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Direct Contacting of 2D Nanosheets by Metallic Nanoprobes †

Filippo Giubileo 1,*, Francesca Urban 1,2, Alessandro Grillo 1,2, Aniello Pelella 1,2, Enver Faella 2 and Antonio Di Bartolomeo 1,2

1 CNR-SPIN Salerno Via Giovanni Paolo II 132, 84084 Fisciano, Salerno, Italy; furban@unisa.it (F.U.); agrillo@unisa.it (A.G.); apelella@unisa.it (A.P.); adibartolomeo@unisa.it (A.D.B.)
2 Physics Department “E. R. Caianiello”, University of Salerno, Via Giovanni Paolo II 132, 84084 Fisciano, Salerno, Italy; efaella@unisa.it
* Correspondence: filippo.giubileo@spin.cnr.it
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Abstract: We present a simple and fast methodology to realize metal contacts on two-dimensional nanosheets. In particular, we perform a complete characterization of the transport properties of MoS2 monolayer flakes on SiO2/Si substrates by using nano-manipulated metallic tips as metallic electrodes directly approached on the flake surface. We report detailed experimental investigation of transport properties and contact resistance in back-gated field effect transistor in which the Si substrate is used as the gate electrode. Moreover, profiting of the n-type conduction, as well as the high aspect ratio at the edge of the MoS2 flakes, we also explored the possibility of exploiting the material as a field emitter. Indeed, by retracting one of the metallic probes (the anode) from the sample surface, it has been possible to switch on a field-emitted current by applying a relatively low external electric field of few-tens of Volts for a cathode-anode separation distance below 1 µm. Experimental data are then analyzed in the framework of Fowler-Nordheim theory and its extension to the two-dimensional limit.

Keywords: two-dimensional materials; transition metal dichalcogenides; molybdenum disulfide; field-effect transistor; transport properties; field emission

1. Introduction

Molybdenum disulfide (MoS2) is one of the most investigated transition-metal dichalcogenides (TMDs) for exploitation in next-generation two-dimensional (2D) devices, including field-effect transistors [1–4], solar cells [5], photodetectors [6,7], field emission devices [8–11], chemical or biological sensors [12,13], etc.

MoS2 has a crystal structure characterized by a hexagonal layer of Mo atoms between two layers of S atoms. Layers are bonded together by van der Waals forces. MoS2 flakes can be fabricated either by mechanical exfoliation or chemical vapor deposition [14]. Bulk MoS2 has 1.2 eV indirect bandgap, while mono-layer (1 L) and bilayer (2 L) MoS2 have 1.8 eV and 1.6 eV indirect bandgap, respectively [15]. Consequently, both 1 L and 2 L MoS2 can be used to realize field-effect transistors with high On/Off ratio and photoresponse [16]. On the other hand, carrier mobility is typically limited to few-tens cm2 V−1 s−1. Moreover, ohmic contacts (with low resistance) are crucial to improving device performance [17].

In this paper, we demonstrate a simple method of realizing electrical contacts on MoS2 flakes by using nanomanipulated metallic probes inside a scanning electron microscope (SEM). We show that this technique allows complete characterization of the back-gated field-effect transistor (FET), as well as checking the field emission properties of the MoS2 flake.
2. Materials and Methods

The MoS₂ flakes studied in this work have been grown on Si/SiO₂ substrates by means of a chemical vapour deposition technique, in which S powder and a saturated ammonium heptamolybdate solution have been used as precursors. Few-layer MoS₂ flakes have been characterized by micro-Raman spectroscopy (λ = 532 nm). The experimental setup for electrical characterization is realized inside a SEM chamber (see Figure 1a) provided with two piezo-driven nano-manipulators for precise positioning (step resolution ~5 nm) of metallic probes (tungsten tips). A semiconductor parameter analyzer (Keithley 4200-SCS) is then used as a source-measurement unit, to apply bias up to ±100 V and to measure current with resolution better than 0.1 pA. Electrical measurements are performed at room temperature and in high vacuum (10⁻⁶ mbar) after gently approaching the tungsten tips on the MoS₂ flake (a real image taken inside the SEM chamber is shown in Figure 1b) and using the Si substrate as a back gate.

![Figure 1. (a) Schematic and real image of the nanomanipulators contacting the MoS₂ flake inside the SEM chamber. (b) SEM image of a contacted MoS₂ flake. (c) Raman spectrum of MoS₂ flake.](image)

Micro-Raman analysis of the MoS₂ flake has shown a spectrum (see Figure 1c) with two peaks corresponding to the E₂g and A₁g modes, separated by about 20 cm⁻¹, indicating that the sample under investigation is a monolayer.

3. Results and Discussion

In Figure 2a, we report the output characteristics (I_dS−V_dS) measured in the range of ±0.5 V for different values of the gate voltage (V_g). We notice a slight rectification that can be explained as the result of asymmetric Schottky barriers forming at the tungsten/MoS₂ interfaces [3]. By varying the distance between the two tungsten tips, we can modulate the channel length of the FET, thus realizing an experiment based on the Transfer Length Method (TLM) [18,19] to evaluate the contact resistance at the tungsten/MoS₂ interface. In Figure 2b, we show the measured total resistance R_tot versus d, with R_tot = 2R_c + \frac{R_0}{W}d, where R_c is the contact resistance, R_s is the MoS₂ sheet resistance, W is the channel width (assumed to be equal to the tip diameter, 200 nm), and d is the channel length, i.e., the separation between the two tips. Experimental data have linear behavior, from which specific area contact resistivity and sheet resistance can be evaluated as \( \rho_c \approx 4 \times 10^{-2} \Omega \cdot \text{cm}^2 \) and \( R_s \approx 10^6 \Omega/\square \) from the intercept and the slope of the linear fit, respectively.
The transfer characteristics ($I_{ds} - V_{gs}$) reported in Figure 2c have been measured for different gate voltage ranges up to ±60 V, with $V_{ds} = -5$ V, and by positioning the tungsten tips at separation of 13 μm. The device has n-type behavior, with a threshold voltage of about −10 V, and it can be explained in terms of chemisorption of oxygen on MoS$_2$ or sulphur vacancies [20–22].

From the transfer characteristic measured in the range ±50 V, we have estimated the on/off ratio as $\sim 10^5$, a subthreshold swing of $SS \approx 4 \frac{V}{\text{decade}}$, and a mobility of $\mu = 1 \text{ cm}^2 \text{V}^{-1} \text{s}^{-1}$, a value within the typical range (0.02 – 100 cm$^2$ V$^{-1}$ s$^{-1}$) reported for MoS$_2$-based FETs on SiO$_2$ [23,24]. The low mobility can be attributed to the high contact resistance and to high defects or traps density [25].

In Figure 3c, we show the transfer characteristics measured by sweeping the gate voltage different ranges, from ±20 V up to ±60 V. The curves have a clear hysteresis that we explain as being caused by negative charge trapping [20]. We observe that the hysteresis width ($H_{Vg}$), estimated at $I_{ds} = 0.1$ nA, has linear dependence on the $V_{gs}$ sweeping range (See Figure 3d). This behavior can be ascribed to the trapping process driven by the gate voltage and the effects on the MoS$_2$/Si-substrate capacitor.

Finally, we also investigated the field emission (FE) properties of the MoS$_2$ flake, profiting of the n-type conduction and the high aspect ratio of the flake side. By retracting the tip-anode at a distance $h = 900$ nm from the MoS$_2$ edge, we can measure the current emitted from the flake under the application of an external electric field (Figure 3a). More precisely, we applied a voltage bias of up to 120 V on the anode, and we measured the current emitted from the flake (cathode) with a resolution better than 0.1 pA. The current–voltage ($I_{ds} - V_{ds}$) curves have been measured at fixed cathode-anode separation $h$ and for two different values of gate voltage.
The FE characteristics have been measured by applying a bias voltage on the anode up to +120 V, by keeping a fixed gate voltage of 10 V and 40 V, respectively. The measured curves are reported on a linear scale (Figure 3b) and on a logarithmic scale (Figure 3c). Interestingly, we observe that the FE current is larger for $V_{gs} = 40$ V, suggesting that the gate voltage increases the n-doping of the MoS$_2$ flake [26].

We analyzed the FE curves in the framework of the Fowler-Nordheim (FN) theory [27], for which the FE current is expressed as

$$I_{ds} = A \left(\frac{\beta V_{ds}}{\Phi}\right)^2 \frac{S}{\Phi} \exp\left(-B \frac{\Phi^2}{(\beta V_{ds}/h)}\right)$$

where $A = 1.54 \times 10^{-6}$ A V$^{-2}$ eV and $B = 6.83 \times 10^7$ V cm$^{-1}$ eV$^{-3/2}$, $\Phi$ is the work function of the emitter, $S$ is the emitting surface area, and $\beta$ is the field enhancement factor. Accordingly, for FE curves, it is expected that $\ln(I_{ds}/V_{ds}^2)$ versus $1/V$ is linear (FN plot), and $\beta$ can be evaluated from its slope.

In Figure 3d, we report the FN plots that demonstrate the FE nature of the measured current. For $V_{gs} = 40$ V, we found a turn-on field $E_{on} = 40$ V $\mu$m$^{-1}$ (defined as the field to obtain a FE current of 1 pA) and $\beta \approx 200$.

4. Conclusions

We demonstrate a simple and fast methodology to realize metal contacts on two-dimensional nanosheets by gently approaching nanomanipulated tungsten tips inside a scanning electron microscope. We contacted a MoS$_2$ monolayer to form a back-gated FET, and we performed complete electrical characterization, reporting specific area contact resistivity of $4 \times 10^{-2}$ $\Omega$ cm$^2$, sheet resistance of $10^3 \Omega/\square$, on/off ratio of $10^5$, subthreshold swing of 4 V/decade, and mobility of 1 cm$^2$ V$^{-1}$ s$^{-1}$. Finally, by retracting the tip-anode, we performed field emission characterization of the MoS$_2$ flake, reporting that the FE current can be modulated by the gate bias.
Data Availability Statement: Data available on request.

Conflicts of Interest: The authors declare no conflict of interest.

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