Supporting Information (SI):

Electrical transport properties and ultrafast optical

nonlinearity of rGO-metal chalcogenide ensembles

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1. XPS full surface survey spectrum:



Figure S(I): Full surface survey XPS spectrum of sample (a) C1 and (b) C2

Figure S(I)(a) shows the XPS spectrum of rGO-ZnSe (C1) in a wide binding energy range of 0-1100 eV and this shows the presence of different elements like Zn, Se, C and O. The full surface survey spectrum of rGO-ZnTe (C2) composite (figure S(I)(b)) confirms the presence of Zn, Te, C and O.

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2. SEM images:



Figure S(II): SEM images of sample (a) C1 and (b) C2

3. UV-vis and PL spectra:



Figure S(III): (a), (c) UV-vis absorption and (b), (d) PL spectra of ZnSe, C1, ZnTe and sample C2. Inset of figure (a) show the tauc plot of ZnSe and C1 with a band gap of ~ 2.65 eV and ~ 1.8 eV, respectively. Inset of (c) indicates ~ 2.4 eV and ~ 1.6 eV band gap of ZnTe and C2, respectively. The excitation wavelength for PL measurement was 300 nm for ZnSe, C1 and 460 nm for ZnTe, and C2.

To get information about the interaction of rGO with ZnSe and ZnTe, UV-vis and PL spectroscopy measurements have been performed as shown in figure S(III). Pure ZnSe and ZnTe show a characteristic absorption peak at around 290 nm whereas after incorporation

of rGO, the absorption coefficient of the composites have been improved over the whole visible range (figure S(III)(a) and S(III)(c)). Steady state PL spectra of ZnSe (figure S(III)(b), excited at 300 nm) depicts an emission maxima at \sim 510 nm which is arising from deep level emission of Zn vacancies or impurities present in the sample [1]. The PL spectrum of ZnTe (figure S(III)(d), excited at 460 nm) exhibits a strong emission at 548 nm, assigned to the band to band electronic transition of ZnTe [2]. Incorporation of rGO causes efficient charge transfer from conduction band of ZnSe/ZnTe to rGO sheets which results in quenching of emission band in the composites [2].

4. J - V characteristics of rGO-ZnSe and rGO-ZnTe composites at 380K temperature:



Figure S(IV): J - V characteristics of different (a) rGO-ZnSe (A1, E1), (b) rGO-ZnTe (A2, E2) composites both under dark and illumination at 380K temperature.

Sample	Values of m		Sample	Values of m		
Sample	Sample Dark Illumination	Dark		Illumination		
A1	1.97	2.14	A2	2.08	1.94	
B1	2.05	2.14	B2	2.09	2.14	
C1	2.13	2.16	C2	2.14	2.18	
D1	2.11	2.22	D2	2.13	2.20	
E1	1.93	2.01	E2	2.17	2.09	

5. Table ST(I): Values of *m* for different compositions at 380K in dark and illumination conditions:

6. J - V characteristics at different temperatures:



Figure S(V): J - V characteristics at different temperatures for rGO in (a) dark, (b) illumination and for rGO-ZnSe (C1) in (c) dark and (d) illumination conditions.

7. Capacitance- frequency (C - f) response:

Frequency dependent (40Hz-70MHz) capacitance (C) has been measured for rGO, sample C1 and C2. It is observed from figure S(VI) that capacitance decreases with increasing frequency as higher frequency causes drift of space charges accumulated at the



Figure S(VI): Capacitance-frequency characteristics of (a) rGO, (b) C1 and (c) C2 at room temperature.

Al/semiconductor junction. At higher frequency sample capacitance becomes independent of frequency and attains the value of geometrical capacitance (C_g) of the sample. Using the value of C_g dielectric constant of the sample can be calculated with the help of following formula

$$C_g = \frac{\epsilon_0 \epsilon_r A}{d} \tag{1}$$

Where ϵ_0 is the free space permittivity, ϵ_r is the dielectric constant of the active material, A represents the active sample area and d is the thickness. The values of ϵ_r for rGO, C1, C2 have been tabulated in table ST(II).

8. Table ST(II): Sample thickness (d), ϵ_r of rGO, C1, and C2

sample	ϵ_r	d (nm)		
rGO	3.2	900		
C1	4.5	1050		
C2	5.1	1000		

9. Extrapolation of J - V curves:



Figure S(VII): Extrapolation of J - V curves at different temperatures for rGO and C1 in (a), (c) dark and (b), (d) illumination conditions. It gives V_C ~19V and ~12V for rGO and ~45V and ~30V for sample C1 in dark and illumination, respectively.

10. Temperature dependent current-voltage response in dark and illumination:



Figure S(VIII): Current-voltage (I-V) response of (a) rGO, (b) C1 and (c) C2 in dark and illumination condition at different temperatures.

Temperature	Value of m in dark			Value of m under illumination		
(K)	rGO	C1	C2	rGO	C1	C2
87	3.28	-	-	3.39	-	-
100	3.15	3.12	3.49	3.32	3.23	3.60
110	3.09	3.06	3.43	3.25	3.13	3.58
120	3.04	2.96	3.36	3.21	3.07	3.56
140	2.81	2.91	3.38	3.02	3.05	3.43
160	2.66	2.74	3.23	2.89	2.84	3.29
180	2.57	2.64	3.07	2.73	2.79	3.23
200	2.45	2.48	2.96	2.54	2.74	3.09
220	2.29	2.43	2.89	2.40	2.61	3.04
240	2.16	2.31	2.74	2.32	2.50	2.95
260	2.07	2.23	2.67	2.23	2.43	2.82
280	2.04	2.16	2.54	2.11	2.25	2.72
300	1.93	1.99	2.49	2.02	2.19	2.57
320	1.90	1.95	2.37	1.96	2.10	2.46
340	1.76	1.91	2.28	1.82	2.02	2.38
360	1.78	1.83	2.24	1.80	1.97	2.34
380	1.63	1.76	2.15	1.73	1.86	2.21
400	1.50	1.64	2.08	1.64	1.83	2.20
420	1.47	1.50	1.89	1.59	1.73	2.11
440	1.44	1.45	1.78	1.48	1.62	1.85
460	1.42	1.37	1.65	1.42	1.55	1.75
473	1.37	1.29	1.62	_	_	-

11. Table ST(III): Variation of exponent (m) with temperature of rGO, C1 and C2 samples in region II both under dark and illumination conditions.

12. Frequency dependent conductivity of sample C1 and C2 in dark:



Figure S(IX): Frequency dependent conductivity of C1 and C2 at room temperature. The solid lines are the best fits to equation 2.

The conductivity isotherms can be described with the following empirical formula

$$\sigma_{ac} = \sigma_{dc} + B\omega^s \tag{2}$$

where σ_{ac} and σ_{dc} are ac and dc conductivity respectively, *B* is a prefactor and *s* is an exponent. The frequency dependent conductivity data have been fitted with equation 2 and it gives $s \sim 0.96$ and ~ 1.3 for sample C1 and C2 at room temperature, respectively.

References

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