Journal of Advances and Scholarly Researches in Allied Education
Vol. 21, Issue No. 5, July-2024, ISSN 2230-7540

DOI: https://doi.org/10.29070/yrgpqt98

Optimization of Synthesis Parameters for Mgb2/Graphene Bilayer Films: Enhancing Superconducting Transition Temperature

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Abstract: Over the last few years, there has been a boom in interest in the development of hybrid structures that combine graphene with superconductors. This might be attributed to the newly found superconducting proximity effect as well as the need for superconducting devices that are at the cutting edge of technology. Increasing the superconducting transition temperature (Tc) is the major objective of this research project, which aims to optimise the synthesis parameters for bilayer films composed of graphene and magnesium boride (MgB2). The first step in the procedure involves transferring monolayer graphene that has been generated on copper foil to the substrate that is being targeted. The subsequent step is to produce thin films of magnesium bicarbonate by using a hybrid technique that combines physical and chemical vapour deposition. Both of these steps are essentially important to the manufacturing process as a whole. By fine-tuning parameters such as precursor flow rates, annealing conditions, and deposition temperature, the MgB2 film is optimised for a dominant c-axis orientation, a smooth and continuous surface, and a sharp superconducting transition at an improved Tc of 36 K, which is close to the bulk value of 39 K. This is accomplished by optimising the film for these characteristics. This increase in crystallinity and Tc, which illustrates the usefulness of the improved production strategy, makes it feasible to undertake more research into the proximity effect in graphene and to create high-temperature superconducting devices such as Josephson junctions. Both of these developments show that the optimised manufacturing approach is effective. The creation of these innovations is made possible by advancements in the manufacturing process.

Keywords: Parameters , superconducting, Transition, Temperature

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INTRODUCTION

Superconducting materials that have been merged with graphene have cleared the way for exciting new breakthroughs in materials science and condensed matter physics. These developments have provided a firm platform for future research, both theoretical and practical. Magnesium diboride (MgB2) is a superconductor that is especially noteworthy due to its high critical current density (Jc) and low anisotropy. Additionally, it has a very high superconducting transition temperature (Tc) of 39 K, which makes it one of the most remarkable superconductors. Graphene is a two-dimensional material that has extraordinary mechanical, electrical, and thermal properties. Some of these properties include great carrier mobility and flexibility. There is just one layer of carbon atoms that are arranged in a hexagonal lattice to make up this structure. In the process of researching novel quantum phenomena that are made feasible by the combination of graphene and magnesium boron tetrachloride, it is likely that innovative superconducting devices will be produced. The phenomenon known as the superconducting proximity effect is one example of this.

There is a potential of exploring the superconducting proximity effect, which would include producing the superconducting features of MgB2 in graphene. This is an exciting component of the hybrid structure that consists of MgB2 and graphene. The use of this characteristic is essential for the development of nanoscale quantum devices, such as Josephson junctions and superconducting quantum interference devices (SQUIDs), which are capable of operating at temperatures that are greater than those of conventional superconductors. The integration of graphene with magnesium boron tetragonal is expected to result in enhanced device performance due to the combination of graphene's remarkable transport properties and the strong superconducting properties of magnesium boron tetragonal.

In spite of the fact that they have a lot of promise, superconducting transition temperature (Tc) maximisation of MgB2 and graphene bilayer films is still a difficult challenge to accomplish. The production of these hybrid structures is a multi-step process that needs the careful deposition of MgB2 thin films onto the surface of the graphene and the effective transfer of monolayer graphene onto the target substrate. Both of these steps are necessary for the manufacture of these hybrid structures. There are a variety of synthetic parameters that impact the quality, orientation, and superconducting features of the MgB2 film. These factors include the deposition temperature, the precursor flow rates, the substrate selection, and the post-deposition annealing.

Ideal synthesis parameters for the production of bilayer films consisting of graphene and magnesium

The major purpose of this study was to determine the ideal synthesis parameters for the production of bilayer films consisting of graphene and magnesium boron tetrachloride in order to increase the temperature at which they made their superconducting transition. Through the use of the high-performance chemical vapour deposition (HPCVD) method, thin films of magnesium boron tetrachloride (MgB2O5) are produced on the graphene layer. While doing so, the parameters that are of utmost importance are fine-tuned. When it comes to enhancing the stoichiometry and superconductivity of the film, the annealing conditions are quite important. Additionally, the deposition temperature has an effect on the crystalline structure and c-axis orientation of the magnesium magnesium boronide film.

Providing that these synthesis parameters are adjusted, it is anticipated that high-quality MgB2 and graphene bilayer films would be formed. These films should exhibit improved crystalline properties and a higher Tc. These advances will not only assist us in gaining a better understanding of the superconducting proximity effect in graphene, but they will also pave the way for the creation of next-generation superconducting devices that are capable of withstanding higher extremes of temperature. This work motivates both academic research and practical applications in fields like as energy-efficient technologies, superconducting electronics, and quantum computing. These are all topics that are currently being researched.

OBJECTIVES

1. To create high-quality MgB_2/graphene bilayer films by optimising synthesis parameters as annealing conditions, precursor flow rates, and deposition temperature.

2. To improve MgB_2 films' crystallinity and superconducting transition temperature (Tc), which will enable the creation of sophisticated superconducting devices that can function at greater temperatures.

METHODOLOGY

An illustration of the method of covering graphene with a thin layer of magnesium boride (MgB2) may be seen in Figure 1. Graphene is first produced by chemical vapour deposition (CVD) on copper foil as the starting material. A massive expanse of monolayer graphene is produced as a consequence of this process. This procedure consists of two significant phases, which are as follows: The first thing that has to be done is to transfer graphene between copper foil and the substrate of your choosing. The transfer is followed by the application of a thin layer of magnesium boride (MgB2) to the graphene, which is then thoroughly coated. In comparison to past investigations, the process for transferring graphene has not undergone any modifications. The whole procedure is shown in Figure 1, which you may see.

Figure 1. Process for graphene mgb2 thin film manufacturing.

To begin, a layer of polyvinyl butyral (PVB) with a thickness of 500 nanometres and a mass concentration of 5% was spin-coated onto the monolayer graphene that was placed on copper foil (XF023, XFNANO Materials Co.). After that, it took around half an hour for the coating to turn into a solid state at ambient temperature. Following the aforementioned step, an aqueous solution of iron nitrate with a concentration of 1 mol/L was used to progressively remove the 20 µm thick copper foil over a period of several hours. After the PVB/graphene layer that was created was washed with deionised water, it was then deposited onto the substrate that was needed, which in this case was a c-cut 6H-SiC substrate measuring 5 mm by 5 mm. For the purpose of preparing the graphene for the subsequent stage, which included depositing a thin coating of magnesium boride (MgB2), the PVB was heated to 60 degrees Celsius for around ten minutes in order to soften it. A solution of alcohol was then used to dissolve the PVB.

Previous studies provided specific information on the use of the hybrid physical-chemical vapour deposition (HPCVD) technique for the production of the MgB2 thin film. Using this technique, four magnesium ingots were positioned around the graphene/SiC substrate inside of a container that was maintained at a high pressure (about 5 kPa) and supplied with a continuous flow of pure hydrogen gas (300 sccm). Around 680 degrees Celsius was the temperature at which the magnesium ingots melted, which resulted in the release of a cloud of high-pressure magnesium vapour over the surface. It was concurrently cycled through the chamber at a rate of 6 sccm that B2H6 gas, which contained 5% hydrogen, was being pumped through. It was the thermal disintegration of the boron in this gas, which was a pure source of boron, that interacted with the magnesium vapour to start the process of depositing the MgB2 layer. With a deposition time of two minutes, a layer thickness of about thirty nanometres of magnesium bicarbonate was accomplished.

In order to gain a more accurate evaluation of the film deposition on graphene, it is possible to compare the findings with those of MgB2 films that were generated on SiC substrates that were devoid of graphene under the identical conditions. A variety of experimental techniques, including as electrical resistance tests, scanning electron microscopy (SEM), Raman spectroscopy, and X-ray diffraction (XRD), were used in order to investigate the samples that were acquired. The acquisition of Raman spectra was carried out at room temperature with the assistance of a Renishaw inVia Raman spectrometer equipped with a 532 nm laser. For the purpose of recording the XRD patterns, the Rigaku D/max-RA X-ray diffractometer was carried out. The range of its scanning was from 20° to 80° (2θ), and the rate of scanning was 4° per minute. Cu Kα was the radiation that was used. An FEI Quanta 200 microscope was used to acquire a scanning electron micrograph of the substance so that the form of the material could be analysed. A Keithley 6221 current source, a Lakeshore-calibrated DT670 silicon diode thermometer, and a 2182A nanovoltmeter were the components of the custom-built apparatus that was used for the purpose of measuring electrical resistance across a temperature range that extended from 5 to 300 Kelvin.

RESULT

The several stages of the manufacturing process that were responsible for the acquisition of Raman spectra are shown in Figure 2. The Raman spectra of the sample were obtained prior to the deposition of the MgB2 thin film in order to verify that the graphene had been effectively transferred onto the SiC substrate. In Figure 2(d), the findings are shown at a higher laser intensity, while in Figure 2(a), the results are displayed in the spectral area of 1400-3200 cm^{-1}. Figure 2(a) displays spectra that include several peaks ranging from 100 to 2000 cm^{-1}. Within these spectra, the two peaks that are most prominent are located at 788 and 968 cm^{-1}, respectively. The fact that these peaks are in excellent accord with previous experimental research on the Raman spectra of SiC and that they are extremely comparable to those reported for 6H-SiC is a strong indication that they are originated from the SiC substrate. In the sequence shown above, the two most significant peaks are the ones that correspond to the E2 and A1 phonon modes. In addition, there is a peak in the frequency range that spans from 2000 to 3200 cm^{-1}, with the centre of this peak being 2692 cm^{-1}. This peak is the characteristic two-dimensional peak that is associated with graphene. The graphene was successfully deposited onto the SiC substrate, as shown by the spectra shown in Figures 2(a) and 2(d). It is possible that the peak at around 1580 cm^{-1}, which is another distinctive graphene peak, was concealed by peaks emerging from the SiC substrate at the same locations. This can be seen in the magnified picture shown in Figure 2(d).

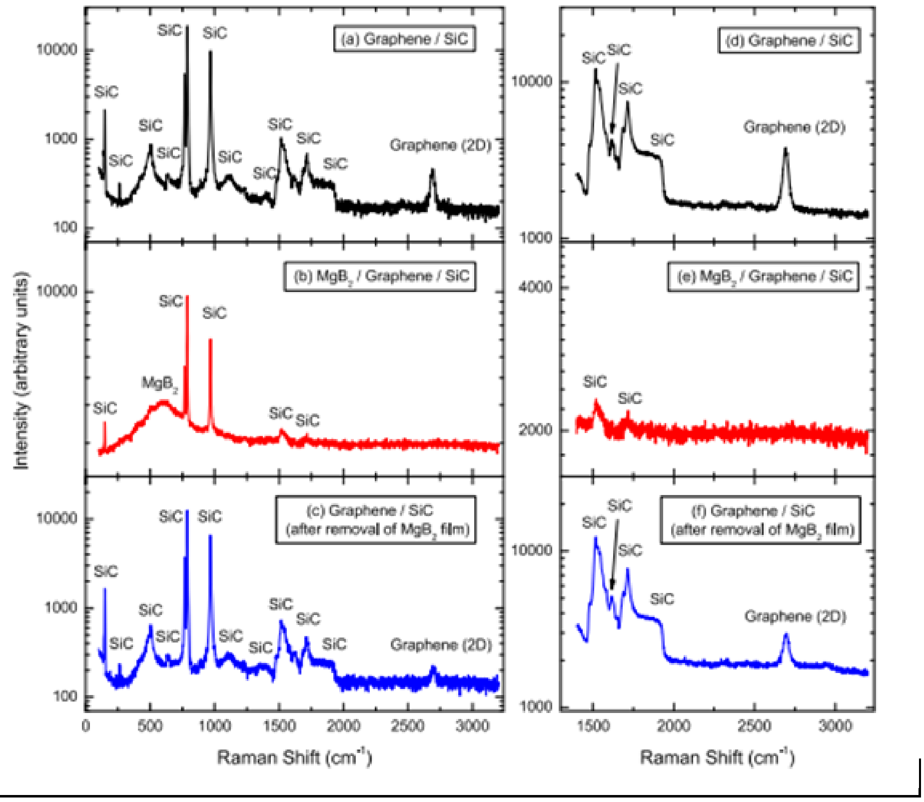

Figure 2. Raman spectra during graphene MgB2 thin film fabrication. (a) Graphene/SiC: after transfer to SiC substrate and before MgB2 layer development. B) MgB2/Graphene/SiC after thin film deposition. After MgB2 thin layer removal, SiC/graphene. Spectra at 1400-3200 cm−1 were collected concurrently with increasing laser intensity (a, b, c, d, e, f).

It is possible that the spectra were occluded by peaks at 1580 cm^{-1} that originated from the SiC substrate, as can be observed in the enlarged version of Figure 2(d). Following the deposition of a thin layer of magnesium bicarbonate (MgB2) onto the graphene/silicon carbide (SiC) substrate, Figure 2(b) displays a broad peak at around 600 cm^{-1}. As a consequence of the E2g phonon mode, this peak is typical of magnesium bicarbonate, according to the findings of earlier studies. As can be seen in Figures 2(b) and 2(e), the MgB2 layer is able to hide the 2D graphene peak as well as a number of SiC peaks. As can be seen in Figures 2(c) and 2(f), the Raman spectra were re-collected for verification after the thin coating of MgB2 was removed by the use of conventional acid etching. In accordance with the prediction, the huge peak at around 600 cm^{-1} disappears, and the weak SiC peaks return. As shown by the fact that the 2D graphene peak was effectively restored, graphene was successfully covered by the thin layer of magnesium bicarbonate. Additionally, this demonstrates that the graphene continue to maintain a high level of contact with the SiC base even after the transfer has taken place.

These XRD patterns of MgB2 thin films on silica/graphene, graphene, and silica substrates are compared in Figure 3, which illustrates the results of the comparison. The confirmation of the c-cut orientation of the SiC substrate is based on the fact that during the 2θ scan, only the peaks (006) and (0012) are visible. When MgB2 is directly deposited on SiC, the c-axis of the film is orientated in a direction that is perpendicular to the substrate. This is due to the fact that the only peaks that are present are the MgB2 (001) and (002) peaks, in addition to the peaks obtained from the substrate. Considering the close lattice match between hexagonal 6H-SiC (a = 3.081 Å) and MgB2 (a = 3.086 Å), it can be concluded that the epitaxial growth of MgB2 on SiC is in accordance with the observations that have been made in the past. The c-axis lattice constant of the MgB2 film is somewhat less than the bulk value of 3.524 Å, which validates prior results of in-plane tensile strain in MgB2 films produced on SiC substrates using HPCVD. This is based on peak locations, which indicate that the lattice constant is slightly less than the bulk value.

As can be seen in Figure 3, the development pattern of the MgB2 thin film on the graphene/SiC substrate is similar to that of the film created directly on SiC. This is because the c-axis is perpendicular to the surface of the film. There is a substantial difference in the lattice mismatch between hexagonal graphene and MgB2 (a = 2.460 Å) compared to the match between 6H-SiC and MgB2. This difference indicates that compressive strain is produced in the film. It is important to note that the MgB2 peaks on the graphene/SiC substrate, which are remarkably comparable to those on the film formed directly on SiC, serve as an indication of the in-plane tensile strain.

Disagreements between the substrate and the film lattice, differences in the growth mode of the film, and differences in the thermal expansion coefficient have all been identified as possible causes of strain in MgB2 films. There is a need for more research in order to get a reliable measurement of strain in MgB2 layers on graphene. When compared to the other two SiC substrates, Figure 3 demonstrates that the MgB2 layer on the graphene/SiC substrate exhibits two peaks that are close to SiC (006) and one peak that is near SiC (0012), both of which are at higher 2Θ values. It is possible that these variations are the result of differences in the lattice properties of the n-type nitrogen-doped 6H-SiC substrates from one batch to the next.

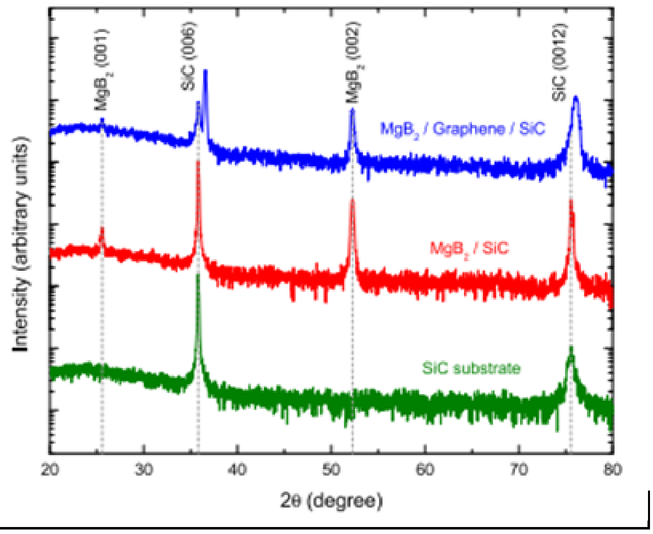

Figure 3. XRD pattern of MgB2 thin layer over graphene/SiC substrate (blue) compared to direct SiC substrate deposit (red) and SiC substrate alone (green).

XRD pattern (Fig. 4) and scanning electron microscopy (SEM) photographs provide further evidence that the film growth features that were discussed earlier provide support for. According to Figures 4(c) and 4(d), the MgB2 film that was directly deposited on SiC is thick, smooth, and contains grains that are densely packed and have an average size of between 500 and 700 nm. They are also tightly packed. There are a few microscopic hexagonal grains that can be observed on the surface of the film, which is consistent with the XRD pattern. This pattern also reveals that the film is orientated along the c-axis, as can be seen in Figure 4(c).

As can be seen in Figure 4(b), the grain sizes of MgB2 films that are normally generated on graphene/SiC substrates are typically less than 200 nm. The appearance of films that are deposited directly on SiC gives the impression that they are denser; nonetheless, these films also include grains that are packed extremely tightly together. This observation lends credence to the c-axis orientation as the predominant alignment, which suggests that the majority of the film's grains are aligned on its surface. This conclusion is in line with the findings of the XRD analysis. In addition, as can be seen in Figure 4(b), there are a few grains that are not parallel to the surface of the film but rather protrude outward, giving the impression of brilliant spots in the image.

Examining Figure 4(a) in further detail reveals that these brilliant spots are dispersed over the whole of the video. In terms of epitaxial quality, the findings suggest that the MgB2 film that was created on graphene/SiC is not as high-quality as the film that was formed directly on the SiC substrate. Despite the fact that the layer on graphene/SiC has good surface continuity and c-axis orientation, the differential in epitaxial quality is presumably explained, at least in part, by the larger lattice mismatch between graphene and MgB2. When compared to films grown directly on SiC substrates, it is possible that the MgB2 films that are generated on graphene/SiC may not have the greatest epitaxial growth performance. This may be due to the fact that the graphene that was transferred is not flat on the surface of the SiC.

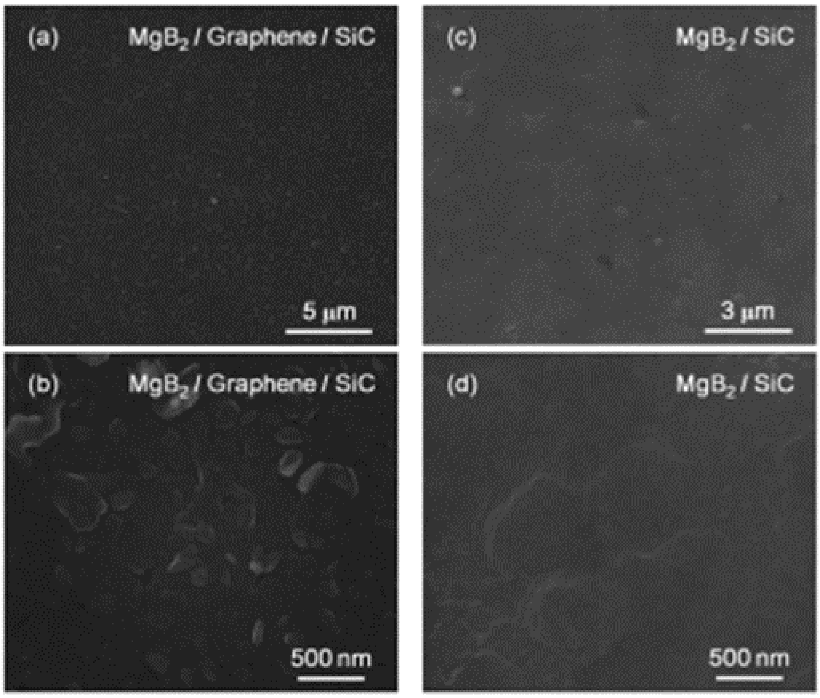

Figure 4. Compare the scanning electron microscopy (SEM) images of MgB2 thin films deposited directly on SiC substrates and graphene/SiC substrates [(c) and (d)].

Figure 5 illustrates the relationship between temperature and the electrical resistance of MgB2 films that were produced on graphene/SiC substrates as well as directly on SiC. The region of the superconducting transition is shown in more detail in Figure 5(b), which may be found here. Using high-pressure chemical vapour deposition (HPCVD), MgB2 films that are produced on SiC substrates often exhibit metallic behaviour, as seen in Figure 5(a). When the film is in its normal condition, it displays metallic behaviour because its resistance decreases with decreasing temperature. This occurs prior to the shift from a superconducting to a conventional state. It can be seen in Figure 5(b) that the critical temperature (Tc) is recorded in the superconducting state at 39.2 K. This state is defined as the transition point that is halfway between the two states. To determine the transition width, ΔTc, which is about 1 K, the difference between the temperature at which zero resistance is recorded (Tc, 0) and the onset temperature (Tc, onset) is used. This difference is employed to construct the transition width. As a result of these discoveries, the MgB2 film demonstrates a superconducting transition that is narrow, and its Tc value is comparable to that of bulk MgB2 samples. Previously conducted research has shown that the HPCVD technique has the potential to generate high-quality MgB2 thin films on SiC substrates. This is achieved by achieving high Tc values and a lower ΔTc. The fact that this is the case suggests that the current deposition settings have space for improvement, and that the performance of these films might likely be improved even more.

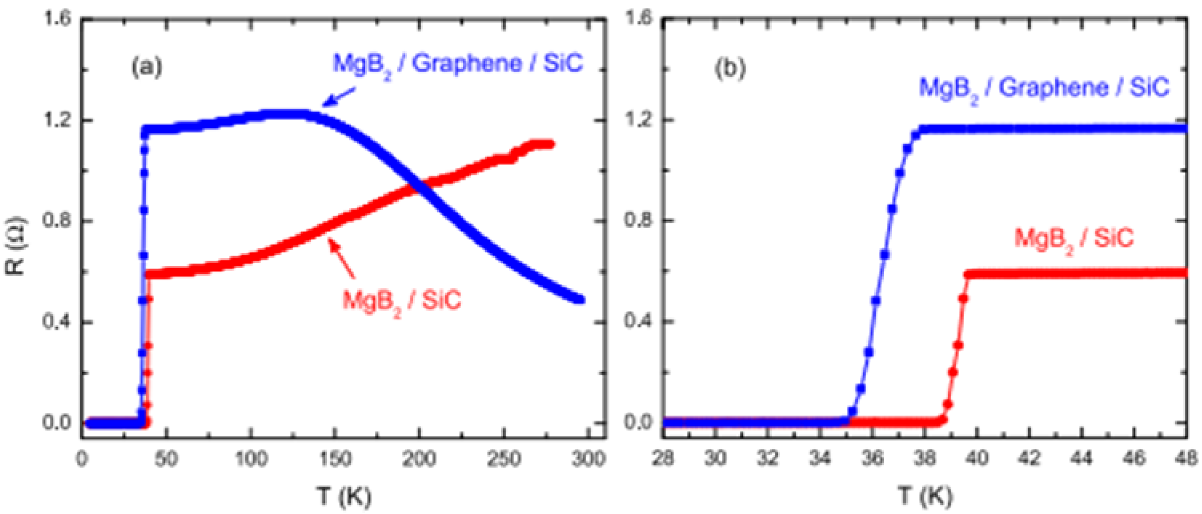

Figure 5. (a) Temperature dependence of MgB2 thin film resistance on graphene/SiC substrate (blue) vs directly on SiC substrate (red). (b) Magnified superconducting transition from (a).

when can be seen in Figure 5(a), the MgB2 film that was produced on the graphene/SiC substrate exhibits semiconducting behaviour when the temperature decreases from 300 K to 120 K. Below 120 K, the resistance decreases, much as it does in the film that was made on SiC. The semiconducting behaviour of films of magnesium bicarbonate (MgB2) that were created utilising a variety of experimental procedures on conventional substrates has also been observed at increased temperatures by the films. The semiconducting resistance pattern that emerges on MgB2 films produced on Si or SiC substrates by electron beam evaporation followed by post-annealing is an example of this phenomenon. This pattern is seen when the temperature drops from the ambient temperature to Tc or from higher temperatures. During the vacuum post-annealing process, magnesium is lost, which results in a shortage of magnesium in the film, as stated by a number of specialists. This behaviour is related to the magnesium loss. Due to the fact that the constant stoichiometry of the film is typically maintained by the high magnesium vapour pressure that is near to the substrate, this reasoning is not valid in this situation. On the other hand, post-annealing is not required with the use of HPCVD. It is possible that the slanted grains of the film, which are seen in Figures 4(a) and 4(b), are partially responsible for the observed semiconducting property. As a result of these grains, grain boundaries are formed, which may make it more difficult for electrical current to go between grains that are aligned on the surface of the film. There is a possibility that the intricate resistance behaviour of the MgB2 layer on graphene might be further influenced by the dispersion of electrical current inside the coating. At some point in the future, further study will be conducted in these areas.

As seen in Figure 5(b), the MgB2 layer that is deposited on the graphene/SiC substrate displays a temperature of 36.3 K and a temperature difference of 2.7 K. One of the differences between the film created indirectly and the one grown on the SiC substrate is that the former has a Tc that is 3 K lower and a ΔTc that is 2 K broader. The increased disorder in the MgB2 coating on graphene/SiC seems to be the possible source of the decreased Tc, as shown by the behaviour of the normal-state resistance and the images obtained from the scanning electron microscope (SEM). A pairbreaking effect may be accomplished by increasing disorder in MgB2 thin films, as shown by previous research. This enables a reduction in Tc by increasing the interband scattering between the and bands, hence reducing the intensity of the transmission line. In addition, this might be the explanation for the superconducting transition that is rather widespread. In spite of the fact that the Tc values of MgB2 films or bulk samples formed on SiC substrates are higher, the Tc of the film can be lower because of the near proximity of the graphene layer underneath it. The present study demonstrates that it is possible to reach Tc values that are close to 39 K by developing thin films of magnesium bicarbonate on graphene. Although more research is necessary to fully appreciate the little decrease in Tc, the results of this work indicate that it may be possible to investigate high-temperature superconductivity via the combination of MgB2 and graphene.

In this study, a thin layer of magnesium bicarbonate (MgB2) was put on top of graphene in order to create the hybrid structure. For the purpose of researching the superconducting proximity effect in these materials, micro- or nano-patterning procedures are necessary. As seen in Figure 2, etching has the potential to reveal the graphene layer that lies behind the more substantial MgB2 layer. Conventional lithography techniques might thus be used in order to build microstructures or connections that are composed of graphene and magnesium bicarbonate. The goal shape may be accomplished by depositing a thin layer of magnesium bicarbonate (MgB2) over graphene. This would be done after the deposition zone had been determined via the use of stencils and lithography. We wish to examine this approach further since it is analogous to the one that is used to manufacture graphene-based Josephson junctions with low-Tc superconductors. This is because the current findings indicate that this technology needs to be investigated further.

When compared to monolayer graphene, the MgB2 film used in this investigation, which is made up of around one hundred monolayers of MgB2, is noticeably more substantial. There has been a recent spike of interest in ultrathin superconducting films that are as thin as a few monolayers, which has been a potential new area for study into multiband superconductivity. According to theoretical studies, surface states in atomically thin MgB2 films have the potential to modify the superconducting gap spectrum. This spectrum is distinct from the bulk σ- and π-gaps, and it may be controlled by adjusting the film strain and the number of monolayers. It has been shown via experimental testing on few-monolayer MgB2 films that were created by molecular beam epitaxy (MBE) that surface states that include an additional gap do in fact exist. Using atomically thin MgB2 films, this presents a great opportunity to investigate the proximity effect in MgB2/graphene hybrids and to investigate how the various characteristics of these hybrids impact the proximity effect. HPCVD has been proven to be capable of generating MgB2 films with a thickness of 6 nm, which is equivalent to around 20 monolayers, according to previous research. It is possible that hitherto undiscovered frontiers might be revealed via the further refinement of this growth approach or by the use of MBE to produce atomically thin MgB2 on graphene.

COCNLUSION

For the purpose of enhancing the superconducting transition temperature (Tc), we have conducted research into the most effective methods for optimising the synthesis parameters of MgB2 and graphene bilayer films. Transferring CVD-grown monolayer graphene from copper foil to a target substrate allowed for the successful formation of MgB2 thin films on graphene. This was accomplished via the use of a hybrid physical-chemical vapour deposition (HPCVD) technique employed. Both the narrow superconducting transition of the film at an enhanced Tc of 36 K and the prevalence of the c-axis orientation with well-connected grains demonstrated that the optimised synthesis technique was successful. Because of the versatility of the method, it is feasible to transfer graphene onto a variety of substrates. Additionally, by simply repeating the transfer operations, it is able to alter the number of graphene layers. Additionally, the manufacturing of more sophisticated designs, such as alternating graphene and MgB2 layers, may provide new chances for increasing device performance. These opportunities may allow for the enhancement of device performance. With further optimisation of the synthesis parameters and the application of sophisticated patterning methods, these hybrid structures of MgB2/graphene could be used as platforms to study the proximity effect and build superconductors with higher temperatures, like Josephson junctions.

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