim 9.2$ MΩ is the nanoflake device of the least disorder. In the strong disorder regime, the resistivity described by the Mott's VRH model is [14].

$$\rho(T) = \rho_0 \exp\left(\left(\frac{T_0}{T}\right)^s\right), \tag{1}$$

where T is temperature, ρ_0 is a parameter of weak temperature dependence, T_0 is the disorder parameter, and s is the exponent depending on the dimension of the electron system. In order to check the dimension of the electron system in ZnO nanoflakes, another parameter $W(T)$ is defined as [16].

$$W(T) = -\frac{\partial(\ln(\rho(T)))}{\partial(\ln(T))}, \tag{2}$$

thus leading to a linear behavior between $\ln(W(T))$ and $\ln(T)$, and giving a slope equal to the dimensional parameter s. Fig. 1(b) presents the experimental data of $\ln(W(T))$ versus $\ln(T)$ for the Z10 device. It obviously reveals a linear dependence with the slope s of 0.34 very close to the ideal value (1/3) predicted by the Mott's 2D VRH model. The inset to Fig. 1(b) indicates the deposited electrodes on ZnO nanoflakes and the circuit connecting to the nanoflake device for electrical measurements. The source-drain current as a function of the back gating voltage is plotted in Fig. 1(c). It dictates a *n*-type gating behavior and the conduction band edge in the gate voltage range between 0 and 20 V. The electron mobility in the ZnO nanoflake is estimated to be $\sim 1.1\ \mathrm{cm}^2 / \mathrm{V\ s}$ and hence the carrier concentration of the Z10 device is $\sim 6.2\times 10^{11}\ \mathrm{cm}^{-2}$. The Fermi wavelength of ~32 nm is then obtained, which is longer than the thickness of the nanoflakes that corroborates again the charge carriers in a 2D system.

Fig. 2(a) presents temperature behaviors of resistivity of ZnO nanoflake devices in the temperature range from 80 to 300 K. The Z1 to Z10 devices have a large variation of RT resistivity from ~10 MΩ to ~1 GΩ whereas all devices show temperature dependent resistivity being well fitted by Mott's 2D VRH model. The fitting lines in Fig. 2(a)

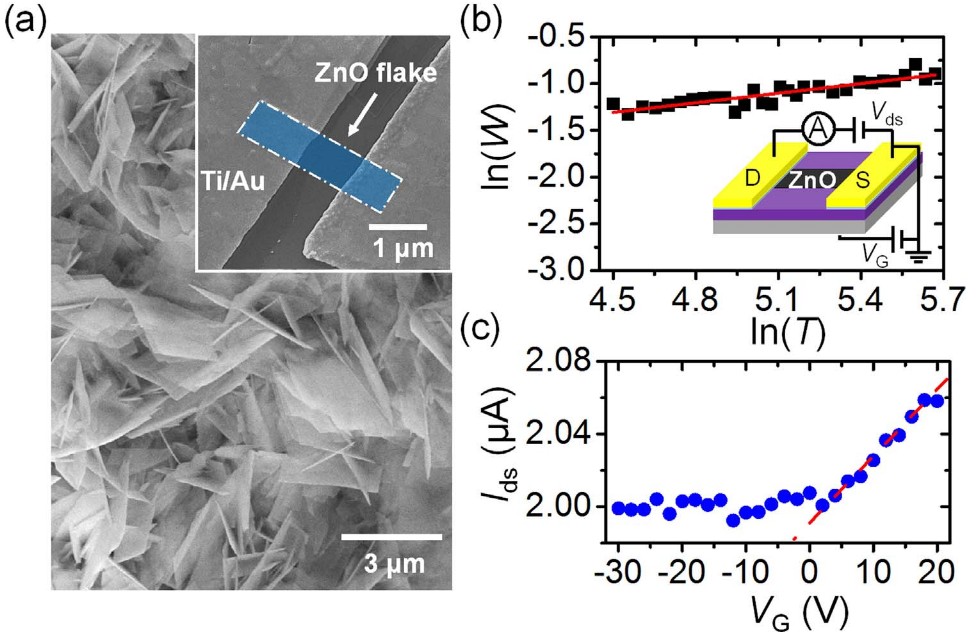

Fig. 1. (a) SEM image of as-synthesized ZnO nanoflakes with a scale bar of 3 μm. The inset shows a top view of the as-fabricated ZnO nanoflake device. (b) Plot of $\ln(W)$ as a function of $\ln(T)$ of the Z10 nanoflake device. The solid line delineates the best fitting result to give an exponent s of 0.34. The inset is a scheme presenting the source (S), the drain (D), and the back gate (G) electrodes and the connecting circuits. (c) Gating behavior of the ZnO nanoflake with a dashed line indicating the fitting to estimate electron mobility of $1.1\ \mathrm{cm}^2 / \mathrm{V\ s}$.