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Non-Hysteretic Magnetoresistance In rGO-PVA Nanocomposite Films Consisting of Chromium Oxides Nanoscale Composite Particles

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Abstract

Reduced graphene oxide (rGO) incorporated *poly*-vinyl alcohol (PVA) polymer nanocomposite films reinforced with multigranular & multiphasic chromium oxides composite nanoparticles have been developed. The composite nanoparticles are loaded in 1 wt% (Film-1) and 2 wt% (Film-2) formulations in the rGO-PVA nanocomposite films. Structural, morphological, electrical, optical & magnetotransport studies have been performed on these nanocomposite films. Electrical studies reveal a resistance of ~600 ohms at 300 K along with existence of Effros-Shklovskii type of variable range hopping in Film-1. Film-2 shows an electrical resistance of ~1200 ohms at 300 K but an enhanced range of hopping mechanism. The transition temperatures for Film-1 and Film-2 are 198 K and 240 K, respectively. After these temperatures, spin-independent hopping (SIH) channel also opens-up in addition to the spin-dependent tunnelling (SDT) channel of transport. As the temperature increases beyond 300 K, SDT is lost slowly, but completely, and pure SIH becomes the dominant mechanism. Non-hysteretic MR curves were observed in both nanocomposite films with -30% MR in Film-1 and mixed MR of 27.5% in Film-2 at room temperature in the out-of-plane geometry of the applied field. Occurrence of negative MR is ascribed to weak localization (WL), while positive MR is attributed to weak antilocalization (WAL). The non-hysteretic MR is particularly useful for the development of magnetic field sensors, sensors for the determination of speed of wheel in automobiles, biomedical magnetometers, and also in case of non-volatile memory devices.

Keywords : 2D materials, rGO, magnetoresistance in graphene, nanocomposites, hopping.

Introduction

Graphene is a highly interesting 2D material slated for the development of high speed nanoelectronics and spintronic devices along with the quality of reduced power operation [1-5]. Graphene can serve the purpose of channel material in a spintronic transistor possessing outstanding features of long spin life time and long spin relaxation lengths [5-10]. Recently, graphene has been experimentally detected to possess higher spin relaxation lengths of the orders of micrometres preferably at room temperature [11-15]. This opens-up the possibilities of the development of spintronic devices functioning at room temperature. Graphene is also interesting for gate-tunable spin-orbit interactions [16, 17], non-traditional superconductivity [18], introduction of magnetism [19, 20], spin Hall effect [21, 22], spin-filtering effect [23, 24], spin injection efficiency [25, 26], proximity-induced ferromagnetism [27, 28] and spin-based long communications [29-32]. Wees *et al.* used a device structure consisting of insulating SiO₂ layer over the *n*-doped Si substrate [33]. A longer spin relaxation length in the range of 1.5-2.0 μm was measured in the graphene at room temperature. This spin-polarized conduction was found to be not dependent on the temperature in the single layered graphene. Thus, the spin-polarized transport and precession were demonstrated for the first time in this paper by adopting the non-local contacting in conjunction with the artificial tunnel barriers. Kawakami *et al.* have demonstrated spin injection in tunnelling format into the single layer graphene by application of MgO barriers having a seed layer of TiO₂ [34]. The study reported a non-local magnetoresistance of 130 Ω in the specific device structure at room temperature. A crux of this study was the implementation of tunnelling contacts in contrast to the transparent contacts in line with the drift-diffusion theoretical account of spin-polarized conduction [35]. Kawakami *et al.* have performed spin-transport measurements in both single- and bilayer graphene and reported contrasting nature in both types of graphene [36]. The spin lifetime (τ_s) was discovered to be changing in a linear manner as a function of the momentum scattering time (τ_p) with the

variation of carrier concentration in case of single layer graphene at low temperature. This particular nature demonstrated the prevalence of Elliot-Yafet (EY) spin relaxation mechanism at low temperatures. Özyilmaz *et al.* conducted reportedly to be the first study on spin-polarized transport in form of charge density, minimum conductivity, mobility and temperature [37]. D'yakonov-Perel' spin relaxation mechanism prevailed in bilayer graphene because the spin relaxation time (τ_s) showed inverse nature to the mobilities both at low and room temperatures. The study reported the spin relaxation times to a maximum of 2 ns at room temperature in the bilayer graphene with minimum mobilities.

Pure graphene is largely diamagnetic in nature and shows orbital diamagnetism in the vicinity of the Dirac point [39]. It is at this point that a strong negative magnetic susceptibility behaviour becomes more pronounced. Graphene has inherent spin polarization of a very weak nature as well as weak spin-orbit interactions [1]. Despite diamagnetic behaviour, graphene has many applications in spintronics, just as highlighted above. There are occurrences of defects & disorders in graphene structure as well. The defects- and disorders ridden structure of carbon nanomaterial has the honeycomb lattice identical to the graphene. The reduced graphene oxide or rGO possesses sp^2 hybridization in its structure, but, unlike graphene, it consists of sp^3 hybridization as well. This is the basic difference in pure graphene and rGO in terms of structure. The rGO is obtained by the reduction of graphene oxide which is heavily oxidized and is insulating in nature. The purity of the derived structure depends on the degree of reduction of plenty of -OH, -COOH and epoxy groups. Complete removal of the oxygen functional groups from graphene oxide by reduction is never possible, in principle. The residual functional groups, such as, -OH, -COOH and epoxy groups, still keep on attaching oxygen atoms to the carbon plane forcing to get arranged in sp^3 -hybridization. Also, there are involved severe oxidation as well as reduction reactions in the making of graphene. Thus, the carbon lattice has the high susceptibility of getting vacancies, structural defects as well as distorted

Stone-Wales bonds [40]. The distorted Stone-Wales bond is a type of crystallographic defect that involves rotation of the two carbon atoms by 90° with special reference to the central point of the bond connecting them [41]. Thus, it involves the alteration in linkages of the two C-atoms that are connected together by means of a π -bond. Thus, rGO has sound electrical nature, good thermal conductivity as well as mechanical strength, but, consists of defects and disorders so that the lattice is not fully sp^2 -bonded. Moments from defects and those from residual oxygen groups contributing through unpaired electrons are the main source of inducing magnetic properties in rGO. Besides, graphene can be intentionally doped with magnetic elements by physical methods, doping by chemical routes, and there may be other methods as well, including thermal reduction. Graphene can be produced by a number of techniques that including and sputtering, chemical vapour deposition (CVD) and even chemical synthesis techniques [42-55]. The rGO in sp^2 -carbon network can be retained by several sustainable & non-toxic reducing agents. These reducing agents can be L-ascorbic acid or vitamin C [56], green methods of reduction from natural plants [57] as well as phyto-extracts [58, 59] hydrothermal route to optimize good crystallinity and reduced defect densities [60], sonication assisted exfoliation [61].

This is pertinent here to discuss few reports of rGO mentioning electrical & magnetoresistive properties. Yang *et al.* in-situ deposited FeCo nanoparticles over rGO flakes for the development of a hybrid magnetic sensor [62]. It was discovered that the quantum magnetoresistance (MR) of the hybrid material can be explained by Abrikosov's model applied for layered materials. Interestingly, MR value was tunable by modifying the density of nanoparticles over the rGO flakes. The study reported an MR value of $21.02 \pm 5.74\%$ under the application of $H = 10$ kOe at room temperature. Further, the study reported the integration of hybrid magnetic sensor in a wireless system for actual estimation of electromagnetic radiation produced by a working mobile phone. Chen *et al.* decorated rGO nanosheets with

Fe₅₀Co₅₀ nanoparticles that showed MR of -7% under $H = 9.5$ kOe at room temperature [63]. It was deciphered that increasing the negative MR-value enhances the performance of the sensor so that much lower concentration of glucose could be detected. Xie *et al.* prepared nanocomposite aerogel of rGO/Fe₃O₄/PANI that showed an MR of -4.37% under $H = 8.0$ T [64]. The authors claimed that this is the first report that mentions negative MR in the rGO-based nanocomposite aerogel. Roy *et al.* detected negative MR of -1.4% in CoFe₂O₄ (CFO):rGO nanocomposites in ratio of 1:1. On the other hand, negative MR of $\sim -1\%$ for 2:1 and 3:1 compositional ratios. During the electrical transport, charge transport happens because of conducting phase of rGO. However, CFO is magnetic in nature but is electrically insulating which induces electron-spin scattering. This electron-spin scattering is reduced under the application of H because parallel alignment of magnetic moments happens under H . Kartik *et al.* have deposited rGO thin films by the technique of pulsed laser deposition (PLD). Large positive and negative MR were observed at high and low applied H at room temperature. Electrical transport mechanism in these thin films consisted of two regimes. The first mechanism is the 2D Efros-Shklovskii variable range hopping happening typically at low temperatures. The second mechanism is the Arrhenius-type electrical transport happening at temperatures above 200 K. The reason for this crossover in transport regimes was ascribed to reductions in the characteristic lengths in the 2D system in a typical disordered configuration. The origin of MR in the rGO films was attributed to the vacancies, magnetic moments due to defects and disorders and to the diffuse scattering occurring at the crystallite boundaries. The films showed a high value of MR of 160% along with positive MR ascribed to the Lorentz force at high applied fields.

In the current work, our approach is to develop rGO-polymer films incorporated with multiphasic chromium oxide nanophase composite particles by comparatively easier simple solution chemistry techniques basically for magnetic field sensors.

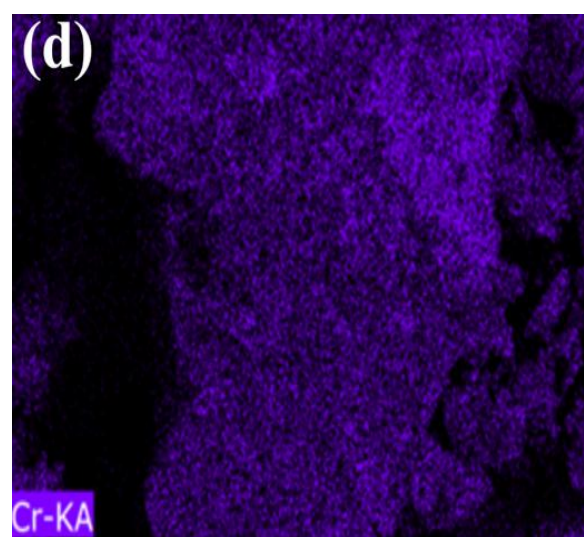
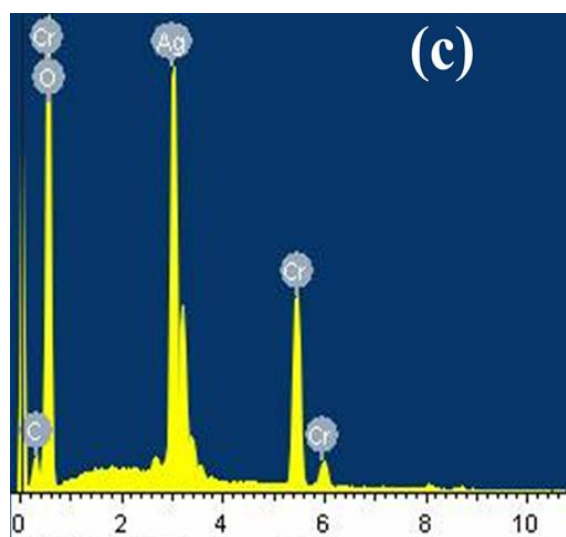
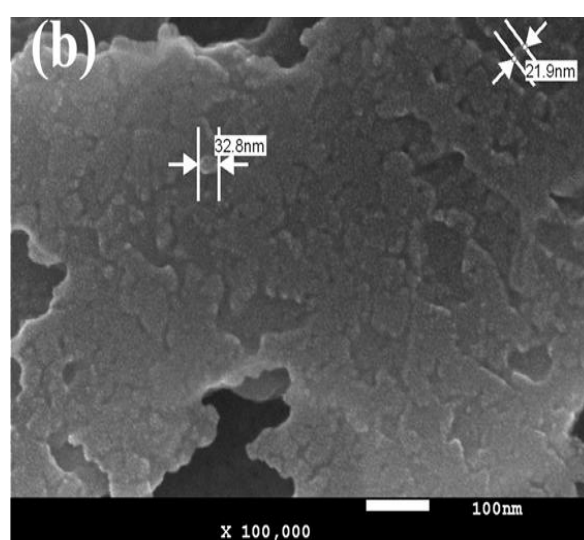
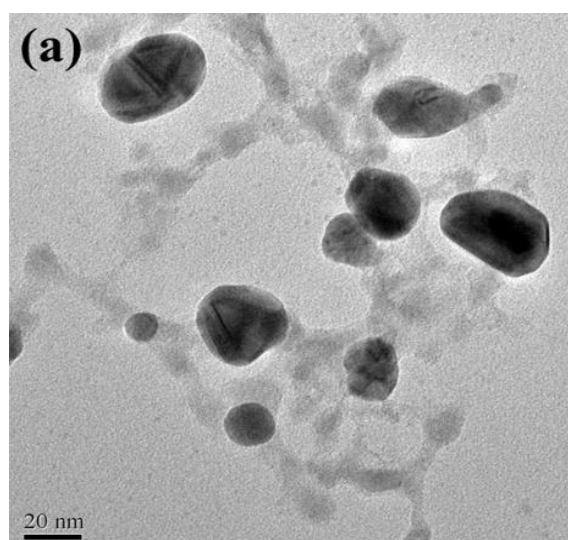
Experimental

Multigranular chromium oxides nanophase composite particles were synthesized by a technique as mentioned by us in previous research reports [67, 68]. These composite nanophase particles with loadings of 1 wt% and 2 wt% were dispersed in the 2 wt% aqueous solution of PVA consisting of 3 wt% rGO nanoflakes for the development of nanocomposite films. All the above mixing and stirring processes were conducted at room temperature only. After proper mixing of the above materials in aq. phase for sufficient time, the mixture solution was dried at $\sim 40^{\circ}\text{C}$ to get nanocomposite films.

X-ray diffractogram from PANalytical X'Pert Pro possessing $\text{CuK}\alpha$ radiation of wavelength 0.15406 nm was used for the determination of crystal structures of different phases contained in the nanocomposite films. For the observation of shape and size of the composite nanoparticles, high resolution transmission electron microscopy (HRTEM) from JEOL JEM-2100 was used which was operated at 200 kV of accelerating voltage. Field emission scanning electron microscope (FESEM) from Nova Nano FE-SEM 450 (FEI) model was used for imaging of surface morphological features contained in the nanocomposite films along with energy-dispersive spectrum (EDS) and colour mapping. STR series Raman spectrometer with a laser wavelength of 532 nm offering a resolution smaller than 0.5 cm^{-1} was employed to collect a representative Raman spectrum of the nanocomposite film possessing 2 wt% of composite nanoparticles. Electrical measurements were done with the help of Physical Quantities Measurement System (PQMS) of XPLORE 1.2 Model from Quazar Technologies, India. Magnetoresistance (MR) curves were acquired using an electromagnet from Polytronic Inc., Mumbai, India along with a standard Keithley 2400 SourceMeter for measuring voltages at constant current. A digital gaussmeter from Scientific Equipment, Roorkee, India of Model DGM-102 with a standard probe were used for magnetic field measurements.

III. Results And Discussions

Figure 1(a), (b) and (c) show the HRTEM image, FESEM image and EDS of the multigranular chromium oxides nanophase composite particles. HRTEM images in Figure 1(a) showed average particle size of ~ 35 nm of nearly spherical shape. Figure 1(b) shows the FESEM image of nanophase composite particles studded in the PVA polymer typically forming an agglomeration matrix. Figure 1(c) shows the energy dispersive spectrum (EDS) of the nanophase composite particles. EDS quantified the Cr at K-edge as 13.44 at.%, O at K-edge as



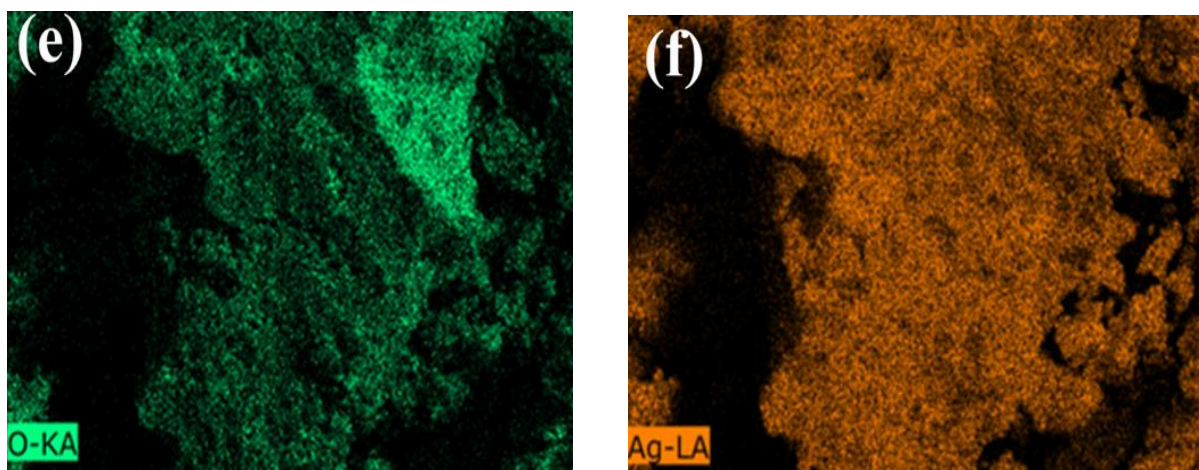


Figure 1. (a) shows HRTEM images of the multigranular chromium oxides nanophase composite particles. An average size of ~ 35 nm of the nanophase composite particles was estimated from the HRTEM image. (b) shows the surface morphological features of the nanophase composite particles. Effect of polymer is apparent from the image since agglomeration leads the whole imaging process. (c) shows the EDS of the composite particles which depicts the presence of elements of Cr at K-edge as 13.44 at.%, O at K-edge as 72.48 at.%, Ag at L-edge as 10.75 at.% and C at K-edge as 3.33 at.% forming a total of 100% atomic composition. Colour mapping of the composite nanoparticles for Cr at K-edge, O at K-edge and Ag at L-edge is shown in (d), (e) and (f), respectively. The readers may refer to [67, 68] for a detailed description of the multigranular chromium oxides nanophase composite particles.

72.48 at.%, Ag at L-edge as 10.75 at.% and C at K-edge as 3.33 at.%. Further, Cr at K-edge was estimated to be 22.86 wt%, O at K-edge as 37.92 wt%, Ag at L-edge as 37.91 wt% and C at K edge as 1.31 wt% in a typical composition. Images of colour mapping as acquired from EDS for Cr K-edge, O K-edge and Ag L-edge are shown in Figure 1(d), 1(e) and 1(f), respectively. An even distribution of these elements can be observed for these elements in a typical sample as given from Figure 1(d), 1(e) and 1(f), respectively.

X-ray diffractograms of the nanocomposite films consisting of rGO and reinforced with 1 wt% nanophase composite particles, and, nanocomposite films with 2 wt% composite particles are shown in Figure 2(a) & 2(b), respectively. The peaks identified for the two nanocomposite films have been marked in Figure 1(a) & 1(b) with their respective crystal orientations. In these

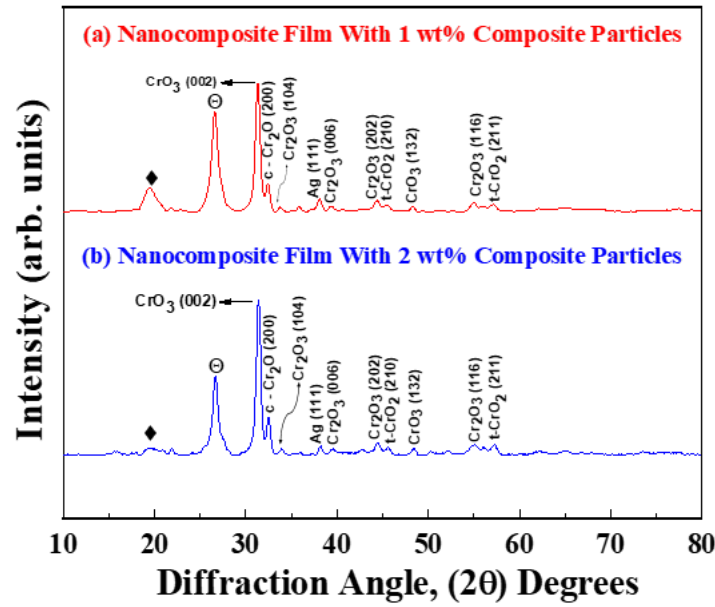


Figure 2. (a) and (b) show the X-ray diffractograms of the nanocomposite films formed by inclusion of 1 wt% and 2 wt% multigranular chromium oxides nanophase composite particles, respectively. In (a) and (b), $\blacklozenge$ symbol is used for hump associated with the amorphous PVA polymer and Θ symbol represents that related to the reduced graphene oxide or rGO. With increase in particle loading from 1 wt% to 2 wt% in the nanocomposite films, peak reflection intensity of CrO_3 (002) rises. On the other hand, reflection intensity from rGO comes down slightly because of higher particle loading.

rGO-enhanced PVA nanocomposite films, hump associated with the amorphous *poly*-vinyl alcohol (PVA) phase appears at a diffraction angle of $2\theta = \sim 19.5^\circ$. Ricciardi *et al.* reported the major peak at $2\theta = 19.4^\circ$ in PVA with a side peak at $2\theta = 20^\circ$ [69]. Guirguis *et al.* reported the semicrystalline peaks at $2\theta = 19.45^\circ$, 20.65° and 23.52° in case of pure PVA polymer [70]. Bhargav *et al.* also discovered a broader peak at $2\theta = 20^\circ$ in PVA which showed a decline in intensity on inclusion of NaI into the PVA [71]. Hema *et al.* confirmed a broader peak at $2\theta = \sim 19.7^\circ$ identified with amorphous behaviour of the PVA films [72]. The second peak appears at $2\theta = 26.6^\circ$ in good reflection intensities in both the nanocomposite films. This peak of sound reflection intensity arising from reduced graphene oxide or rGO shows the higher degree of crystallinity of the carbon nanomaterial embedded in the amorphous polymer phase. This peak

is of slightly higher intensity in 1 wt% nanocomposite (Film-1) film than the 2 wt% nanocomposite film (Film-2) owed to lesser wt% of the nanophase composite particles inclusion. Other peaks in Film-1 are CrO_3 (002), $\text{c-Cr}_2\text{O}$ (200), Cr_2O_3 (104), Ag (111), Cr_2O_3 (006), Cr_2O_3 (202), $t\text{-CrO}_2$ (210), CrO_3 (132), Cr_2O_3 (116) and $t\text{-CrO}_2$ (211). Similarly, other peaks in Film-2 are CrO_3 (002), $\text{c-Cr}_2\text{O}$ (200), Cr_2O_3 (104), Ag (111), Cr_2O_3 (006), Cr_2O_3 (202), $t\text{-CrO}_2$ (210), CrO_3 (132), Cr_2O_3 (116) and $t\text{-CrO}_2$ (211). Various parameters of the two nanocomposite films related to these phases have been summarized in Table I. For Rietveld analysis, ICDD (International Centre For Diffraction Data) card of number 01-075-7969 was employed for tetragonal CrO_2 , 04-005-4336 for hexagonal Cr_2O_3 , 01-073-6548 for orthorhombic CrO_3 , 04-013-6546 for cubic Cr_2O and 00-003-0921 for metallic Ag phase. This

Table I. Different parameters pertaining to X-ray diffractograms of Film-1 are presented in a tabular form.

S. No.	2θ	Phase	hkl	d -spacing (nm) (Observed)	Reflection Intensity (%) [Film-1]
1.	31.33°	CrO_3	(002)	0.2853	100.00
2.	32.43°	$\text{c-Cr}_2\text{O}$	(200)	0.2758	22.00
3.	33.82°	Cr_2O_3	(104)	0.2648	3.80
4.	38.10°	Ag	(111)	0.2359	10.06
5.	39.37°	Cr_2O_3	(006)	0.2286	4.59
6.	44.40°	Cr_2O_3	(202)	0.2039	8.87
7.	45.48°	$t\text{-CrO}_2$	(210)	0.1992	5.31
8.	48.34°	CrO_3	(132)	0.1881	4.44
9.	55.05°	Cr_2O_3	(116)	0.1682	7.69
10.	57.15°	$t\text{-CrO}_2$	(211)	0.1610	5.86

Table II. Different parameters pertaining to X-ray diffractograms of Film-2 are presented in a tabular form.

S. No.	2θ	Phase	hkl	d -spacing (nm) (Observed)	Reflection Intensity (%) [Film-2]
1.	31.39°	CrO_3	(002)	0.2848	100.00
2.	32.49°	$\text{c-Cr}_2\text{O}$	(200)	0.2753	24.31
3.	33.90°	Cr_2O_3	(104)	0.2642	3.75
4.	38.22°	Ag	(111)	0.2353	5.58
5.	39.55°	Cr_2O_3	(006)	0.2277	4.16
6.	44.48°	Cr_2O_3	(202)	0.2035	8.24
7.	45.57°	$t\text{-CrO}_2$	(210)	0.1989	4.66
8.	48.46°	CrO_3	(132)	0.1877	3.91
9.	55.10°	Cr_2O_3	(116)	0.1665	6.83
10.	57.29°	$t\text{-CrO}_2$	(211)	0.1607	6.58

is highly desirable to mention about the strain in tetragonal or t -CrO₂ lattice in the two nanocomposite films since it is the major phase imparting spin-polarized electrons. The W-H or Williamson-Hall plots for Film-1 and Film-2 in case of t -CrO₂ lattice are shown in Figure 3(a) & 3(b), respectively. The plots can be analyzed using the following given formula in a linear form, whereby, slope represents the strain (ϵ) and intercept represents the crystallite size (D) [73, 74],

$$\beta \cos \theta = \frac{k\lambda}{D} + 4\epsilon \sin \theta \dots \dots \dots (3)$$

where, β is the full width at half maximum (FWHM) in radians, ϵ is the strain, D is the effective crystallite size and θ is the diffraction angle. Strain of t -CrO₂ lattice comes out to be -0.0033 and $D = 10.23$ nm in case of Film-1. Strain of t -CrO₂ lattice comes out to be 0.0021 and $D = 12.66$ nm in case of Film-2. All the data pertaining to strain (ϵ) and crystallite size (D) in case of two films is summarized in Table II.

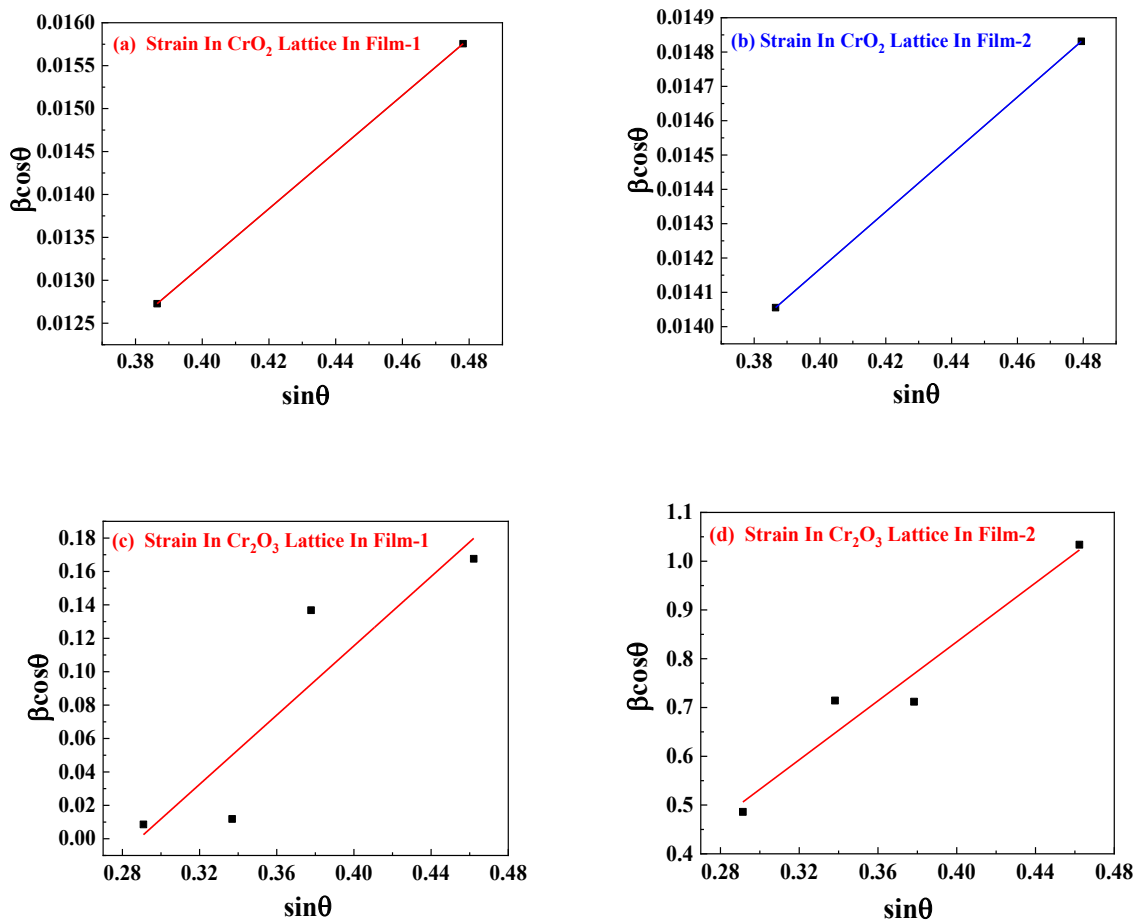


Figure 3. (a) and (b) show the Williamson-Hall (W-H) plots of $t\text{-CrO}_2$ lattice contained in Film-1 and Film-2, respectively. Compressive strain was detected in $t\text{-CrO}_2$ lattice in Film-1, while tensile strain was determined in Film-2, respectively. The crystallite size (D) was estimated to be 10.23 nm in Film-1, while D was determined to be 12.66 nm in Film-2.

Table II. Strain (ϵ) and crystallite size (D) in $t\text{-CrO}_2$ lattice in Film-1 & Film-2 samples.

Physical Quantity	CrO ₂ Lattice In Film-1 Sample	CrO ₂ Lattice In Film-2 Sample
Strain (ϵ)	-0.0033 (Compressive Strain)	0.0021 (Tensile Strain)
Crystallite Size (D)	10.23 nm	12.66 nm

Field emission scanning electron microscopy (FESEM) based images of Film-1 and Film-2 samples are shown in Figure 4(a) & Figure 4(b). Nearly similar surface morphological features are visible from the electron micrographs of the two films acquired at the magnification of 50,000X. However, more clustering of composite nanoparticles is observed in Film-2, which is formed with 2 wt% nanophase composite particles.

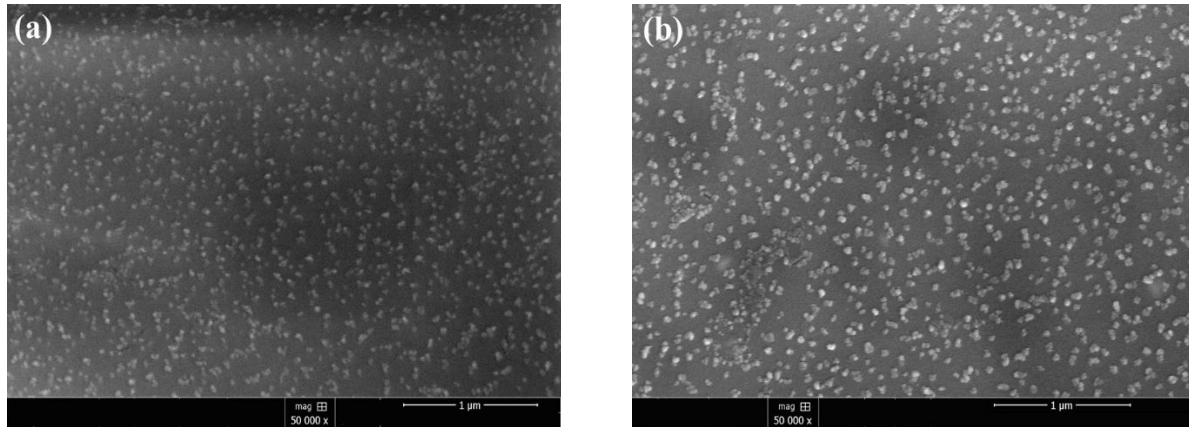


Figure 4. (a) shows the FESEM image of Film-1 *i.e.*, nanocomposite film prepared by loading of 1 wt% of nanophase composite particles. (b) shows the surface morphology of Film-2 *i.e.*, nanocomposite films prepared by loading of 2 wt% nanophase composite particles. It is clearly evident that the density of composite nanoparticles is significantly enhanced in Film-2 in comparison to Film-1.

Raman spectrum of Film-2 has been given in Figure 5 as a representative spectrum of the nanocomposite films. Raman spectrum clearly shows the presence of D- and G-bands related to graphene as well as other Raman bands related to chromium oxides phases of CrO_2 , Cr_2O_3 , and Cr_3O_8 [75-91]. All the representative bands in the Raman spectrum have been given in

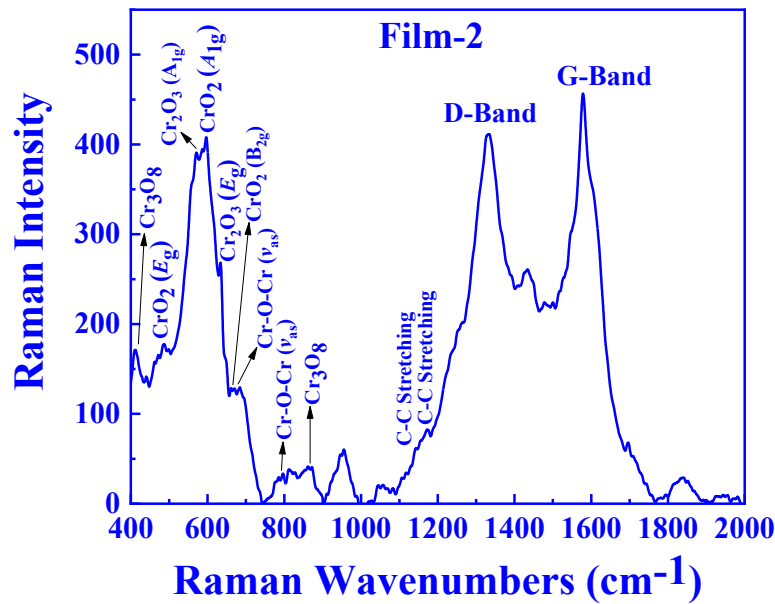


Figure 5 shows a representative Raman spectrum of graphene-enhanced nanocomposite films reinforced with 2 wt% multigranular chromium oxides composite nanoparticles. The presence of characteristic D- and G-bands in the Raman spectrum symbolizes the rGO. Other Raman bands from rutile type tetragonal half-metallic ferromagnetic CrO_2 and other phases of chromium oxides are also evident from the spectrum.

Table IV. Raman bands in graphene-enhanced PVA nanocomposite films with 2 wt% composite nanoparticles.

S. No.	Raman Wavenumber	Raman Band	Assignment in Literature	Reference	Ref. No.
1.	413 cm^{-1}	Wagging vibrations of -CO groups	421 cm^{-1}	Montana <i>et al.</i>	[75]
2.	472.1 cm^{-1}	CrO_2 (E_g)	458 cm^{-1}	Coey <i>et al.</i>	[76]
3.	488.2 cm^{-1}	Bending vibrations of CO groups	475 cm^{-1}	Montana <i>et al.</i>	[75]
4.	557.2 cm^{-1}	Shoulder band	524 cm^{-1}	Montana <i>et al.</i>	[75]
5.	572.9 cm^{-1}	Cr_2O_3 (A_{1g})	551 cm^{-1}	Ahlberg <i>et al.</i>	[77]
6.	583.9 cm^{-1}	CrO_2 (A_{1g})	570 cm^{-1}	Coey <i>et al.</i>	[76]
7.	595.2 cm^{-1}	Wagging vibrations of OH groups	580 cm^{-1}	Montana <i>et al.</i>	[75]
8.	637.1 cm^{-1}	Cr_2O_3 (E_g)	609 610 cm^{-1}	Ahlberg <i>et al.</i> Monnereau <i>et al.</i>	[77] [78]
9.	670.5 cm^{-1}	OH wagging vibrations	521 - 695 cm^{-1}	Montana <i>et al.</i>	[75]

			646 cm ⁻¹	Rao <i>et al.</i>	[79]
10.	683.8 cm ⁻¹	CrO ₂ (B _{2g})	682 cm ⁻¹	Coey <i>et al.</i>	[76]
11.	697.3 cm ⁻¹	Cr ₂ O ₃ (B _{2g})	700 cm ⁻¹	Ahlberg <i>et al.</i>	[77]
12.	796.3 cm ⁻¹	Cr ₃ O ₈ vibrations	743 cm ⁻¹	Maslar <i>et al.</i>	[80]
			732 cm ⁻¹	Monnereau <i>et al.</i>	[78]
13.	812.6 cm ⁻¹	Cr-O-Cr vibrations	809.2 cm ⁻¹	Dwivedi <i>et al.</i>	[67]
			772 cm ⁻¹	Ramsey <i>et al.</i>	[81]
14.	861.5 cm ⁻¹	Symmetric stretching vibrations of C-C skeleton	852 cm ⁻¹	Montana <i>et al.</i>	[75]
15.	871.9 cm ⁻¹	Cr ₃ O ₈ vibrations	886 cm ⁻¹	Maslar <i>et al.</i>	[70]
			878 cm ⁻¹	Monnereau <i>et al.</i>	[78]
16.	943.6 cm ⁻¹	C ₃ symmetric stretching of CrO ₃	942 cm ⁻¹	Vuurman <i>et al.</i>	[82]
		CH ₂ rocking vibrations	914 cm ⁻¹	Montana <i>et al.</i>	[75]
		Twist mode of isotactic PVA	912 cm ⁻¹	Prosanov <i>et al.</i>	[83]
17.	956.7 cm ⁻¹	Cr ₃ O ₈ vibrations	968 cm ⁻¹	Maslar <i>et al.</i>	[80]
18.	1045.7 cm ⁻¹	C-O stretching vibrations	1023 cm ⁻¹	Montana <i>et al.</i>	[75]
19.	1057.3 cm ⁻¹	C-O stretching vibrations	1093 cm ⁻¹	Montana <i>et al.</i>	[75]
20.	1143.5 cm ⁻¹	C-O stretching vibrations	1124 cm ⁻¹	Montana <i>et al.</i>	[75]
			1121 cm ⁻¹	Rao <i>et al.</i>	[79]
21.	1172.3 cm ⁻¹	C-O-C and C-C & C-O stretching vibrations	1141 cm ⁻¹	Montana <i>et al.</i>	[75]
22.	1229.9 cm ⁻¹ - 1245.6 cm ⁻¹	C-H wagging vibrations	1235 cm ⁻¹	Montana <i>et al.</i>	[75]
23.	1305 cm ⁻¹	C-H & O-H stretching vibrations	1295 cm ⁻¹	Montana <i>et al.</i>	[75]
24.	1332 cm ⁻¹	sp ² -hybridized carbon (C) atoms related vibrations in graphene	1353 cm ⁻¹	Jung <i>et al.</i>	[84]
25.	1374 cm ⁻¹	CH ₂ wagging vibrations	1376 cm ⁻¹	Montana <i>et al.</i>	[75]
			1343 cm ⁻¹	Rao <i>et al.</i>	[79]
26.	1390.3 cm ⁻¹	Wagging vibrations of -CH ₂	1372 cm ⁻¹	Montana <i>et al.</i>	[75]
27.	1447.3 cm ⁻¹	O-H bending vibrations	1428 cm ⁻¹	Rao <i>et al.</i>	[79]
28.	1480.9 cm ⁻¹	Deformation vibrations of -CH	1490 cm ⁻¹	Prosanov <i>et al.</i>	[83]
29.	1502.8 cm ⁻¹	Overtone of D-band	1500 cm ⁻¹	Couzi <i>et al.</i>	[95]
		D'-band	1530 cm ⁻¹	Yoshimura <i>et al.</i>	[86]
30.	1540.0 cm ⁻¹	C=O stretching vibrations belonging to deficiencies in rGO	1567 cm ⁻¹	Wang <i>et al.</i>	[87]
				Geim <i>et al.</i>	[88]
31.	1578.3 cm ⁻¹	Breathing vibrations of κ-point phonons of type A _{1g} symmetry in graphene	1605 cm ⁻¹	Jung <i>et al.</i>	[84]
			1590 cm ⁻¹	Couzi <i>et al.</i>	[85]
32.	1611.9 cm ⁻¹	Overtone of D-band	1604 cm ⁻¹	Couzi <i>et al.</i>	[85]
		D'-band	1615 cm ⁻¹	Yoshimura <i>et al.</i>	[86]
33.	1696.8 cm ⁻¹	Broad band	1721 cm ⁻¹	Montana <i>et al.</i>	[75]
34.	1780.0 cm ⁻¹	C=O stretching vibrations	1723 cm ⁻¹	Srikanth <i>et al.</i>	[89]
		Residual acetate	1720 cm ⁻¹	Montana <i>et al.</i>	[75]
35.	1840.2 cm ⁻¹	Linear carbon chains	1830 cm ⁻¹	Mariotto <i>et al.</i>	[90]

Table IV. Raman bands from half-metallic ferromagnetic CrO₂ are present at 472.1 cm⁻¹ (E_g), 583.9 cm⁻¹ (A_{1g}) and 683.8 cm⁻¹ (B_{2g}) in the nanocomposite films [76]. Coey *et al.* have marked these Raman bands of half-metallic ferromagnetic CrO₂ at 458 cm⁻¹, 570 cm⁻¹ and 683.8 cm⁻¹

in their exemplary work on Raman spectroscopy of CrO₂ [76]. Raman bands related to antiferromagnetic Cr₂O₃ are present at 572.9 cm⁻¹ (A_{1g}) and 697.3 cm⁻¹ (B_{2g}) in the spectrum [77]. Ahlberg *et al.* reported these bands of Cr₂O₃ at 551 cm⁻¹ and 700 cm⁻¹, respectively [77]. CrO₃ also shows its presence in nanocomposite films at 812.6 cm⁻¹ for Cr-O-Cr vibrations and at 943.6 cm⁻¹ related to C₃ symmetric stretching of CrO₃ [67, 81]. Raman bands related to Cr₃O₈ phase appear at 796.3 cm⁻¹, 871.9 cm⁻¹ and 956.7 cm⁻¹. These bands have been assigned at 743 cm⁻¹ by Maslar *et al.* [80] and at 732 cm⁻¹ by Monnereau *et al.* [78], at 886 cm⁻¹ by Maslar *et al.* [80] and 878 cm⁻¹ by Monnereau *et al.* [78], and at 968 cm⁻¹ by Maslar *et al.* [80], respectively. The bands at 1332 cm⁻¹ (D-band) and 1578.3 cm⁻¹ (G-band) are assigned to *sp*²-hybridized carbon (C) atoms related vibrations in E_{2g} phonon mode in graphene and breathing vibrations of κ -point phonons of type A_{1g} symmetry. These bands have been assigned at 1353 cm⁻¹ by Jung *et al.* [84] and at 1600 cm⁻¹ by Jung *et al.* [84] and at 1590 cm⁻¹ by Couzi *et al.* [85], respectively. The D-band owes its presence to the inter-valley double resonance mechanism of electron-hole by the defects that is responsible for the generation of transverse-optic (TO) mode of phonon dispersion in the vicinity of the Γ -point. D-band signifies the presence of anomalies in the carbon nanomaterial that include vacancies, disorders in grain boundaries and even non-crystalline or semicrystalline carbon linkages. The ratio of I_D/I_G gives 0.90 which indicates the presence of defects in the carbon nanomaterial only to an extent. Jung *et al.* have reported the I_D/I_G ratio of 0.96 in case of graphene and a ratio of 1.00 in case of graphene oxide [84]. The band at 1502.8 cm⁻¹ is assigned to the overtone of D-band, while the band at 1611.9 cm⁻¹ is assigned to the overtone of G-band. Couzi *et al.* mentions an overtone of D-band at 1500 cm⁻¹, while another overtone at 1604 cm⁻¹ [85] Yoshimura *et al.* mentions the presence of D''-band at ~1530 cm⁻¹ and D'-band at ~1615 cm⁻¹ [86]. The D'-band happens due to an intra-valley double resonance mechanism of electron-hole induced by the defects generating a longitudinal-optic (LO) phonon mode in the vicinity of the Γ -point. These bands,

D''-band and D'-band, are usually connected to the presence of amorphous carbon or D₃ and to the integration of aliphatic moieties on fundamental graphite parts or D₄. In the current study, the presence of D₃ band is owed to amorphous carbon chains contained in the polymer and D₄ band is ascribed to the connection of aliphatic parts onto the graphitic chains in the nanocomposite films. The I_G-peak is owed to the stretching vibrations of *sp*²-bonded C-C chains across the different planes. Normal first order Raman scattering mechanism happens in graphene so that a phonon mode of double-degenerate nature gets associated at the Brillouin zone center. In contrast, I_{2D}-peak is owed to a second-order mechanism in which in-plane transverse optic (iTO) configurational phonons are associated closer to the K-point. I_D-peak is also a second-order phenomenon in which one phonon in iTO mode along with one defect are involved [91]. The band at 1540 cm⁻¹ is assigned to C=O stretching vibrations which happens due to the occurrence of left-out O₂-containing functional groups in rGO [73, 74]. Wang *et al.* ascribes this band at 1567 cm⁻¹ in a work on PVA/rGO hydrogel composite materials [87]. All other bands, their assignments and correlated references are listed in Table IV.

Intergranular tunneling or spin-dependent tunneling (SDT) in an associated Coulomb gap and the spin-independent hopping (SIH) phenomenon based spin-polarized transport can be expresses in form of a phenomenological equation given as follows [67, 68, 92-101],

$$R_{\text{tun}} \propto (1 + P^2 m^2)^{-1} \exp(\Delta/T)^{1/2} \dots\dots\dots(4)$$

In the above equation (4), *m* is the relative magnetization with *m* = *M*/*M_s*, where *M* denotes the magnetization and *M_s* is saturation magnetization. Δ-value represents the Coulomb blockade and is a direct function of the Coulomb electrostatic charging energy as well as insulating barrier width. Thus, the electrical conduction happens by (i) spin-dependent channel with the

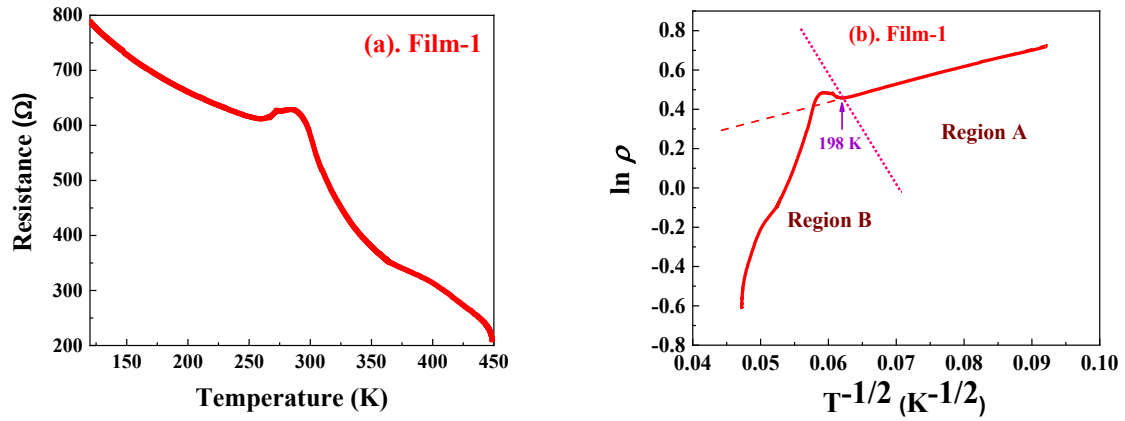


Figure 6. (a) shows the variation of resistance (R) as a function of advancing temperature (T) which depicts as a typical semiconductor nature of Film-1. Resistance is $800 \, \Omega$ at $77 \, \text{K}$ which decreases to $200 \, \Omega$ at $450 \, \text{K}$, which is 75% decrement in case of Film-1. (b) shows the Efros-Shklovskii variable range hopping character contained in Film-1 by plot of $\ln \rho$ vs. $T^{-1/2}$. The linear character is followed till $198 \, \text{K}$, after which the resistivity varies in a non-linear manner as a function of $T^{-1/2}$.

conduction varying as exponential of $T^{-1/2}$ typically at low temperatures, and (ii) spin-independent channel obeying the dependence on power law temperature. Figure 6(a) & 6(b) show the electrical behaviours of Film-1 by plotting R vs. T and $\ln \rho$ vs. $T^{-1/2}$. Figure 6(a) reflects a typical semiconductor nature of Film-1 since resistance decreases as the operating temperature increases. Resistance is $800 \, \Omega$ at $77 \, \text{K}$ which decreases to $200 \, \Omega$ at $450 \, \text{K}$ in case of Film-1. Efros-Shklovskii variable range hopping appears to be the major mechanism as visible from Figure 6(b) [87]. Linear nature of $\ln \rho$ vs. $T^{-1/2}$ plots suggest the prevalence of spin-polarized electrical transport, while non-linear part gives information about the SIH type of electrical transport.

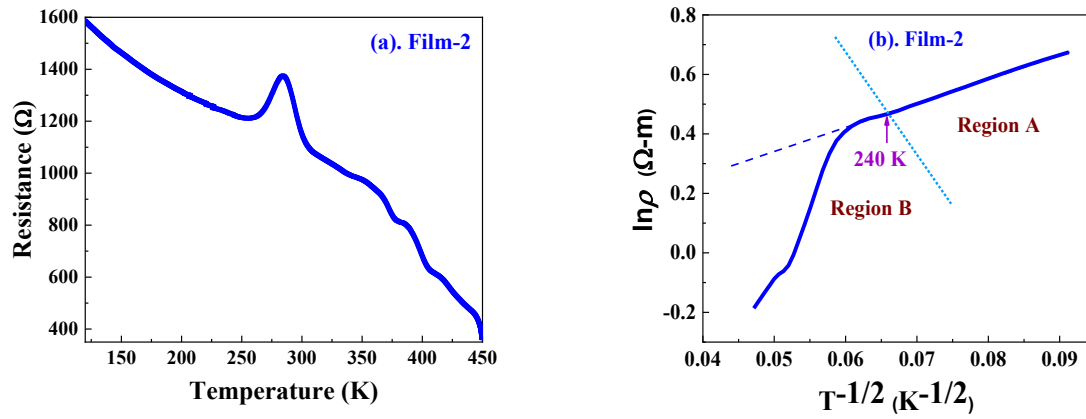


Figure 6. (a) shows the variation of resistance (R) as a function of advancing temperature (T) which depicts in form of a typical semiconductor nature in Film-2. Resistance is $1585 \, \Omega$ at $120 \, \text{K}$ which decreases to $366 \, \Omega$ at $450 \, \text{K}$, which is $\sim 77\%$ decrement in case of Film-2. (b) shows the Efros-Shklovskii variable range hopping character contained in Film-2 by plot of $\ln \rho$ vs. $T^{-1/2}$. The linear character is followed till $240 \, \text{K}$, after which the resistivity varies in a non-linear manner as a function of $T^{-1/2}$.

Figures 7(a) and 7(b) show the magnetoresistance (MR) curves of Film-1 and Film-2, respectively, acquired in an out-of-plane geometry of application of the magnetic field (H). The

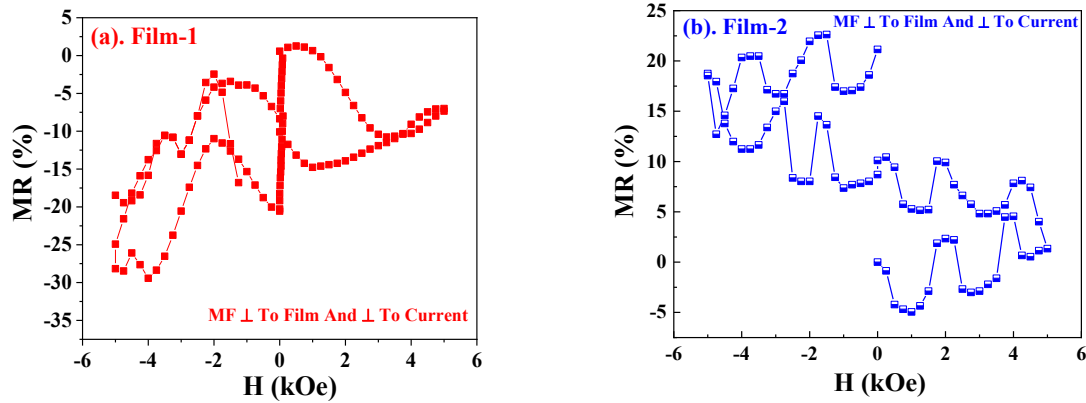


Figure 7. (a) shows a total change of 31% in MR% of Film-1 in an out-of-plane geometry of application of the magnetic field with applied current also in a perpendicular direction to H . (b) shows a change of 27.5% in MR% of Film-2 with H applied in a perpendicular direction to the film plane and also to the current. This time the large contribution comes from $+22.5\%$ MR change with -5% on the negative side of MR.

MR curve in Fig. 7(a) shows a closed loop MR change of -31%. Although the closed loop is present in the MR curve of Fig. 7(a), yet it does not show a full hysteretic signature. This means that coherent rotation of domains as a function of sweep of H does not follow a singular pattern. The loop shapes for both forward and reverse sweeps of H are not the identical ones. Film-2 also shows an MR of $\sim -27.5\%$ but in a mixed mode in which $+22.5\%$ and -5% happens. The notable thing is the appearance of closed loop curve of MR in Film-1 but with different loop shapes. Different loop-shapes on positive and negative sweeps of H occur due to the difference in resistances on the two sides of H sweep. A major contribution to MR comes from the magnetic moments of the defected graphene (rGO). Besides, magnetic moments contribution from half-metallic ferromagnetic CrO_2 , paramagnetic chromium oxides & ordering from canted spins of antiferromagnetic Cr_2O_3 under H are other inclusive factors. In the totality of the structure, Film-1 shows the loop nature with both the loops being uneven. This means the traced path in both sweeps of the H is completely different. Film-2 does not show any type of loop formation in either of the H sweeps, and an open-loop is traced out of the electrical signal under the H -effect. This clearly shows that the linkages between magnetic components do not follow any serial connection to result in retracing of the MR signal.

Negative MR of 30% in Fig. 7(a) is attributed to the weak localization effect, while positive contribution of $+1\%$ is due to less effective weak antilocalization (WAL) [104]. Negative MR has been ascribed to weak localization in previously published reports on both pure [103-106] and intercalated graphite fibres [107-110]. Similarly, appearance of positive MR in Film-2 is attributed to weak antilocalization (WAL) and negative MR to weak localization (WL). The 2D weak-localization is related to magnetic field in the following manner [102, 103],

$$\Delta R_{sq}(H) = -(R_{sq})_0^2 F(H, \tau) \dots\dots\dots(5)$$

Here, F is a function that depends on H and the different collision times τ for elastic and inelastic scattering. In the geometry of H perpendicular to film plane and also to the current, a mixing of

MR can be observed owed to the interplay of stronger weak localization (WL) for negative MR and to less effective weak antilocalization (WAL) for positive MR. Wyder *et al.* ascribes the mixing of MR in polymer-graphite composite materials to the sum of positive classical along with a negative quantum-mechanical contribution due to the localization effect, expressed as follows [88],

$$\frac{\Delta R}{R_0} \cong \frac{[\Delta R_{sq}(H)]_{classical} + [\Delta R_{sq}(H)]_{quantum}}{(R_{sq})_0 + (R_s)_0} \dots\dots\dots(6)$$

Here, R_{sq} is the square resistance and R_s is the magnetic field independent contact resistance.

Wyder *et al.* have discussed the electrical transport mechanism in polystyrene-graphite material system for fields < 10 T and at low temperatures. Resistivity of the polystyrene-graphite material system was more than 10^4 for high concentrations of graphite in comparison to that of the pristine graphite. Negative MR heavily depended on graphite concentrations in the system, and decreased as a function of increasing concentration of the graphite. Subramanian *et al.* observed negative MR of -1.4% in CoFe₂O₄:rGO ratio of 1:1, while an MR of -1% for 2:1 and 3:1 material composition at room temperature [49]. In CoFe₂O₄:rGO material system, CoFe₂O₄ is electrically insulating and electrical transport happens in the material system through the mediation of rGO among the nanoparticles. CoFe₂O₄ nanoparticles possess magnetic moments and act as the scattering centers during the conduction of electrons to develop electron-spin scattering. This electron-spin scattering is controlled under the application of a magnetic field which induces parallel orientation of the moments. Magnetocapacitance effect of CoFe₂O₄ nanoparticles was also performed which further confirmed the parallel alignment of magnetic moments under an applied field. Ando *et al.* have critically reviewed the weak localization phenomenon in graphene [100]. Heremans *et al.* have performed an experimental study in conjunction with theoretical insights on MR in graphitic fibres [115]. Bright's theory successfully explains the MR nature at low fields i.e., $H < 1.0$ T in this work [116], while field-based impurity scattering is the dominant mechanism at high fields. As per Bright's theory,

$\Delta\rho(H)/\rho_0$ assumes a flat shape in vicinity of 1.4 T, while it begins acquiring less negative values as a function of increasing H . Bright's theory was applied by the inclusion of magnetic field dependent scattering phenomenon to the MR data to explain the nature in graphitic fibres.

Substantially large positive MR of 22.5% in Film-2 is ascribed to the weak antilocalization (WAL), while negative MR of 5% is attributed to weak localization (WL). Thus, it is a combination of WAL and WL which is responsible for the appearance of mixed MR in significant values in Film-2. This is contrary to the appearance of majorly negative MR values in Film-1 in which WL dominates substantially throughout the H sweep. However, there is no loop formation in MR in Film-2 in the same geometry of measurement, and the large difference in MR on the positive and negative sweeps of MR is also evident from Figure 7(b). This is a completely non-hysteretic character of MR of Film-2 in comparison to Film-1 which shows some sort of loop formation as a function of H sweep. This is pertinent here to shed light on the research reports matching closely to the current work. Zhang *et al.* have in-situ deposited FeCo nanoparticles over rGO to develop magnetic field sensor [117]. Abrikosov's quantum model for layered materials was detected to be suitable to establish the quantum magnetoresistance in hybrid rGO. High MR-values of $21.02\% \pm 5.74\%$ were achieved at room temperature at $H = 10$ kOe. The authors claimed the hybrid-rGO material based sensor to be useful to integrate in a wireless system for actual resolution of light doses of electromagnetic radiation occurring from a working mobile phone. Sarkar *et al.* have studied magnetodielectric effects in rGO-*poly*(vinyl alcohol) (PVA), rGO-*poly*(acrylic acid) (PAA) and rGO-*poly*(methyl methacrylate) (PMMA) nanocomposite materials [118]. The magnetodielectric properties of these nanocomposite materials have been studied in magnetic field upto a maximum of 1.2 T. For the enhancement of H from 0 to 1 T, ϵ' reduces by 5% at 100 kHz of operational frequency in rGO-PVA nanocomposites, and reduction happens by 4% in rGO-PAA nanocomposite materials. On the other hand, there is increment of 4% in rGO-PMMA nanocomposites. The

typical magnetodielectric properties were ascribed to interfacial effects happening between the polymers and rGO.

The main reasons for the appearance of MR in both Film-1 and Film-2 can be summarized as follows :

- (i) Defects and/or disorders in rGO are the source of unpaired electrons that impart magnetic moments which prefer to align in the direction of applied H
- (ii) Crystal structure and crystallite size of chromium oxide nanophase composite particles in a typical local symmetry, especially for ferromagnetic & antiferromagnetic phases
- (iii) Distribution of the rGO lattice in addition to CrO_2 lattice in a typical microstructure
- (iv) Complex exchange interactions between CrO_2 particles; disorder-induced spins & CrO_2 -rGO magnetic moments in a typical local symmetry
- (v) Spin-dependent scattering and also intergranular tunneling across the whole matrix
- (vi) Hopping in rGO-induced polymer in case of electrical transport
- (vii) Magnetic domain or interfacial effects at the conductor-insulator boundaries.
- (viii) Canted spins arising from various phases included in the whole specimen, chiefly from antiferromagnetic phase; due to specific alignment of magnetic moments
- (ix) Macroscopic coupling of CrO_2 and rGO magnetic moments within the matrix
- (x) Intercrossing conduction trajectories are detrimental to the appearance of hysteretic MR
- (xi) Non-hysteretic MR suggests that the MR response is not directly linked to the orientation of magnetic domains with respect to sweep of H
- (xii) Lesser effective coherent rotation of domains as a function of sweep of H which generates non-hysteretic MR curve

All the aforementioned points are important in the resolution of an equilibrium spin-band structure in the nanocomposite films. A distinct spin-entropy along with the spin-dynamics is determined by (i) - (xii) points in the nanocomposite films of rGO reinforced with chromium

oxides nanophase composite particles. Thermodynamically, overexposed surfaces, *e.g.*, rGO in 2D enhanced surfaces along with nanophase composite particles of high spin-entropy, can affect the final spin-architecture including spin-dynamics in such nanocomposite materials.

Conclusion

The work highlights the appearance of room temperature MR in rGO-polymer nanocomposite films with (a) 1 wt% and (b) 2 wt% inclusions of multigranular & multiphasic chromium oxides composite nanoparticles consisting of half-metallic ferromagnetic CrO₂ phase. Film-1 composed of 1 wt% composite nanoparticles shows a total change in MR of 31%. The total MR change of 31% is basically a mixed MR consisting of -30% and +1% in case of Film-1. On the other hand, Film-2 does not reflect any loop-type response in MR change during the field sweep. Basically, in case of Film-2, MR% happens in form of open loop. A total change of mixed 27.5% MR happens in case of Film-2 with 22.50% of negative MR and 5.0% of positive MR.

PVA polymer offers sound matrix for the formation of flexible platform for spin-memory & spin-logic devices magneto-optical devices and magnetic sensors. However, PVA is an insulating phase offering higher resistances and inclusion of rGO with PVA brings down resistance to few hundreds of ohms. In this way, the problem of lower electrical conduction is sorted out in a 2D nanocomposite formation. Magnetic moments are imparted by both rGO and the chromium oxides nanophase composite particles consisting of half-metallic ferromagnetic CrO₂. Appearance of MR in the two films happens because of magnetic moments imparted by defects and/or disorders in rGO, crystal structure of CrO₂ in a local symmetry, crystallite size as well as morphological features of CrO₂, distribution of disordered lattice and CrO₂ of ordered lattice in a typical symmetry of the composite microstructure, complex exchange interactions between different constituents of the nanocomposite films, intergranular tunneling of electrically conductive carriers, phenomenon of hopping in transport mechanism, interfacial

effects at the conductor-insulator boundaries and canted spins in a local geometry. Besides, lower response of the domains to coherent rotation as a function of H -sweep and poor linking of MR signal to the orientation of domains are the factors responsible for the appearance of non-hysteretic MR. Besides these, other factors are the contribution from paramagnetic spins of Cr_3O_8 , as well as from canted spin generating from antiferromagnetic Cr_2O_3 and Cr_2O , and those from magnetic moments of defect-ridden rGO. Magnetic moments from different contributory factors are not linked in a serial chain manner or in an ordered percolative manner so that the non-hysteretic MR curves happen.

Although closed loop MR happens in Film-1, MR is non-hysteretic in nature because the curves do not retrace the original path in back and forth sweeps of the magnetic field. This type of MR is useful for the development of sensors for determination of speed of wheels in automobiles, biomedical magnetometers, and also finds applications in non-volatile memory devices, such as, magnetic random access memory (MRAM). Besides, spin-dependent tunneling is a major mechanism of electrical transport in both types of nanocomposite films. This is also helpful in bringing down the operating temperature of magnetoresistance signals down to room temperatures by preserving spin-polarized conduction. In the near future, we plan to develop materials or films which can show much efficient magnetoresistance signals at room temperature under much lower fields.

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Data Availability Statement

All data generated or analysed during this study are included in this published article.

Declarations

Competing Interests : The authors declare no competing interests.

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