Supporting Information

Sensitive photo-thermal response of graphene oxide for mid-infrared detection

Jung Jun Bae,[a] Jung Hyun Yoon,[b] Sooyeon Jeong,[c] Byoung Hee Moon,[a] Joong Tark Han,[c,d] Hee Jin Jeong,[c] Geon-Woong Lee,[c] Ha Ryong Hwang,[b] Young Hee Lee,[a,e] Seung Yol Jeong,[c,d] Seong Chu Lim*[a,e]*

[a]*IBS Center for Integrated Nanostructure Physics, Institute for Basic Science (IBS), Sungkyunkwan University, Suwon, 440-746, Republic of Korea.*

[b]*WISE CONTROL Inc., R&D Division, 199, Sanggal-dong, Giheung-gu, Youngin-si, Gyeonggi-do, 446-905, Republic of Korea.*

[c]*Nano Hybrid Technology Research Center, Nano Carbon Materials Research Group, Korea Electrotechnology Research Institute, Changwon, 641-120, Republic of Korea.*

[d]*Department of Electrical Functionality Material Engineering, University of Science and Technology (UST), Daejon, 305-333, Republic of Korea.*

[e]*Department of Energy Science, Department of Physics, Sungkyunkwan University, Suwon, 440-746, Republic of Korea.*

*Corresponding authors. E-mail: (S. C. Lim) seonglim@skku.edu and (S. Y. Jeong) syjeong@keri.re.kr

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Bolometer effect

1. Raman spectroscopy of graphene, GO, and reduced-GO

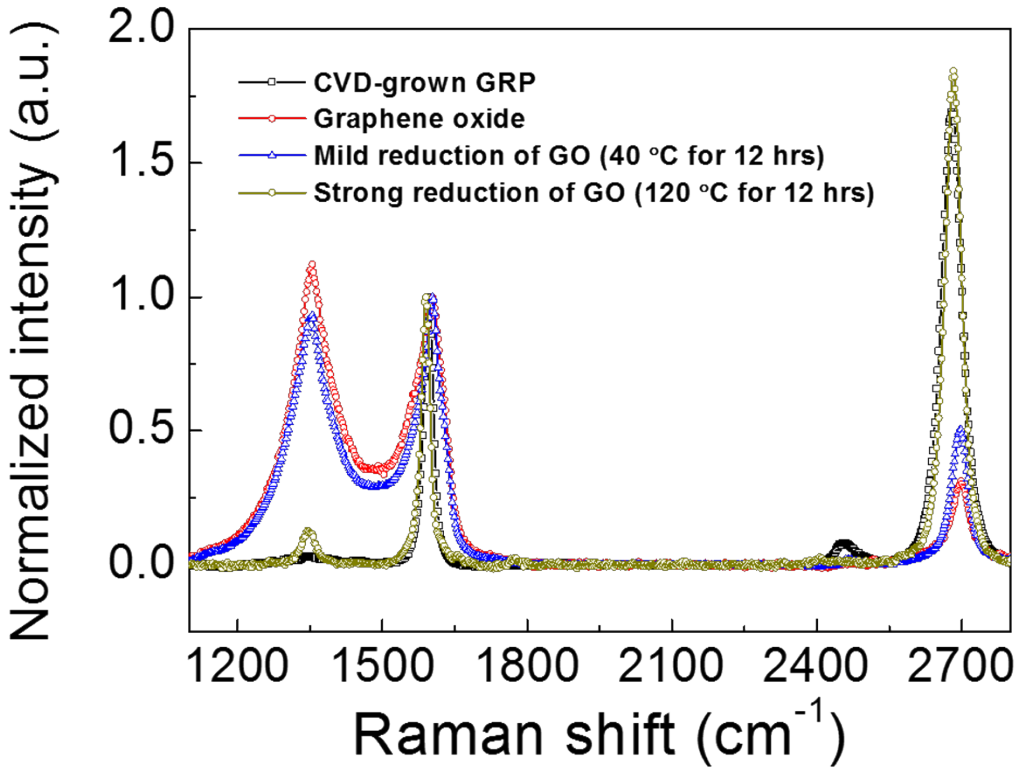


Figure S1. Raman spectroscopy of graphene, GO, and reduced-GO.

Raman analysis provides further insight into doping effect by oxygen moieties and structural changes of graphene due to a chemical treatment, as shown in Fig. S1. The second-order zone boundary phonon (2D) peak of pristine graphene was observed at 2450 cm^{-1} with a symmetric line shape and no shoulder peaks, and high intensity ratio of I(2D)/I(G) confirms a single-layered graphene. In order to describe the effect of oxidation level, four different samples were prepared; pristine CVD-graphene, GO, and annealed GO at 40 °C and 120 °C for overnight in a vacuum condition. Subsequent oxidation treatment of graphene layer, the relative intensity changes of I(D)/I(G) and I(2D)/I(G) and additional G peak shift have several reasons in terms of oxygen moieties with epoxy, hydroxyl, carbonyl, and carboxyl groups. First, the I(D)/I(G) ratio is increased after acid treatment compared to that of the pristine graphene. This is due to chemical functional groups which generates intervalley scattering. This is due to large amount of smaller sp^2 domains of graphene on the basal plane with oxygen functional groups, which indicated a phonon confinement. Subsequent mild reduction by annealing, the ratio decreases due to desorption of oxygen functional groups. This is due to larger domains by reduction on the

basal plane. Second, the intensities of second order Raman peaks (2D) were sensitive to the ordering structure on the basal plane of graphene. These peaks detected changes not only in the polarizability but also in the symmetric and asymmetric vibrational energies. Thus, the broadened FWHM and weak intensities of 2D peaks indicated the disordered carbon rings structure on the basal plane of graphene with oxygen functional groups. In addition, the spectra support the doping effect by oxygen moieties. The charge transfer occurred from graphene to oxygen which corresponds to *p*-type doping. Thus, the I(2D)/I(G) ratio is dramatically decreased and the G band reveals red shift. Generally, the phonon frequency of 2D peak can be decreased by electron-phonon coupling which is described by phonon softening.[1] Therefore, oxidation level of graphene can be described by Raman analysis. The extracted I(D)/I(G) and I(2D)/I(G) ratio from Fig. S1 are summarized in Table S1.

	GRP	Graphene oxide	Mild reduction	Strong reduction
I(D)/I(G)	0.03	1.12	0.93	0.13
I(2D)/I(G)	1.69	0.31	0.51	1.84

Table S1. I(D)/I(G) and I(2D)/I(G) ratio of graphene, GO, and reduced-GO.

2. Resistance vs. temperature curves of GO at V_{ds}

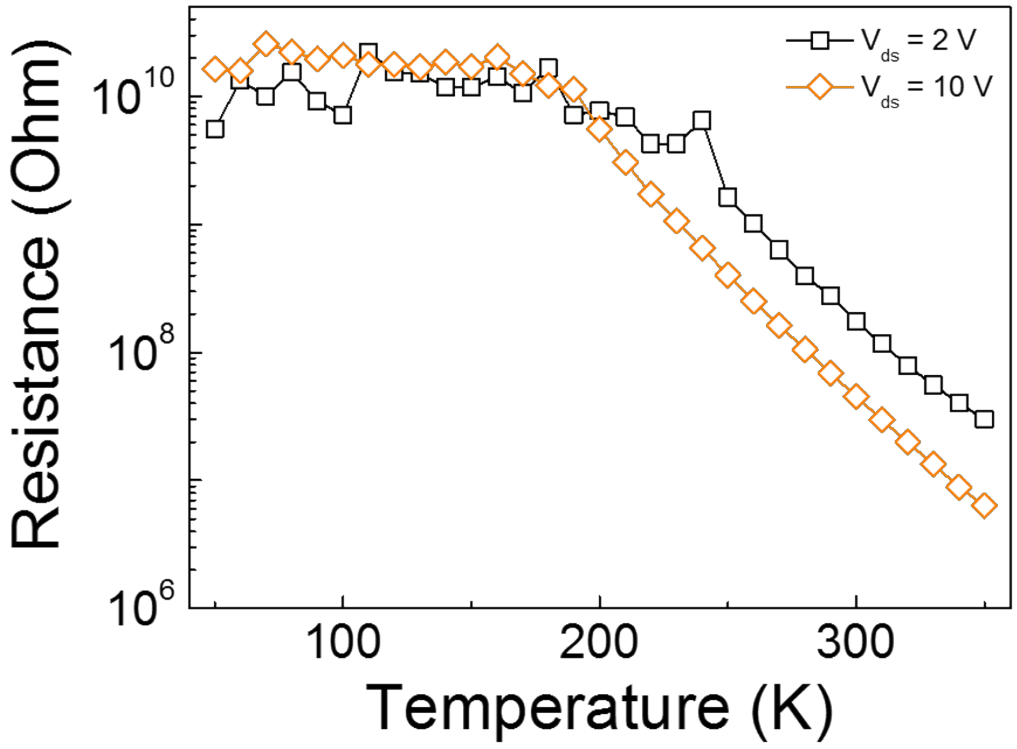
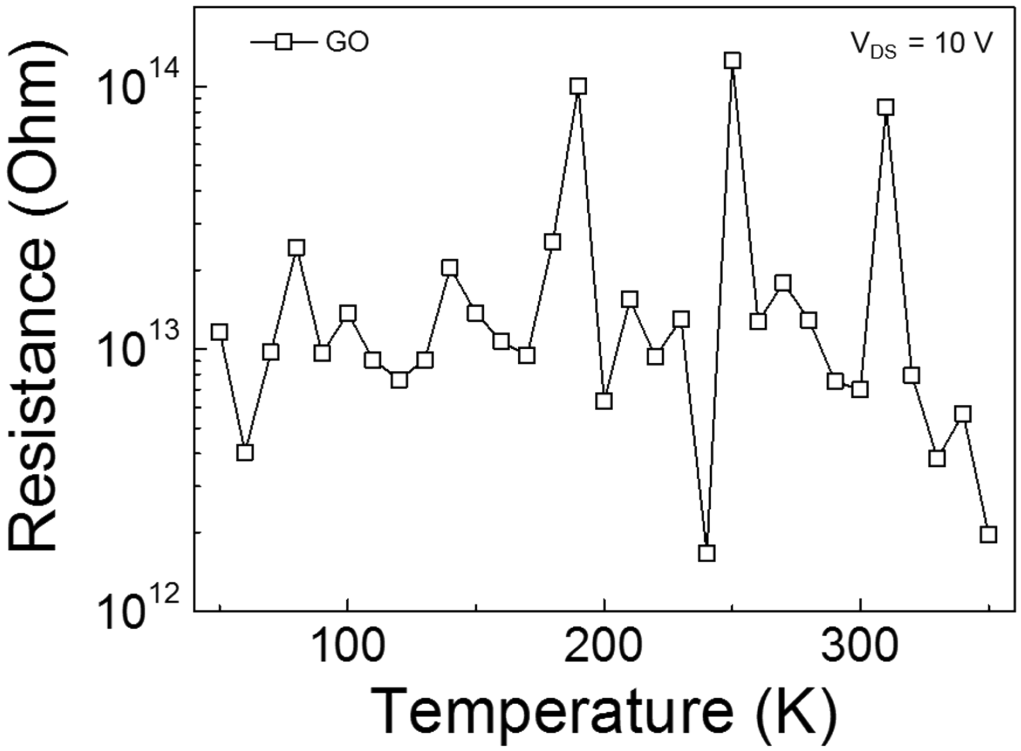


Figure S2. Resistance-temperature plot of m-GO between 50 K to 350 K measured with different V_{ds}.

The resistance of m-GO was measured at different V_{ds} as a function of temperature. Ln(R)-T curve measured at V_{ds} = 2 V, shows a rather higher temperature of resistance-lowering about 250 K, whereas this temperature appears at 200 K for V_{ds} = 10 V. After an initiation of resistance lowering, the logarithm of resistance shows a nearly linear decrease to the temperature with a slightly different slope for 2 volt and 10 volt. The reduction of resistance-lowering temperature and lower slope of plot measured with V_{ds} = 10 V is due to a weak field-driven transport of m-GO.

3. Resistance of GO as a function of temperature



The resistance of GO was measured from 50 K to 370 K using Keithley 6517A electrometer. The resistance of GO fluctuates with a large degree and does not change noticeable at all through the temperature range.

4. Resistance vs. temperature curves of GO at different V_{ds} under IR illumination

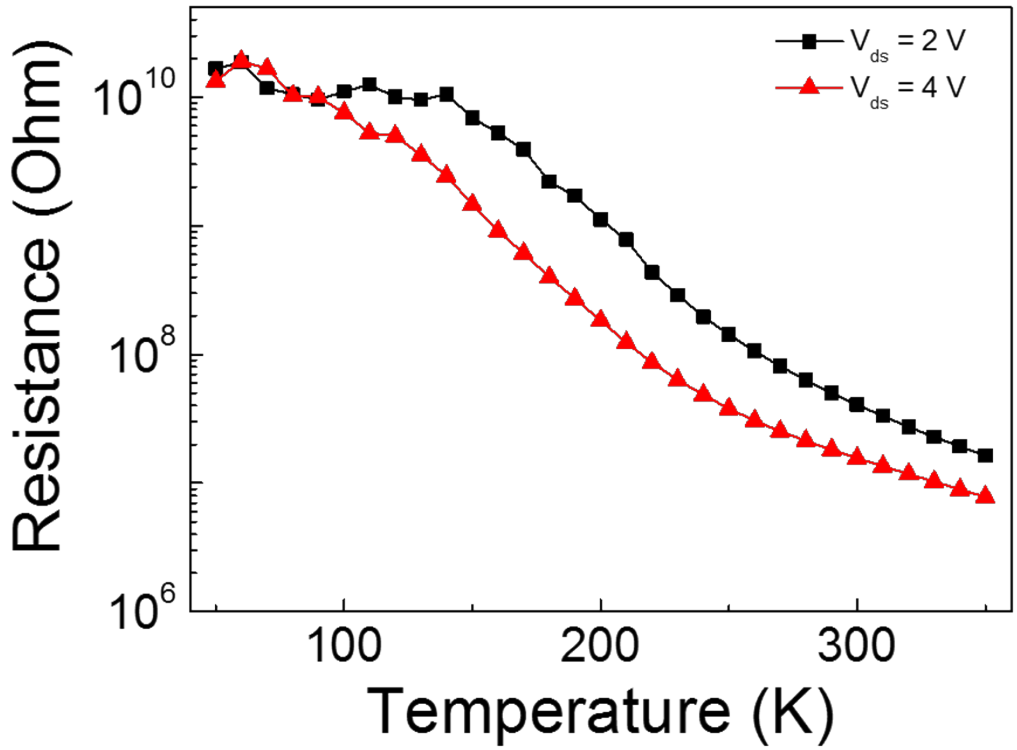


Figure S4. Resistance-temperature plot of GO under IR illumination between 50 K to 350 K measured with different V_{ds}.

The resistance of GO was measured at different V_{ds} as a function of temperature while IR source was shining a GO device. Ln(R)-T curves measured under IR illumination show a more gradual change of transport behavior comparing to Fig. S2. In the temperature range between 250 and 350 K, the transport behavior is observed to be nearest neighboring hopping (NNH). The temperature range for NNH is expected to be wider for V_{ds} = 2 volt than V_{ds} = 4 volt. In particular, it is well manifested in Fig. 3d that the NNH range becomes lower and narrower in temperature when V_{ds} = 10 volt. Below 200 K, non-exponential decrease of resistance measured below 200 K may be caused by field-driven transport[2], which is more prominent under IR illumination comparing to Fig. S2.

[1] Y. Zhu, S. Murali, W. Cai, X. Li, J. W. Suk, J. R. Potts, R. S. Ruoff, *Adv. Mater*., 2010, **22**, 3906.

[2] C. Y. Cheah, A. B. Kaiser. *Int. J. Nanotechnol*., 2014, **11**, 412.