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Graphene and its Phononics: A Review

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Abstract

Graphene, a unique allotrope of carbon, has garnered a huge amount of attention amongst researchers as its unique properties and promising applications in various real-life domains like in efficient batteries, solar cells, medicinal technologies, environmental remediation, circuit boards, lighting and display, and anti-corrosion has resulted in its commercial exploitation and implementation in everyday life. Since 2004 graphene has been one of the most beautiful scientific and technological achievements. The unique electronic cloud forming the bond between different carbon atoms in graphene leads to several inquisitive questions raised in the field of quantum physics. Till now graphene has been exploited for its electronic and optical properties but new research has shown that phononic properties of graphene will lead to the development of "killer" practical applications in the future. In this review article we have explored the structure, properties and the phononics of graphene with a special reference to few-layer graphene (FLG) and graphene Thermal Interface Materials (TIMs).

Key words : Half-integer Quantum Hall Effect (QHE), phononics, Raman Optothermal Spectroscopy, Few-layer Graphene (FLG), Thermal Interface Materials (TIMs).

1. Introduction

Material Sciences research has greatly focused upon the mass production, characterization and applications in the real world of ultra-thin carbon films, of which graphene is the thinnest. Graphene has attracted a lot of attention of the scientific community due to its unique properties and wide-ranging promising applications. Its various real-life applications include its use in efficient batteries, solar cells, medicinal technologies, environmental remediation, circuit boards, lighting and display, and anti-corrosion etc [1]. The possibility of widely ranged applications has given a way for its commercial exploitation and implementation in everyday life. Since 2004, when graphene was first isolated, it has been considered the best scientific and technological accomplishment. Graphene has also found well tested and established applications in thin-film transistors, transparent conductive films (TCF), ultra-sensitive chemical sensors and quantum dot (QD) devices. Single layer graphite which is hexagonal in shape and is crystalline is the simplest and most basic crystalline allotrope of carbon with carbon atoms having a bond distance of 0.142 nm. The broad category of application areas includes sensors, microelectronics, composite materials and biomedical applications[2].

2. Structure of Graphene

Graphene is a unique allotrope of carbon in which each carbon atom is bonded to its neighbors tightly by a unique electronic cloud that leads to several inquisitive questions raised in the field of quantum physics[3]. It gives validity to the quantum hall phenomenon which is also quite unique. Various forms in which graphene exists include graphene nanoribbons, nanosheets, nanoplates and 3D graphene. Each of them displays promising and overwhelming applications. The electronic and quantum properties of graphene are still under an inquisitive lens of fundamental research. Every carbon atom in graphene is in a state of sp^2-hybridization, having three bonds, closely in touch with different neighbouring carbon atoms[4,5].

The sp^2-hybridization in graphene arises due to the combination of s, p_x, and p_y orbitals. In the hexagonal phase, three distinct carbon atoms attach themselves in a covalent bond with each carbon atom in the vicinity and all of them are specifically sp^2 hybridized, resulting for each carbon atom in one free electron. The p_z orbital supports this free electron and this p-orbital is located above the plane, forming the pi bond[6]. Strikingly, the p_z orbital of graphene plays a very important role in the chemical and physical dynamics of this unique material. The presence of a zero bandgap is both a shortcoming and a unique feature of graphene, which leads to several new opportunities to develop artificial human made materials with tunable band gaps that can be utilized for the next-generation of computing. The two pi-electrons that are located in every hexagon of the graphene sheets are the reason for the exceptional conductivity of graphene[7]. Also, due to the closed packing of atoms in the crystal lattice of graphene, it is very stable, but only if its size is of the order of 20 nm or less, otherwise it is very unstable thermodynamically, the exception being some specific conditions[8].

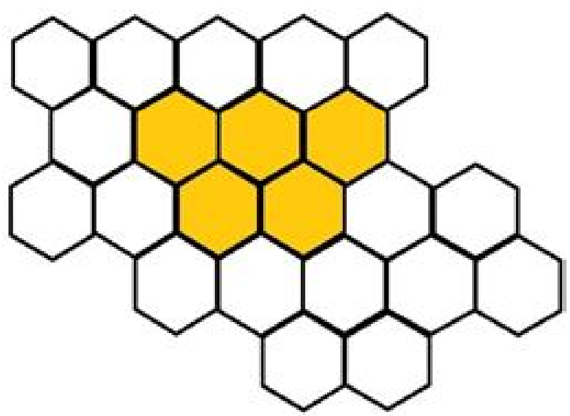

Fig. 1: Graphene as an allotrope of carbon having a single atomic layer in a two-dimensional lattice resembling a honeycomb.

3. Properties of Graphene

Graphene is a two-dimensional sheet-like material with several remarkable properties. It is quite interesting that the structural holes allow the phonons, the quanta of lattice vibrations, to go unobstructed, which leads to remarkable thermal conductivity in graphene[9,10]. However, this property has been unobserved in graphene oxide and other derivatives of graphene owing to the manipulated band structure. The classification of graphene as metal, non-metal or semimetal is still debatable and requires further probing. But, due to graphene's metallic layers with very low bandgaps, it can be considered as a semimetal with an exceptional theoretical background. On the whole, it has lots of remarkable characteristics that are not noticeable for other non-metallic materials as well as for the existing ideal semimetals. The properties of graphene completely depend on the number of layers and the defects present in the graphene layers[11-13]. For example, the surface area of pristine graphene is ~2630 m^2 /g (theoretically) which is quite higher than the surface area of carbon black (850 to 900 m^2 /g), CNTs (100 to 1000 m^2 /g), and many other carbon-based analogues. Whereas, the surface area of few-layer graphene (FLG), graphene oxide, and various derivatives are much less in comparison to single-layer graphene (SLG). Due to these extraordinary properties, graphene behaves as the most suitable material for many modern technologies such as electronic applications besides acting as a substrate or template for other materials[14]. One of the most prominent properties of graphene is its brilliant electrical conductivity, which is unparalleled and highly essential for next-generation technologies. The zero bandgap of graphene and its engineered analogue with little overlap between valence and conduction band opened great potential for applied and fundamental research. It is due to electrons mostly, which behave as massless relativistic particles.

Recently, lots of scientists and researchers are working globally in the same area for the development and production of next generation 2D materials with unique properties and promising applications. Many researchers documented that graphene could display several charge transporters and carriers up to 10^{13} cm^{-2} having a mobility of 1×10^4 cm^2 V^{-1} s^{-1} at room temperature which could be tuned according to real-time applications[15]. It has also been discovered that this mobility can rise up to 2×10^5 cm^2 V^{-1} s^{-1} at low temperature under specific boundary conditions. Since the charge carriers in graphene behave as in semi metals, commonly termed as massless Dirac Fermions, graphene displays a half-integer Quantum Hall Effect (QHE)[16]. In this way, the Quantum Hall Effect in graphene is unique and showcases an exceptional relation between charge, thickness, and speed of the charge carriers. Due to these properties of graphene, it has been discovered that the electrical resistance of a graphene sheet is much less than that of silver, which provides for its high favorability for electronic applications[17]. Such prototype devices and electronic gadgets are in the development stages right now and are expected to enter the market within the next 5 years. It has also been observed that when the magnetic field starts acting perpendicular to the conducting materials, especially to the 2D system's plane and along the axis, then the QHE is observed which strongly supports the exceptional Hall Effect in the case of graphene. The electron repression results in discrete band levels due to the two-dimensional nature of graphene. Such levels, which are occupied by charge carriers, are known as Landau levels. But, deeper research must be carried out for graphene and their analogous materials to completely explore their potential.

Graphene has exceptional optical, thermal, and mechanical properties[18]. It has been discovered that up to 2.3% of white light is absorbed by each layer of graphene with a reflectance rate of less than 0.1% [19]. Thus, the pure single graphene layer is high in transparency along with a high degree of flexibility. The relationship between the absorbance and the number of layers of graphene is linear in nature; as a consequence, as the number of layers increases in graphene, the absorbance increases rapidly. Theoretically, at room temperature, single layer graphene can display a thermal conductivity of 3000-5000 $Wm^{-1}K^{-1}$ [20]. Scientists are still searching for the roots of such an exceptionally high thermal conductivity in graphene. Also, depending on the nature of the substrate, this thermal conductivity can be reduced to 600 $W\,m^{-1}K^{-1}$ even for single-layer graphene. Further, several studies have established that the mixture of ^{12}C and ^{13}C has a quite high value of thermal conductivity of graphene. Such a kind of dissimilar variation of thermal conductivity is because of the dissemination of phonons at the interface of the graphene gallery which blocks their movement. In single layer graphene the pathway of phonons differs from that in other forms. Indeed, graphene performs much better as compared to copper (Cu) even at this lower conductivity. Hence, graphene is the strongest and best conductive material. Particularly, a single layer of graphene can tolerate up to 42 Nm^{-1} of stresses, having Young's modulus of 1.0 TPa[21].

Table 1. Various Properties and Characteristics of Graphene[22]

CHARACTERISTIC	VALUE
Current density	$\sim 10^9$ A cm^{-1}
Electron mobility	$\sim 2 \times 10^5$ cm^2 V^{-1} s^{-1}
Thermal conductivity	~ 5000 W m^{-1} K^{-1}
Transparency	~97.7%
Surface area	~ 2360 m^2 g^{-1}
Elastic limit	~20%
Breaking strength	42 N m^{-1}
Velocity of fermion (electron)	$\sim 10^6$ m s^{-1}
Tensile strength	~1.5 Tpa

4. Phononics: Phonon Engineering

Crystalline solids comprise closely packed atoms placed together in a three-dimensional pattern which repeats itself throughout the crystal known as lattice. The atoms are closely linked with their respective neighbouring atoms[23,24]. They are linked via bonds, be it ionic or covalent in nature. Each atom can vibrate about its mean position in a simple harmonic motion like a tiny spring due to its own thermal energy or outside forces. Hence collectively, the lattice also vibrates as it is made up of these single vibrating atoms grouped together[25]. This results in the generation of mechanical waves in the crystal that carry heat and sound in the form of waves throughout the material. A packet of these waves can propagate through the material carrying a definite momentum and energy. In quantum mechanical terms this packet of waves can be treated as a particle. This particle is known as phonon which is a unit of vibrational energy arising from the oscillating atoms within the crystal lattice. Being a discrete and quantum unit of vibrational mechanical energy, it is analogous to a photon which is a quantum particle of light or electromagnetic energy[26]. Hence the study of phonons and how their characteristics affect and influence the elastic, acoustic and thermal properties of bulk and nanostructured materials is known as phononics[27]. Also known as phonon engineering, it deals with the manipulation and tweaking of elastic, acoustic and thermal phonons in semiconductors to obtain favourable acoustic, elastic and thermal properties in bulk and specifically in nanostructured materials. By engineering of phonons, we can discover a whole new spectrum of functionalities for materials. Electrical properties of a material are a manifestation of the electronic nature and configuration of that material[28]. Likewise, phonons determine the speed of sound in a material or how much heat it will take to change the temperature of the material.

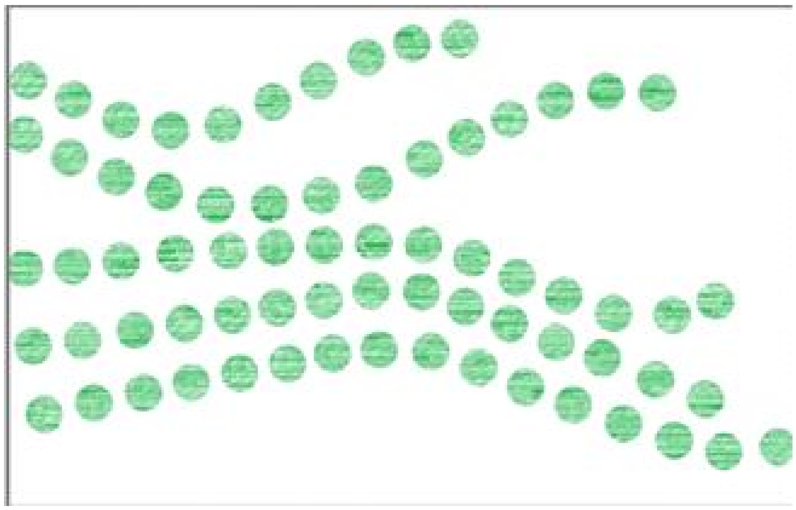

Fig. 2. Phonons propagating through a crystal lattice

5. Phononics of Graphene

Phononics of lower dimensional materials is of quite an interest for scientists and researchers as tweaking of properties at the nanoscale results in exceptional enhancement in thermal, optical and electronic properties of the nanomaterial[29,30]. Graphene is such a low dimensionality system of great interest and focus. A very high interest in graphene started due to its unique electronic dispersion which is linear in nature[31]. It was revealed by a quite high charge carrier mobility and other exotic optical and electronic properties. Apart from electrons, phonons also become highly sensitive as the material heads from the bulk state towards the single atomic plane nanoscale limit[32]. This is quite prominent in optical and acoustic phonons. The most standard technique to identify graphene and count the number of atomic planes in few-layer graphene (FLG) is Raman Spectroscopy, especially in the case of optical phonons[33]. Raman spectral signatures of these phonons are obtained including G peak and 2D band. In the field of graphene research, Raman Spectroscopy became quite instrumental for the proliferation of graphene. Instead of conducting low temperature phonon transport measurements in single-layer graphene, taking a Raman Spectrum became way easier. Thermal conductivity of graphene was first experimentally done with the help of a no contact Raman optothermal method[34]. In FLG it has been observed that acoustic phonons are sensitive to the number of atomic planes changing their ability to conduct heat. Remarkable thermal conductivity characteristics such as high values and dependence on the lateral size of the sample of intrinsic thermal conductivity is caused due to the absence of interatomic plane coupling and a modified phonon density of states (DOS) in graphene. Due to the emergence of FLG, phonon dispersion can be engineered in the whole Brillouin Zone (BZ) and in the energy range from optical to acoustic phonons. This is possible just by a simple twist of one atomic plane about FLG's surface normal vector with respect to another[35]. The discovery of unique phonon transport properties of graphene has led to numerous studies concerning the practical use and application of graphene and FLG in thermal composites and coatings which is discussed ahead. Another possible explanation for graphene's and FLG's excellent thermal conductivity arises from the concept of phonon hydrodynamic transport. As for the hydrodynamic collective phonon transportation, it results in high thermal conductivity of graphene that is above 3000 W/mK at room temperature.

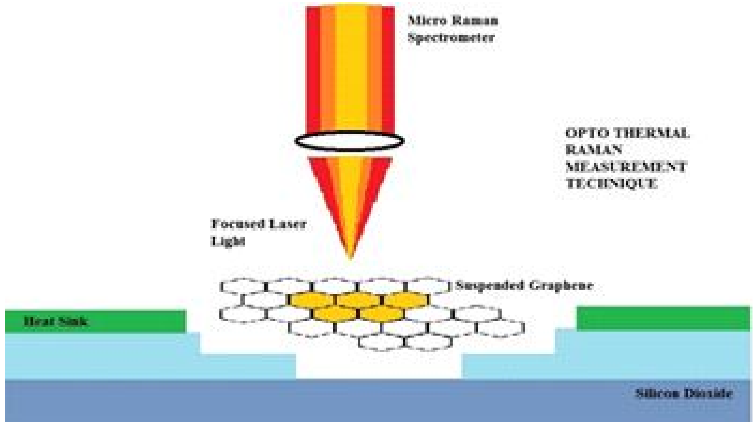

Fig. 3: Schematic Representation of the thermal conductivity measurement of graphene using optothermal Raman Spectroscopy

6. Few-Layer Graphene (FLG) in Thermal Interface Materials (TIMs)

Few Layers Graphene (FLG) is the most promising material when it comes to its practical applications in TIMs[36,37]. The promising applications of FLG in TIMs are inspired by its quality-dependent high levels of thermal conductivity in the range of 500-2000 W/mK which has lesser rate of degradation compared to graphene when exposed to matrix materials in composites; a comparatively large cross-sectional area to conduct heat with respect to graphene; ability to retain its mechanical flexibility required to thermally couple with the matrix material; and low-cost production with the flexibility of good control on its thickness[38,39].

6.1. Preparation of graphene non-curing thermal paste for TIM applications:

There are various practical applications of non-curing thermal interface materials in electronics. To start with firstly we have to develop a non-curing thermal paste using graphene [40]. In this method, graphene is added to the base material with acetone followed by slow-speed sheer mixing. The optimized mixing process separates the graphene and mineral oil mix from the acetone, leaving a smooth graphene paste with proper viscosity that is easy to store and to apply at the interfaces. The synthesis process is scalable and already used by industry.

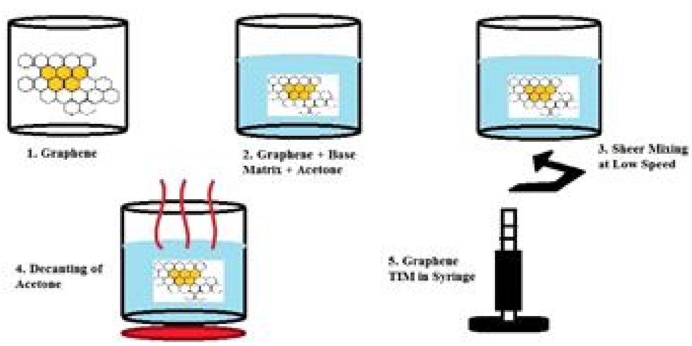

Fig. 4: The process for synthesizing graphene non curing thermal paste for Thermal Interface Materials (TIMs)

7. Conclusions

There seems to be a rapid interest in the phononics of graphene and its scientific research as well as practical applications. The interest originated in the optical and electronic properties of graphene but as we move forward, we can see that phonons showcase sensitivity towards the number of atomic planes and unusual heat conduction properties originating from the two-dimensional nature of graphene plates. In the future we will see exciting new developments in the field of phononics of graphene relevant to phonon scattering, dispersion of acoustic phonons in graphene and further investigation of hydrodynamics phonon transport in graphene, FLGs and thin films. There is a rapid growth in the practical applications of graphene related to its phononic properties. The "killer" applications being awaited to be discovered by researchers owing to graphene's electronic properties have already started to get replaced by the non-electronic i.e., thermal, mechanical, surface and electrochemical properties of graphene in the market. These applications are governed by graphene's phonon transport and heat conduction properties. Here graphene means single-layer graphene and FLG as either layers on a substrate or as a mixture in a composite. Few examples include graphene-enhanced cooling systems for computer processors, epoxy adhesives, coatings, and non-curing TIMs for a wide variety of electronic uses. All of these developments lead us to a realization that it is possible for the true "killer" application, or rather a range of applications, coming from graphene's phononic properties rather than its electronic properties.

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