Nanopages 7 (2012), 9–16
DOI: 10.1556/Nano.2010.00002

Electrical behavior of indium contacted graphene flakes

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Abstract

We studied the electrical transport properties of indium contacted graphene, few layer graphite (FLG) and bulk graphite samples under ambient conditions. Some of the contacted samples showed nonlinear transport characteristics. The initial electrical properties of the samples were changed reversibly by light illumination and irreversibly by argon ion irradiation. Ion irradiation modified the nonlinear I–V characteristics to quasi-linear ones in many cases. The resistance of a graphene sample showed positive temperature coefficient.

Keywords: graphene electrical properties, photosensitivity, indium contact, nanofabrication, nanotechnology

Introduction

Thanking to its outstanding physical properties such as high carrier mobility and large mean free path, graphene, the one-atom-thick single layer of graphite is one of the most promising electronic materials of the future. Contrary to carbon nanotubes, graphene sheets can be processed by conventional lithography [1, 2, 3], or atomic resolution, crystallographically defined STM lithography [4]. This makes graphene a potential successor of silicon in microelectronics and nanoelectronics if the reproducible production of large area graphene sheets can be solved. Smaller, technologically irrelevant thin graphite sheets with lateral dimensions of up to a few hundred microns can be prepared by mechanical cleaving of highly oriented pyrolytic graphite (HOPG) with adhesive tape [5]. For scientific purposes, these graphite flakes with single, double or few layer thicknesses are usually contacted individually by electron beam lithography in multistep, time consuming processes using polymer resist and solvents leaving presumably residues on the samples. The size of the mentioned graphite flakes is large enough to be well visible by optical microscopy. This gives rise to the development of maskless contacting methods [6, 7].

Here we report the investigation of the electrical properties of different graphene and FLG samples contacted with indium using the maskless method developed by Girit and Zettl [6]. I–V curves of the samples as prepared and Ar^+ ion irradiated samples were measured at room and elevated temperatures, besides, the effect of white light illumination on the resistance was studied.

Experimental

All graphene and FLG samples were fabricated by mechanical exfoliation ("scotch tape" method [5]) of HOPG and deposition on SiO_2 (90 nm)/Si substrates. The SiO_2 thickness was adapted to gain the maximal optical microscopy contrast between the graphene flakes with different thicknesses. The number of graphene/graphite layers can be estimated from the

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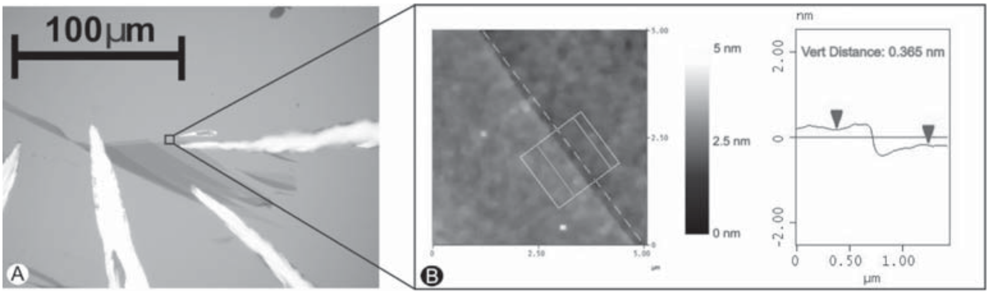

Fig. 1. Optical microscopy image of a thin graphite flake (sample b) contacted with three In solder spikes at places with different thicknesses (A). Tapping mode AFM image (left) and its line profile (right) of a single layer thick edge region of the same graphite flake before contacting (B)

Table 1. The parameters of the prepared samples. Two values separated by semicolon as number of layers mean varying thickness of graphite under the contact

Sample	Nr. of layers			Approx. contact distance [µm]
	contact 1	contact 2	contact 3	
a	1; 2	1; 2	–	22
b	5	2	1	48; 28; 41
c	1; 3	1	–	22
d	1	1; 6	–	22
e	1	1	–	9
f	1	1	–	7
g	2	1	–	6

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colour of the flake (Fig. 1A) [8]. Suitable selected flakes were measured by tapping mode atomic force microscopy (TAFM) to determine the number of layers [9], TAFM image acquired on the edge of a graphene flake is shown in Fig. 1B. In order to study the electrical behavior of the flakes, they were contacted by the method of Girit and Zettl [6]. This method uses a micromanipulator to deposit pure indium solder spikes on the desired positions under the optical microscope. In our case, the solder process was performed at 170 °C. By this technique, numerous structures were prepared, not only single and few layer graphite flakes of uniform thicknesses but pieces where the contacts were placed on positions with different graphite layer thicknesses. The parameters of the prepared samples are presented in Table 1. Due to the finite contact area, some contacts were placed on parts of the graphite flakes with different layer thickness. In these cases both values are presented in Table 1 as number of layers, separated by semicolon. Fig. 1A shows a flake of non-uniform thickness with three contacts, one of them on a single, another on a double and the third on a 5 layers thick part. As a comparison, similar In spikes were deposited on different parts of a macroscopic HOPG crystal. Besides, freshly cleaved HOPG crystal pieces were contacted with different metals as Ag, Al, Au, Cu and In by evaporation in 6.67 10^{-4} Pa background pressure in vacuum. The metal layers were evaporated through a mask onto the HOPG surfaces with varying contact lengths (see Fig. 2). The thickness of the evaporated layers was 150 nm.

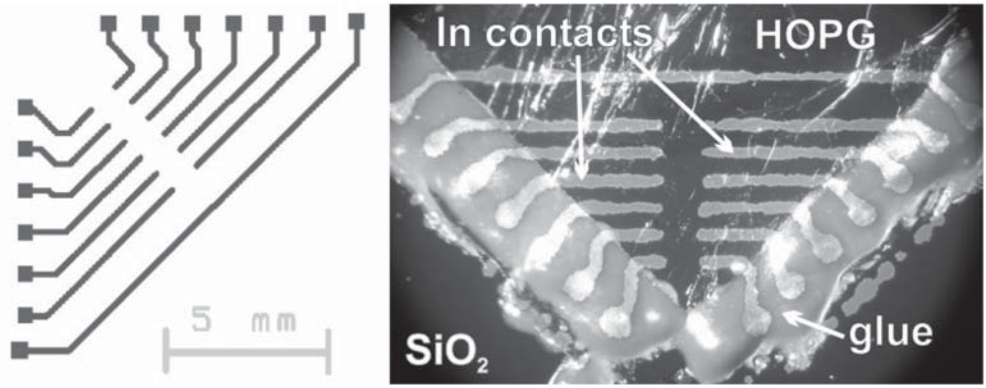

Fig. 2. The Al mask used for metal contact evaporation (left) and the evaporated In metal contacts on HOPG (right). The glue was an insulating epoxy material (Araldite)

The DC I–V characteristics of the prepared samples were measured by using a Keithley 224 programmable current source and a Keithley 617 programmable electrometer as voltmeter.

A Cole Palmer 41720-series illuminator white light source was used to illuminate the samples. The light was focused on the sample surfaces with a spot diameter of 15 mm. The light intensity was periodically changed from dark (0%) to 10 mW/cm^2 (100%) with a period of 50 seconds while a constant current of 1 mA was applied to the samples and the voltage was monitored. In order to estimate the thermal effect of illumination, one of the samples was illuminated with constant maximum intensity while the sample temperature was monitored. We found that the temperature equilibrated after 10 minutes at a 5 °C increase as compared to the ambient.

For the measurement of the temperature dependence of the resistance, the samples were placed on a controlled hotplate in ambient atmosphere. The sample temperature was monitored using an independent PT100 resistor thermometer. The effect of different temperatures in the range of 20 to 140 °C was measured.

Some of the graphene and FLG samples were irradiated with Ar^+ ions at 30 keV using a low dose of $D = 5\times10^{11}$ ions·cm^{-2}. The ion current density was $J = 0.04$ µA·cm^{-2} and the irradiation was done at normal incidence.

Result and discussion

All HOPG samples with 150 nm thick metal contacts showed ohmic characteristic independently from the contact material. The measured resistances can be expressed as the sum of the HOPG resistance (with constant 1 mm length) and the contact resistance between the HOPG and the contact metal. Measuring the resistances in case of varying contact length, the resistance of the HOPG can be determined by extrapolating the values of the sum resistances to the cases of zero contact length. This value was 1.6 Ω with this, the contact resistances for each metal can be calculated straightforwardly. The calculated values are presented in Table 2. Comparing the resistances of different metal contacts, Au, Cu, Ag and Al show similarly low values, while a four times higher resistance was measured in case of In.

Table 2. Resistance of different metals contacted HOPG crystals with different contact lengths

Method	Contact metal	Approx. contact distance [mm]	Total resistance [Ω]
PVD	Ag	1	2.90
	Al	1	3.28
	Au	1	3.12
	Cu	1	1,59
	In	1	8.30
Solder	In (spike)	0.077	0.45
	In (spike)	4.1	1.6
	In (spike)	2	1
	In (spike)	0.19	0.64

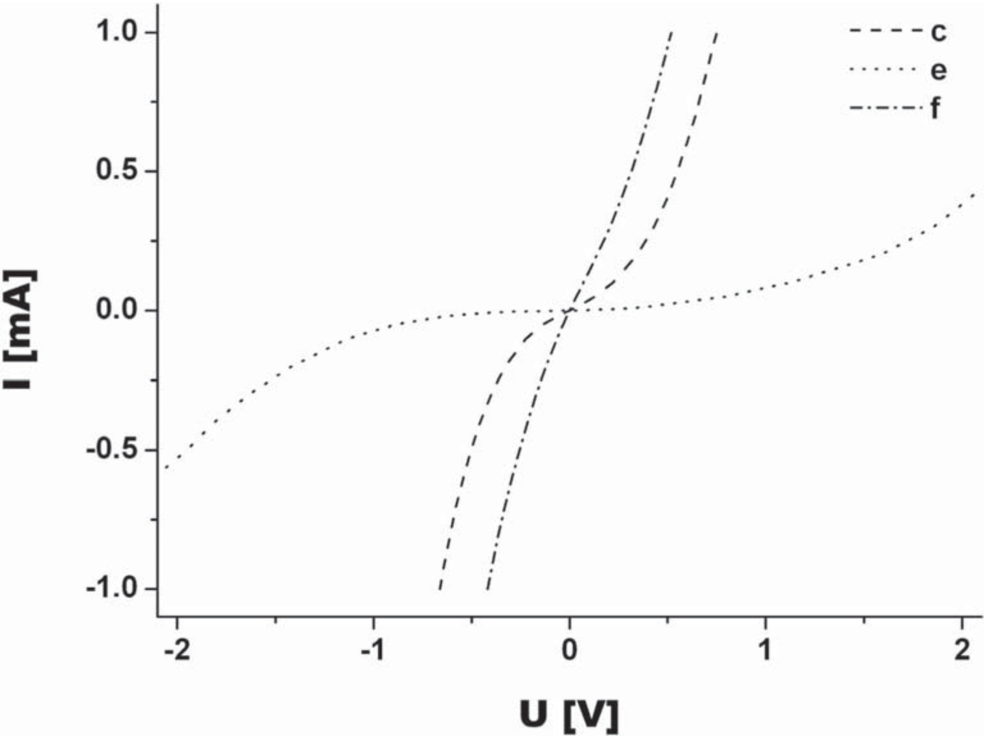

Fig. 3. Some selected, typical I–V curves of different graphene and FLG samples

HOPG samples contacted with In spikes showed ohmic behavior, similarly to the case of evaporated In contacts, but the resistance was lower than in the case of evaporated indium.

The I–V characteristics of spike contacted graphene and FLG samples showed varying behavior. All characteristics can be regarded as linear in the low current range, up to about

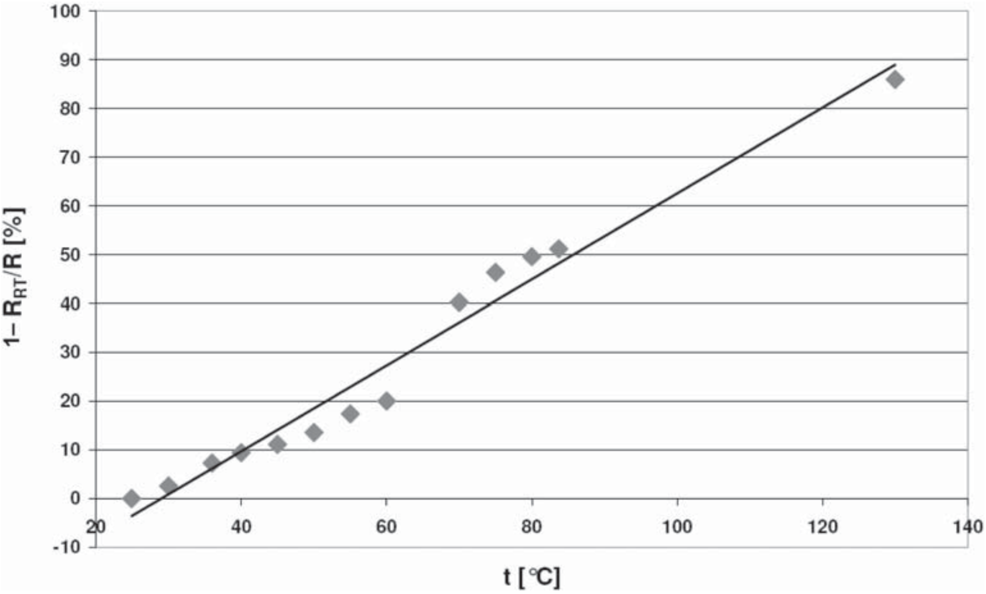

Fig. 4. Temperature dependence of the resistance of an In “spike” contacted FLG (sample g) flake

10 μA but in case of higher measuring currents, some samples show superlinear character, see Fig. 3. Comparing the curves, there is no visible correlation between the sample layer thickness and the degree of nonlinearity. We can find both linear and significantly nonlinear I–V characteristics in case of graphene and FLG samples, too. These nonlinear characteristics resemble to the resultant curve of two reversely connected Schottky diodes. Similar curves were found by J. Zhang et al. [10] in case of gold contacted single wall carbon nanotubes, the authors supposed the formation of Schottky junctions on the two metal/carbon contacts. Another source of nonlinearity can be the thermal effect caused by the higher measuring current. Therefore, the resistance of a contacted graphene sample showing nonlinear I–V characteristics was measured as a function of temperature with 1 mA measuring current. The temperature range was from RT to 135 ºC. Above 140 ºC, an abrupt increase of the resistance was found, presumably because of the melting of the In contacts. The temperature dependence of the sample resistance was found to be linear in the mentioned range with a positive temperature coefficient of $\alpha_{sum} \approx 0.007\ K^{-1}$ (Fig. 4). Accordingly, the thermal effect as the source of nonlinearity can be excluded.

The effect of white light illumination was found to be significant in case of some samples. The change of the resistance of sample “a” in case of a periodic illumination is shown in Fig. 5. As the plot shows, the current immediately starts increasing at the onset of white light with a time constant of about 7.9 s. The falling back of the resistance to its original value when the irradiation is switched off follows a similar slope but the time constant is about 4.7 s. The slow raise of the average resistance during the illumination experiment can be the consequence of temperature effect. This monotonic increase can be characterized by a temperature coefficient of $0.002\ K^{-1}$. The difference between the two temperature coefficients can be the consequence of the 50% duty factor in case of the periodic illumination. No effect of the illumination was found in case of the evaporation and spike-contacted HOPG samples. However, many of the graphite and FLG samples are photosensi-

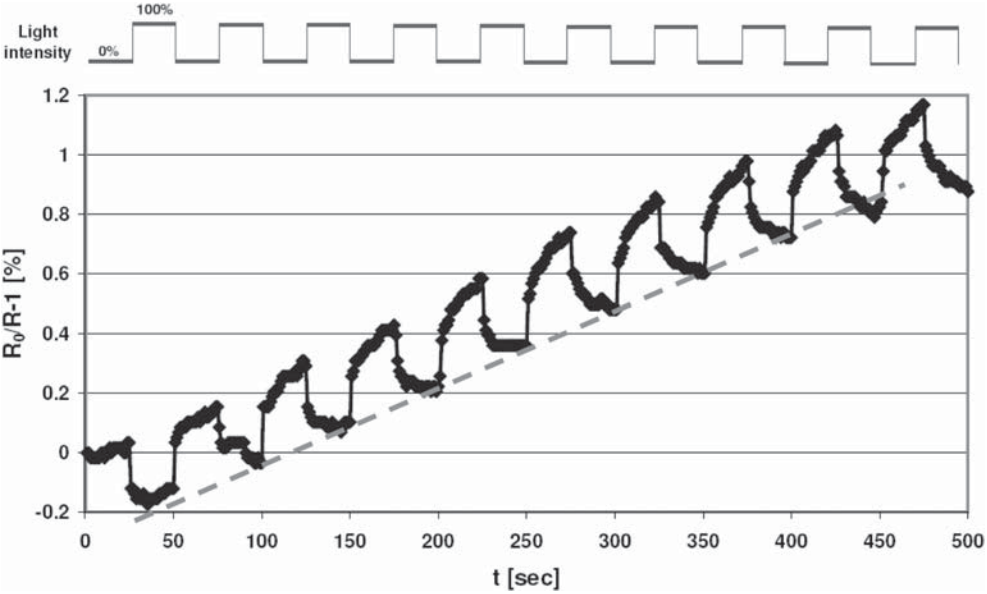

Fig. 5. Relative resistance of an In spike contacted graphene (sample "a") as a function of time in case of periodic white light illumination. The monotonic increase can be characterized by a temperature coefficient of 0.002 K-1.

tive. Similar effect of infrared illumination was found in case of an individual SWCNT [10]. Ideal graphene is regarded as a zero gap semiconductor [1, 11, 12] where the photon induced excess charge carriers can have remarkable role in the conduction. In reality, distortions of the ideal electronic structure due to the finite size, structural defects, especially on the edges and incidental surface contamination can influence the photosensitivity of the individual samples in different ways.

Ar^+ ion irradiation caused significant changes in the I–V characteristics of the samples, see the as prepared and irradiated curve of sample "f" as an example in Fig. 6. In most cases the effect was similar, the nonlinearity of the I–V curves modified towards the linearity but does not reached the completely linear character. In case of the minority of the samples however, this effect cannot be observed, the character of the I–V curves remains the same with small changes in the slope of the curves.

The room temperature I–V measurements, the photoresponse and the effect of ion irradiation may present differences in the behavior of the individual graphene/FLG samples. These differences can be attributed to some extent to the small sizes, which limit the options in placing the contacts and to the particularities of the contact formation when touching the molten In spike to the surface of graphene/FLG layer.

Conclusion

The I–V curves of different In solder spike contacted graphene and FLG flakes were measured and compared to contacted bulk HOPG samples. HOPG samples showed clearly ohmic characteristics while some of the graphene and FLG samples were found to have nonlinear behavior. The degree of nonlinearity does not depend on the number of graphite layers. The effect of nonlinearity can be explained by assuming the presence of Schottky barriers at

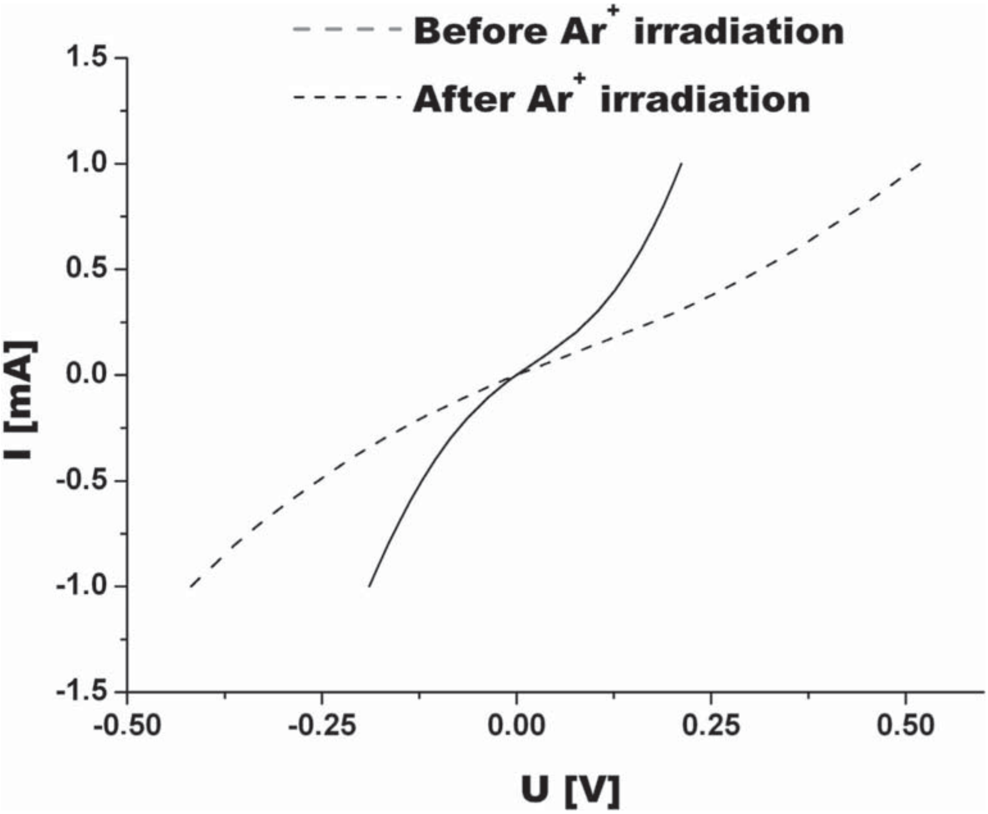

Fig. 6. I–V curves of an In spike contacted graphene flake (sample f) before (dashed line) and after (black line) Ar$^+$ irradiation. The character of the curve remained similar, but the conductivity improved

the contacts. The resistance of the graphene samples showed linear increase as the function of temperature. Some of the graphene and thin FLG samples showed photoconductivity. The differences in the electrical properties of the individual samples can be attributed to the way in which the individual samples were contacted manually.

Acknowledgments

This work was supported by OTKA-NKTH grant No. 67793. One of the authors (Z.E.H.) is a grantee of a Bolyai János Scholarship.

References

[1] A. K. Geim, K. S. Novoselov, Nature Materials 6 (2007) 183–191.
[2] J. H. Chen, M. Ishigami, C. Jang, D. R. Hines, M. S. Fuhrer, E. D. Williams, Adv. Materials 19 (21) (2007) 3623–3627.
[3] J. van den Brink, Nature Nanotechnology 2 (2007) 199–201.
[4] L. Tapasztó, G. Dobrik, Ph. Lambin, L. P. Biró, Nature Nanotechnology 3 (2008) 397–401.
[5] K. S. Novoselov, A. K. Geim, S. V. Morozov, D. Jiang, Y. Zhang, S. V. Dubonos, I. V. Grigorieva, A. A. Firsov. Science 506 (5696) (2004) 666–669.
[6] C. O. Girit, A. Zettl, Appl. Phys. Lett. 91 (2007) 193512.

[7] N. Staley, H. Wang, C. Puls, J. Forster, T. N. Jackson, K. McCarthy, B. Clouser, Y. Liu, Appl. Phys. Lett 90 (2007) 143518.
[8] P. Blake, E. W. Hill, A. H. C. Neto, K. S. Novoselov, D. Jiang, R. Yang, T. J. Booth, K. Geim, Appl. Phys. Lett. 91 (2007) 063124.
[9] P. Nemes-Incze, Z. Osváth, K. Kamarás, L. P. Biró, Carbon 46 (2008) 1435–1442.
[10] J. Zhang, N. Xi, K. Lai, SPIE Newsroom, Doi: 10.1117/2.1200701.0514
[11] M. I. Katsnelson, Materialstoday 10 (1–2) (2007) 20–27.
[12] T. Ando, Physica E 40 (2) (2007) 213–227.