**RESEARCH PAPER** 

# Recent trend in graphene for optoelectronics

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Abstract This study analyzes the scientific knowledge diffusion paths of graphene for optoelectronics (GFO), where graphene offers wide applications due to its thinness, high conductivity, excellent transparency, chemical stability, robustness, and flexibility. Our investigation is based on the main path analysis which establishes the citation links among the literature data in order to trace the significant sequence of knowledge development in this emerging field. We identify the main development paths of GFO up to the year 2012, along which a series of influential papers in this field are identified. The main path graph shows that knowledge diffusion occurs in key subareas, including reduced graphene oxide, chemical vapor deposition, and exfoliation techniques, which are developed for the preparation and applications of

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GFO. The applications cover solar cells, laser devices, sensing devices, and LCD. In addition, the main theme of GFO research evolves in sequence from smallgraphene-sample preparation, to large-scale film growth, and onto prototype device fabrication. This evolution reflects a strong industrial demand for a new transparent–conductive film technology.

Keywords Graphene  $\cdot$  Graphene for optoelectronics  $\cdot$  Literature survey  $\cdot$  Citation analysis  $\cdot$  Main path analysis

#### Introduction

Graphene is the name given to a flat monolayer of  $sp^2$ hybridized carbon atoms arranged in a two-dimensional honeycomb lattice. AK Geim, Professor at University of Manchester, and his team in 2004 obtained monolayer-graphene by means of 3M tape exfoliation and confirmed the existence of graphene (Novoselov et al. 2004, 2005). Today, graphene has become one of the focuses of science and technology research. A single sheet of graphene is a zero-band gap semiconductor having extremely high carrier mobility and very low absorbance (2.3 %) of visible light (Nair et al. 2008; Eigler 2009). Its high conductivity, excellent transparency, flexibility, thinness, and robustness make graphene a very prospective material for optoelectronic devices such as displays, touch screens, light-emitting diodes, and solar cells (Geim

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2009; Eda and Chhowalla 2010; Wassei and Kaner 2010; Bonaccorso et al. 2010; Allen et al. 2010).

The research and development activities of graphene for optoelectronics (GFO) have been growing very fast ever since the discovery of monolayer-graphene. While in early 2004, there was only one academic article in this field, by April 2012, the number of articles had increased up to 1233. At such a fast growth rate, it is difficult to see the whole picture of the field's development for newcomers even with the help of several excellent physical- science-oriented review papers on graphene. A macroscopic and comprehensive view on the development trend of GFO based on a specific statistical analysis of the relevant literature would indeed be insightful. This study achieves such a goal by applying the main path analysis on the citation data of over 1000 GFO academic papers to trace their development paths. Through the analysis, we identify a set of papers that play the central role in the GFO development and also clarify the major GFO activities in recent years.

Citations in academic articles contain rich information on how knowledge disseminates. Garfield et al. (1964) suggested that it is possible to "write the history of science" through analyzing citation relationships among science publications. Hummon and Doreian (1989) adopted Garfield's concept and laid out the groundwork for the main path analysis. The method has since been applied to several science and technology disciplines to uncover their development trajectories (Liu et al. 2013; Liu and Lu 2012; Verspagen 2007). The best feature of the method is that it simplifies a complicated citation network by extracting out important paths in the network. It provides a satellite view of the citation network, in which only significant paths remain and paths of lesser significance disappear. With the main path graph, the important and influential research papers emerge on a timeline, and the main stream of knowledge evolution of GFO is recognizable.

#### Methodology

The main path analysis helps comprehend the GFO development to a more detailed level. This section briefly introduces the method.

Hummon and Doreian (1989) first introduced the main path analysis and used citation information in academic papers or patents to trace the main idea flow in a scientific discipline. When a publication cites a previous work, presumably, knowledge flows from the previous work to the citing publication. The method is network-based; the scientific publications are seen as nodes of a network, and citation information is used to establish links among nodes. The citation network thus created is a non-weighted directional network.

Tracing the flow of ideas in a small citation network may be easy, but the task's difficulty increases as the network grows larger. Hummon and Doreian (1989) suggested a way to simplify the task in a large citation network: tracing only the 'main path'. Identifying the importance of each citation link in the network is the first step in finding the main path. The importance of each citation can be measured by counting the number of times a citation link has been traversed if one exhausts the search from a set of starting nodes to another set of ending nodes. There are several variations of ways to conduct the count. The literature mentions node pair projection count (NPPC), search path link count (SPLC), search path nodes pair (SPNP), and search path count (SPC) (Hummon and Doreian 1989; Batagelj 2003). These counts are similar, but subtle differences exist among them. We choose to use SPC as it is recommended by Batagelj (2003) as the first choice.

In a citation network, a 'source' is a node that is cited, but cites no other nodes; a 'sink' is a node that cites other nodes, but is not cited. In other words, sources are the origins of knowledge, while sinks are the end points of knowledge dissemination. We use a simple citation network in Fig. 1 to demonstrate how SPCs for each link are calculated. The network has two sources, A and B, and four sinks, C, D, E, and F. There are many alternative paths to go from the sources to the sinks. Assuming that one exhausts searching all paths from all the sources to all the sinks, SPC for each link is defined as the total number of times the link is traversed. For example, link J-C has SPC value of 2 because it is passed through by paths A-H-J-C and B-H-J-C. Link B-I's SPC value is 4 as it is traversed by four paths: B-I-F, B-I-G-D, B-I-G-E, and B-I-E. In the example network, B-I and H-J have the largest SPC value. The larger the SPC value is, the more important the link's role is in transmitting the knowledge.

After the SPC value for each citation link is calculated, we begin the key-route search procedure as follows.

1. Select a key-route; it is usually the link that has very high traversal count.

#### Fig. 1 SPC example



- 2. Search forward from the end node of the key-route until a sink is hit.
- 3. Search backward from the start node of the keyroute until a source is hit.

The results of the search are what one calls the keyroute main path (Liu and Lu, 2012). Upon selecting more than one key-routes, one obtain multiple key-route paths.

#### Data and basic statistics

#### Data

We adopt ISI Web of Science (WOS) as the data source of this study. The databases within WOS selected for this study include Science Citation Index Expanded (SCIE), Social Sciences Citation Index (SSCI), and Conference Proceedings Index-Science (CPI-S). In the graphene research, the most difficult issue is how to exfoliate graphite into individual graphene sheets, but this was solved by Geim and his team in 2004 (Novoselov et al. 2004, 2005). Since then, it has aroused an extraordinary amount of interest, accompanied by numerous research reports. We, therefore, take data retrieved from 2004 up until April 2012.

Those papers about GFO are searched and retrieved from WOS with great care. The query to retrieve these studies in the literature is based on the combination of several keywords<sup>1</sup>.

During the search, we found there are several papers that review the progress of GFO rather than propose novel ideas. These papers are heavily cited and offer not much technical contribution. Therefore, we excluded 53 review papers from the dataset after manually examining them. Another issue is that several important papers on the subject of graphene preparation published in the earlier period are not identified through the query mentioned above because these early papers tend to focus on fundamental research without specifying optoelectronic applications. It was recognized later that large-size graphene preparation laid out the basic foundations for the GFO applications and development. These papers are important and should not be excluded in the discussion of GFO development. Therefore, we conducted a second round search using such keywords as "preparation" OR "reduced graphene oxide" OR "CVD graphene" OR "exfoliated graphene" and then selected the top ten highest-citation papers to incorporate into our dataset. The addition of these papers helps us find the technological foundations of GFO. In

<sup>&</sup>lt;sup>1</sup> Graphene AND (optoelectronic\* OR photonic\* OR "transparent electrode\*" OR "transparent conduct\*" OR

Footnote 1 continued

photovoltaic\* OR photodetect\* OR "touch panel\*" OR "touch screen\*" OR "liquid crystal\*" OR "flexible display\*" OR "ultrafast laser\*" OR "fiber laser\*" OR absorber\* OR "terahertz device\*" OR "terahertz emission" OR "optical limit\*" OR "light-emitting device\*" OR "optical device\*" OR "ultrafast optical\*" OR photoconduct\* OR photocurrent\* OR "solar cell\*" OR photoluminescence\* OR "indium tin oxide" OR "ITO" OR "thin-film transistor") (\*denotes the wildcard used in most search engines).

the end, 1233 papers were included in the final dataset for further analysis.

Table 1 displays the top 20 papers with the highest rate of citation. Among them, the paper published by Stankovich et al. (2006) enjoyed the highest number of citations. One may wonder why the most notable papers on graphene research, such as Novoselov et al. (2004) and Novoselov et al. (2005), were not included in this study's dataset. This is because they are out of the scope of this research, i.e., they neither focus on optoelectronic applications, nor do they emphasize chemical preparation such as rGO, CVD, etc.

### Journal statistics

There are many journals that are especially supportive of the GFO field. Table 2 shows six journals that have published more than 50 GFO papers: *Applied Physics Letters*, *ACS Nano*, *Journal of Materials Chemistry*, *Nano Letters*, *Journal of Physical Chemistry C*, and *Physical Review B*.

We apply both the g-index and the h-index to recognize journals' influence. Hirsch (2005) proposed an index to quantify an individual's scientific research output through the use of citation information. The Hirsch index h is defined as "the number of papers with citation number  $\geq h$ ". The index is conceptually simple and has been successfully implemented to capture scholars' influence in various scientific fields (Bornmann and Daniel 2005; Mingers 2009; Saad 2010). The *h*-index, nevertheless, does not take citation scores of a researcher's top articles into account. The g-index is an improvement over the h-index on this specific issue (Egghe 2006). We take the position that the g-index and the h-index are complementary to each other and therefore apply both of them to find out which journal is more influential in this field.

Table 2 presents the top 20 journals according to their g-index and h-index. Nano Letters ranks number one, followed by ACS Nano, Carbon, Advanced Materials, Applied Physics Letters, Journal of Physical Chemistry C, Journal of the American Chemical Society, and Physical Review B.

### Main paths

We apply the main path analysis to explore the development trajectory of graphene research related to

optoelectronics. As with other science and technology disciplines, the trajectory of GFO development is not in single direction. Multiple sets of inter-connected paths form the evolution structure in the GFO field.

Figure 2 presents the main paths constructed from the top ten key-routes in the citation network. The figure is drawn using Pajek software (Batagelj and Mrvar 1998). In the figure, each node represents a paper. Lines connecting the nodes illustrate the citation relationship, while arrows represent the knowledge flow direction of these papers. The line thickness corresponds to its SPC value which reflects the significance of the citation relationship. The thicker a line is, the more important the role it plays in knowledge diffusion. Each paper on the main paths is assigned with a code which begins with the last name of the first author, followed by the initials of the subsequent co-authors and then the publication year. If there are duplicate codes, then lower case alphabet letters are appended at the end.

The top ten key-route main paths consist of 18 papers. The time span from the earliest pioneer to the most recent paper is around 6 years. Therefore, one important paper is published every 4 months on average, indicating that intensive studies prevail in this GFO field. The paths show that the concept of Stankovich's et al. (2006) original work was initially disseminated into several papers and then consolidated in ReinaJHNSBDK2009 (Reina et al. 2009) after diverse contributions from several researchers. Stankovich et al. (2006) were the first in our data to demonstrate a general approach for the preparation of graphene--polymer composites. The simple and scalable method, they provided for efficient production of chemically modified graphene, involves the chemical synthesis of graphite oxide, followed by its exfoliation into individual graphene oxide (GO) sheets, and their subsequent reduction. Easy dispersion and exceptionally low percolation threshold make rGO composites attractive for optoelectronics applications (Eda and Chhowalla 2010). For these reasons, more and more researchers have been attracted to this field. The earliest studies on this main path adopted reduced graphene oxide (rGO) and exfoliated graphene as the preparation method. However, Reina et al. (2009) initiate important research on the graphene fabrication by the chemical vapor deposition (CVD) method, in which high-quality large area graphene films can be grown on metals. From there, a new stream of research

Table 1 Top 20 cite	d papers in the graphene for optoelectronics fi	ble					
Authors	Title	Journal	Total citations	On the main paths	Citations in the citation network (N)	Self-citations in the citation network (n)	n/N (%)
Stankovich et al. (2006)	Graphene-based composite materials	Nature	1788	Y	247	11	4.45
Stankovich et al. (2007)	Synthesis of graphene-based nanosheets via chemical reduction of exfoliated graphite oxide	Carbon	1250	Y	209	6	4.30
Kim et al. (2009)	Large-scale pattern growth of graphene films for stretchable transparent electrodes	Nature	1211	Y	236	26	11.02
Li et al. (2008a)	Processable aqueous dispersions of graphene nanoshcets	Nature Nanotechnology	1146	Y	202	6	4.46
Li et al. (2009a)	Large-area synthesis of high-quality and uniform graphene films on copper foils	Science	898	Y	145	7	4.83
Reina et al. (2009)	Large area, few-layer graphene films on arbitrary substrates by chemical vapor deposition	Nano Letters	749	Y	139	4	2.88
Wang et al. (2008)	Transparent, conductive graphene electrodes for dye-sensitized solar cells	Nano Letters	743	Y	296	12	4.05
Stoller et al.	Graphene-based ultracapacitors	Nano Letters	718	Z	I		I
Eda et al. (2008)	Large-area ultrathin films of reduced graphene oxide as a transparent and flexible electronic material	Nature Nanotechnology	651	Y	195	12	6.15
Dikin et al. (2007)	Preparation and characterization of graphene oxide paper	Nature	645	Y	101	5	4.95
Hernandez et al. (2008)	High-yield production of graphene by liquid-phase exfoliation of graphite	Nature Nanotechnology	561	Y	102	15	14.70
Becerril et al. (2008)	Evaluation of solution-processed reduced graphene oxide films as transparent conductors	ACS Nano	450	Y	195	6	3.08
Bae et al.	Roll-to-roll production of 30-inch graphene films for transparent electrodes	Nature Nanotechnology	441	Z	I		I
Li et al. (2008b)	Highly conducting graphene sheets and Langmuir–Blodgett films	Nature Nanotechnology	428	Y	113	2	1.77
Blake et al. (2008)	Graphene-based liquid crystal device	Nano Letters	295	Y	88	2	2.27

Table 1 continued							
Authors	Title	Journal	Total citations	On the main paths	Citations in the citation network (N)	Self-citations in the citation network (n)	n/N (%)
Watcharotone et al. (2007)	Graphene-silica composite thin films as transparent conductors	Nano Letters	254	Y	102	6	8.25
Sun et al.	Nano-graphene oxide for cellular imaging and drug delivery	Nano Research	229	Z	I		I
Li et al. (2009b)	Transfer of large-area graphene films for high-performance transparent conductive electrodes	Nano Letters	224	Y	67	с,	4.48
Muszynski et al.	Decorating graphene sheets with gold nanoparticles	Journal of Physical Chemistry C	220	Z	I		I
Mohanty et al.	Graphene-based single-bacterium resolution biodevice and DNA transistor: interfacing graphene derivatives with nanoscale and microscale biocomponents	Nano Letters	216	z	I		I

has been devoted to this particular field up until now. As we shall see later in the top 40 key-routes, Kim et al. (2009) also situate at the juncture of initiating CVD method on the main path.

If we select the top 20 key-routes with the highest traversal counts to establish the main paths, then we get main paths that are broadly similar to Fig. 2, except the addition of node KimZJLKKAKCH2009 (Kim et al. 2009). However, if we increase the key-routes to 21, then the number of papers in the main path graph expands to 28 plus a new branch. The new branch has a special feature: all its papers are related to laser mode locking devices. If the key-routes are increased to 22, then the third branch appears and shows that reduced graphene oxide is the preparation method of GFO.

When we gradually increase the number of the keyroutes to 40, the main path graph as shown in Fig. 3 stays virtually the same as the top 22 key-routes. This suggests that the top 40 key-route main paths present a steady and important pattern which Fig. 3 illustrates. Table 3 lists the papers chronologically in the order of the publication date which are present on the top 40 key-route paths. They are the representative papers for the technological development and evolution of GFO because they exert significant influence on the followup research studies.

The top 40 key-route paths provide rich information on the development of GFO. We highlight several observations as below.

Technology evolution and main preparation methods

As shown in Table 4, the papers appearing in the main paths in the early years are those mostly focusing on the preparation of graphene; the application aspect of the material was not mostly discussed until 2010, when researchers started to touch upon preliminary applied products. In about 5 years, GFO technology has advanced from material preparation to applications. This indicates that GFO technological development has been making advances at a very good pace. It is comparable to the development process of the carbon nanotubes (CNT). Papers on electronic devices based on CNT have been published since 1997 (Tans et al. 1997; Bockrath et al. 1997), which is also about 5 years after the original report on CNT by Sumio Iijima in 1991. Not surprisingly, the evolution of GFO

Table 2 Top 20 most influential journals in the graphene for optoelectronics field

g-Index ranking	<i>h</i> -Index ranking	Journals	<i>g-</i> Index	<i>h-</i> Index	Years active	Total number of articles
1	2	Nano Letters	60	26	2007-2012	60
2	1	ACS Nano	48	29	2008-2012	93
3	7	Carbon	39	11	2005-2012	39
4	4	Advanced Materials	38	18	2008-2012	41
5	3	Applied Physics Letters	30	19	2007-2012	97
6	5	Journal of Physical Chemistry C	25	14	2008-2012	58
7	7	Journal of the American Chemical Society	23	11	2007-2012	23
8	6	Physical Review B	22	13	2006-2012	54
9	7	Small	19	11	2009-2012	19
9	7	Nature Nanotechnology	19	11	2007-2012	19
11	12	Advanced Functional Materials	17	9	2005-2012	17
11	15	Nano Research	17	8	2008-2012	17
11	17	Journal of Physical Chemistry Letters	17	7	2010-2012	21
14	12	Physical Review Letters	16	9	2007-2012	16
15	11	Journal of Materials Chemistry	15	10	2010-2012	66
16	12	Nanotechnology	14	9	2009-2012	39
17	15	Chemical Communications	12	8	2009-2012	20
17	17	Langmuir	12	7	2009-2011	12
19	19	Optics Express	11	6	2009-2012	18
20	20	Journal of Applied Physics	8	5	2007-2012	13

The journals are listed in order according to their g-index followed by their h-index and the total number of articles



Fig. 2 Top ten key-routes' main paths of graphene for optoelectronics development. Link weights are indicated with different line thicknesses. *Thicker lines* indicate heavier weights. The network is drawn using Pajek software



Fig. 3 Top 40 key-routes' main paths of graphene for optoelectronics development. Link weights are indicated with different line thicknesses. *Thicker lines* indicate heavier weights. The network is drawn using Pajek software

follows a pattern whereby fundamental research precedes application research, which is in conformity with the development of material technology. In view of the progress and potential of GFO technology, the governments of many countries have been and are still active in investing in this field. For example, the National Graphene Institute at the University of Manchester in UK has been awarded US\$59 million in 2012 from the Engineering and Physical Sciences Research Council (EPSRC) (Brumfiel 2012). The South Korean government is reported to have initiated a five-year program investing US\$20 million to develop graphene-based display panels and other devices. In August 2012, the Graphene Research Center of National Singapore University announced the opening of a micro- and nano-fabrication facility, with an investment of up to S\$15 million. The facility will focus on the development of new technologies and devices based on graphene.

If we classify papers in the main paths by the graphene preparation method adopted, then we find three main methods: rGO, exfoliated graphene, and CVD graphene. Table 4 summarizes the results of the classification, which shows there are seven papers on

exfoliated graphene, 18 papers on reduced graphene oxide, ten papers on CVD graphene, and two papers on others. Those covering rGO account for about 48 % as a whole and constitute the major part of the main paths. Nevertheless, as Fig. 2 illustrates, studies in this field focused on rGO earlier, but switched to CVD later on. Figure 3 shows a growing trend of CVD graphene research (Reina et al. 2009; Kim et al. 2009; Li et al. 2009a, b; Gunes et al. 2010; Wang et al., 2011, Bi et al. 2011a, b) which, along with reduced graphene oxide (Jung et al. 2008; Su et al. 2009; Qi et al. 2010; Yin et al. 2010; He et al. 2010; Wojcik and Kamat 2010; Kamat 2011; Hayashi et al. 2011), forms two important axes of development in this field. In the future, the possibility of more and more research studies on CVD graphene is not ruled out.

#### Citation count and main contributors

Main paths consist of most of the top cited papers in the field. As Table 1 shows, among the top 20 papers, 15 are on the main path. Narrowing down to the top ten papers, nine of them are on the main path. Aside from Stankovich et al. (2006), which has

Table 3 Papers on the main path:	S		
	Authors	Title	Journal
StankovichDDKZSPNR2006	Stankovich et al. (2006)	Graphene-based composite materials	Nature
StankovichDPKKJWNR2007	Stankovich et al.(2007)	Synthesis of graphene-based nanosheets via chemical reduction of exfoliated graphite oxide	Carbon
WatcharotoneDSPJDEWC LNR2007	Watcharotone et al. (2007)	Graphene-silica composite thin films as transparent conductors	Nano Letters
DikinSZPDENR2007	Dikin et al. (2007)	Preparation and characterization of graphene oxide paper	Nature
WangZMK2008	Wang et al. (2008)	Transparent, conductive graphene electrodes for dye- sensitized solar cells	Nano Letters
LiMGKW2008	Li et al. (2008a)	Processable aqueous dispersions of graphene nanosheets	Nature Nanotechnology
BecerrilMLSBC2008	Becerril et al. (2008)	Evaluation of solution-processed reduced graphene oxide films as transparent conductors	ACS Nano
EdaFC2008	Eda et al. (2008)	Large-area ultrathin films of reduced graphene oxide as a transparent and flexible electronic material	Nature Nanotechnology
BlakeBNBJSPMGHGN2008	Blake et al. (2008)	Graphene-based liquid crystal device	Nano Letters
LiZBSWWD2008	Li et al. (2008b)	Highly conducting graphene sheets and Langmuir–Blodgett films	Nature Nanotechnology
HernandezNLBSDMHBGB NDKGHSFC2008	Hernandez et al. (2008)	High-yield production of graphene by liquid-phase exfoliation of graphite	Nature Nanotechnology
JungDPR2008	Jung et al. (2008)	Tunable electrical conductivity of individual graphene oxide sheets reduced at "low" temperatures	Nano Letters
ReinaJHNSBDMK2009	Reina et al. (2009)	Large area, few-layer graphene films on arbitrary substrates by chemical vapor deposition	Nano Letters
KimZJLKKAKCH2009	Kim et al. (2009)	Large-scale pattern growth of graphene films for stretchable transparent electrodes	Nature
LiCAKNYPVJTBCR2009	Li et al. (2009b)	Large-area synthesis of high-quality and uniform graphene films on copper foils	Science
SuPALFM2009	Su et al. (2009)	Composites of graphene with large aromatic molecules	Advanced Materials
BaoZWNYSLT2009	Bao et al. (2009)	Atomic-layer graphene as a saturable absorber for ultrafast pulsed lasers	Advanced Functional Materials
ZhangTZBL2009	Zhang et al. (2009)	Large energy mode locking of an erbium-doped fiber laser with atomic layer graphene	Optics Express
LiZCBHCPCR2009	Li et al. (2009b)	Transfer of large-area graphene films for high-performance transparent conductive electrodes	Nano Letters
TanSKXLT2010	Tan et al. (2010)	Mode locking of ceramic Nd: yttrium aluminum garnet with graphene as a saturable absorber	Applied Physics Letters

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Table 3 continued			
	Authors	Title	Journal
QiPZLLBHZ2010	Qi et al. (2010)	Conjugated-polyelectrolyte-Functionalized reduced graphene oxide with excellent solubility and stability in polar solvents	Small
YinWZHZBZ2010	Yin et al. (2010)	Electrochemical deposition of zno nanorods on transparent reduced graphene oxide electrodes for hybrid solar cells	Small
ZhangTKZBL2010	Zhang et al. (2010)	Graphene mode locked, wavelength-tunable, dissipative soliton fiber laser	Applied Physics Letters
DeC2010	De and Coleman (2010)	Are there fundamental limitations on the sheet resistance and transmittance of thin graphene films?	ACS Nano
HeSYWLBHCZ2010	He et al. (2010)	Centimeter-long and large-scale micropatterns of reduced graphene oxide films: fabrication and sensing applications	ACS Nano
GunesSBHKCCL2010	Gunes et al. (2010)	Layer-by-layer doping of few-layer graphene film	ACS Nano
SunPHTWKTNF2010	Sun et al. (2010)	A stable, wideband tunable, near transform-limited, graphene-mode-locked, ultrafast laser	Nano Research
WojcikK2010	(Wojcik and Kamat 2010)	Reduced graphene oxide and porphyrin. an interactive affair in 2-d	ACS Nano
PopaSTHWF2010	Popa et al. (2010)	Sub 200 fs pulse generation from a graphene mode-locked fiber laser	Applied Physics Letters
Kamat 2011	Kamat (2011)	Graphene-based nanoassemblies for energy conversion	Journal of Physical Chemistry Letters
PopaSHTWF2011	Popa et al. (2011)	Graphene Q-switched, tunable fiber laser	Applied Physics Letters
WangTXOL2011	Wang et al. (2011)	Interface engineering of layer-by-layer stacked graphene anodes for high-performance organic solar cells	Advanced Materials
XuLWHHY2011	Xu et al. (2011)	Graphene saturable absorber mirror for ultra-fast-pulse solid-state laser	Optics Letters
HayashiLTTUK12011	Hayashi et al. (2011)	Electron transfer cascade by organic/inorganic ternary composites of porphyrin, zinc oxide nanoparticles, and reduced graphene oxide on a tin oxide electrode that exhibits efficient photocurrent generation	Journal of the American Chemical Society
BiHLXJ2011	Bi et al. (2011a)	Transparent conductive graphene films synthesized by ambient pressure chemical vapor deposition used as the front electrode of CdTe solar cells	Advanced Materials
BiHLTLXJ2011	Bi et al. (2011b)	Large-scale preparation of highly conductive three dimensional graphene and its applications in CdTe solar cells	Journal of Materials Chemistry
LiuWQZSX2012	Liu et al. (2012)	Graphene oxide absorber for 2 mu m passive mode-locking Tm:YAIO3 laser	Laser Physics Letters

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## Table 4 Categories by different preparation methods

Preparation methods	Corresponding author	Year published
Reduced graphene oxide		
StankovichDDKZSPNR2006	Ruoff, RS	Stankovich et al. (2006)
StankovichDPKKJWNR2007	Ruoff, RS	Stankovich et al. (2007)
WatcharotoneDSPJDEWCLNR2007	Ruoff, RS	Watcharotone et al. (2007)
DikinSZPDENR2007	Ruoff, RS	Dikin et al. (2007)
WangZMK2008 (+device fabrication)	Mullen, K	Wang et al. (2008)
LiMGKW2008	Wallace, GG	Li et al. (2008a)
BecerrilMLSBC2008	Bao, Z; Chen, Y	Becerril et al. (2008)
EdaFC2008	Chhowalla, M	Eda et al. (2008)
JungDPR2008	Ruoff, RS	Jung et al. (2008)
SuPALFM2009	Feng, XL	Su et al. (2009)
TanSKXLT2010 (+device fabrication)	Tang, DY	Tan et al. (2010)
QiPZLLBHZ2010	Zhang, H	Qi et al. (2010)
YinWZHZBZ2010 (+device fabrication)	Zhang, H	Yin et al. (2010)
HeSYWLBHCZ2010 (+device fabrication)	Zhang, H	He et al. (2010)
WojcikK2010	Kamat, PV	Wojcik and Kamat (2010)
Kamat2011	Kamat, PV	Kamat (2011)
HayashiLTTUKI2011	Kamat, PV; Imahori, H	Hayashi et al. (2011)
LiuWQZSX2012 (+device fabrication)	Liu, J	Liu et al. (2012)
Exfoliated graphene		
BlakeBNBJSPMGHGN2008 (+device fabrication)	Novoselov, KS	Blake et al. (2008)
LiZBSWWD2008	Dai, HJ	Li et al. (2008b)
HernandezNLBSDMHBGBNDKGHSFC2008	Coleman, JN	Hernandez et al. (2008)
SunPHTWKTNF2010 (+device fabrication)	Ferrari, AC	Sun et al. (2010)
PopaSTHWF2010 (+device fabrication)	Ferrari, AC	Popa et al. (2010)
PopaSHTWF2011 (+device fabrication)	Ferrari, AC	Popa et al. (2011)
XuLWHHY2011 (+device fabrication)	Hao, XP	Xu et al. (2011)
CVD graphene		
ReinaJHNSBDMK2009	Kong, J	Reina et al. (2009)
KimZJLKKAKCH2009	Hong, BH	Kim et al. (2009)
LiCAKNYPVJTBCR2009	Ruoff, RS	Li et al. (2009a)
BaoZWNYSLT2009 (+device fabrication)	Loh, KP	Bao et al. (2009)
	Tang, DY	
ZhangTZBL2009 (+device fabrication)	Tang, DY	Zhang et al. (2009)
LiZCBHCPCR2009	Ruoff, RS	Li et al. (2009b)
GunesSBHKCCL2010	Choi, JY	Gunes et al. (2010)
WangTXOL2011 (+device fabrication)	Loh, KP	Wang et al. (2011)
BiHLXJ2011 (+device fabrication)	Huang, FQ	Bi et al. (2011a)
BiHLTLXJ2011 (+device fabrication)	Huang, FQ	Bi et al. (2011b)
Others	-	
ZhangTKZBL2010 (+device fabrication)	Tang, DY	Zhang et al. (2010)
DeC2010	Coleman, JN	De and Coleman (2010)

 $+ device \ fabrication, \ indicates \ that \ a \ device \ fabrication \ study \ is \ included$ 

a total of 1788 citations, other important papers with high citation counts include: Stankovich et al. (2007), Kim et al. (2009), (Li et al. 2008a), (Li et al. 2009a), Wang et al. (2008), Reina et al. (2009), Eda et al. (2008), and Dikin et al. (2007). This means that the papers on the main path are generally consistent with those highly cited papers. We do note that, other than the number of total citations, Table 1 also includes the number of citations from the papers in this study's dataset. This number (N) is smaller than the number of total citations because not all citations are from the papers within the dataset. To estimate the effect of self-citation for the papers on the main paths, we count the number of self-citations for each paper as n. We note a selfcitation when at least one author appears on the author list of both the citing paper and cited paper. The ratio of n/N indicates the effect of self-citation on the citation number. The ratios are low, and their effects on the main paths are expected to be insignificant.

From the Corresponding Author column in Table 4, one observes several research teams that have made significant contributions to the field, like Ruoff (seven papers), Tang (four papers), Zhang (three papers), Ferrari (three papers), Kamat (three papers), Loh (two papers), Huang (two papers), and Coleman (two papers). The groups of scholars like Ruoff focused more on graphene preparation and characterization; Zhang, Loh, and Huang on applications; while Tang and Ferrari focused on laser applications research.

### Device applications

The papers related to graphene applications on the main paths are mainly for the production of transparent films or transparent electrodes. These accessories have been used in optoelectronics devices including: solar cells (Wang et al. 2008; Yin et al. 2010; Wang et al. 2011; Bi et al. 2011a, b), LCD (Blake et al. 2008), sensing devices (He et al. 2010), laser mode locking devices (Bao et al. 2009; Zhang et al. 2009; Tan et al. 2010; Zhang et al. 2010; Sun et al. 2010; Popa et al. 2010, 2011; Xu et al. 2011; Liu et al. 2012), and so on. The above mentioned data also show that solar cells and laser mode locking devices encompass the items that most researchers choose to study in the GFO field up until today.

## Conclusions

In this study, we present the graphene for optoelectronics development scenario from a perspective different from that of previous studies. For the first time, we identify the main development paths of GFO. The strong growth of the GFO literature has been on such a scale that it is necessary to adopt a quantitative statistical method to conduct a general review and to find out the main stream of development. The main path method based on citation helps us identify the significant development paths and important papers in this field. It provides a comprehensive view of the whole science development in which the Ruoff group is found to be the earliest pioneer, while teams led by Tang, Zhang, Ferrari, Kamat, Loh, Huang, and Coleman have made considerable contributions, respectively. Most notably, the main path graph shows undergoing knowledge diffusion of the key sub-area, such as rGO, CVD, and exfoliation techniques has been developed for the preparation and applications of GFO, covering solar cells, laser devices, sensing devices, and LCD.

There are several limitations to this study. For example, the dataset is taken from the WOS database. Although it is the largest citation-based academic database available, there are, however, some GFO papers published in journals not included in the WOS. Presentation and interpretation of the results should be accompanied by a warning on the limitation of the data source.

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