MICROWAVE ABSORPTION IN GRAPHENE FILMS: THEORY AND EXPERIMENT

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The interaction of Kα microwave radiation with ultrathin graphene films is studied. Although the thickness of these films is thousands of times smaller than the skin depth, they can absorb a significant fraction of the incident radiation. The possibility of controlling the amount of absorption and reflection of waves incident on graphene is demonstrated. In particular, by choosing the substrate parameters and the angle of incidence, it is possible to increase the absorption in graphene to >50%. For certain angles of incidence it is possible to have the TE-wave reflected, while the TM-wave is transmitted. These effects can be used to create ultrathin (atomic thicknesses) absorbers and polarizers.

Keywords: graphene, thin film, electromagnetic response, absorber, polarizer.

Introduction. The transmission of radiation through plane-parallel metallic films with a thickness much less than the skin layer can be understood intuitively by assuming that almost all the radiation passes through this kind of film without loss. This notion is, however, incorrect. It turns out that thin metallic slabs can absorb up to 50% and reflect up to 25% of the power of an incident wave. The impetus for a new understanding of the propagation of electromagnetic radiation through thin plane-parallel slabs of this kind arose from the discovery that the costly output windows in expensive microwave systems were unexpectedly breaking at intensities which calculations showed should be optimal [1–3]. Here the initial and most rational assumption regarding the contribution of impurities in the diamond, which are centers of elevated heat release, to the breaking of the windows was not confirmed, i.e., the diamond windows that were used were quite pure. After a complete analysis, it was proposed that the radiation had been absorbed in thin metallic layers which are deposited during operation of these devices. The transmission of electromagnetic waves through ultrathin metallic films has been studied in detail [4, 5]. Ultrathin metallic films of this kind on the window surfaces are efficient absorbers of electromagnetic radiation and the heat released as the energy is absorbed leads to fracture of diamond windows. Rapid advances in graphene materials have given a new impetus to research on absorption effects in ultrathin films. Thus, for example high absorption of electromagnetic radiation was first demonstrated experimentally and theoretically in thin films of pyrocarbon [6, 7]. The high absorption of electromagnetic waves in atomically thin layers of graphene was even more striking [8, 9]. In particular, it has been shown [6, 8, 10] that when graphene films are deposited on a substrate, the absorption in the film can be higher than in "free" graphene.

In this paper we propose methods for additional absorption of incident electromagnetic radiation in graphene by optimizing the thickness of the substrate and the angle of incidence of the wave.

Basic Expressions for the Transmission of Radiation through Ultrathin Metallic Films and Their Consequences. Features of transmission and absorption in thin metallic films. We consider the passage of a plane electromagnetic wave through a plane-parallel slab. The reflectivity $R$ (ratio of the intensity of the wave reflected from the slab to the incident intensity) and transmission $T$ (ratio of the intensity of the wave transmitted through the slab to the incident intensity) are found on solving the boundary value problem by equating the tangential components of the electric and magnetic fields at the two boundaries of the slab [11]:

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expression for reflection, transmission, and absorption in thin films:

\[
R = \left| 1 - \alpha^2 \right| \frac{\exp(-ikz_1l_1) - \exp(i\alpha)}{(1 + \alpha)^2 \exp(-ikz_1l_1) - (1 - \alpha)^2 \exp(i\beta)} \right|^2, \tag{1}
\]

\[
T = \left| \frac{4\alpha}{(1 + \alpha)^2 \exp(-ikz_1l_1) - (1 - \alpha)^2 \exp(i\beta)} \right|^2, \tag{2}
\]

where \(\alpha = k_z/c\) for the TM-wave and \(\alpha = k_z/k_{0z}\) for the TE-wave; \(k_z = (\epsilon - \sin^2 \theta)^{1/2}\); \(k_{0z} = (1 - \sin^2 \theta)^{1/2}\); \(k = \omega/c\); \(\omega\) is the frequency of the electromagnetic radiation; \(\theta\) is the angle of incidence; and \(\epsilon\) and \(l_1\) are the dielectric constant and thickness of the slab. The thickness of the ultrathin slabs considered here is much less than the skin layer, so that the phase shift over the thickness of the slab is very small. In this case, Eqs. (1) and (2) can be simplified by expanding in terms of the small parameter \(k_zl_1 \ll 1\). This expansion for the case of \(\epsilon' \ll \epsilon'' = 4\pi\sigma/\omega\) yields very simple expressions for reflection, transmission, and absorption in thin films:

\[
T = \left| 1/(1 + l_1/l_\alpha) \right|^2, \quad R = \left| 1/(1 + l_\alpha/l_1) \right|^2, \quad A = 1 - T - R = (2l_1/l_\alpha)(1 + l_1/l_\alpha)^2, \tag{3}
\]

where \(l_\alpha = 2\epsilon\sigma_0/(\sigma \cos \theta)\) for the TM-wave and \(l_\alpha = (2\epsilon\sigma_0 \cos \theta)/\sigma\) for the TE-wave. Equations (3) for normal incidence \((\theta = 0)\) are given in [5].

An analysis of Eqs. (3) shows that as the thickness \(l_1\) of the slab increases the transmission \(T\) decreases while the reflectivity \(R\) increases. This behavior is fully to be expected, but the "unexpected" thing is that the transmission is substantially below unity, while reflection becomes significant by thicknesses \(l_1 \sim l_\alpha\). The parameter \(l_\alpha\) for metals is several orders of magnitude smaller than the thickness of the skin layer. Even more "unexpected" is the high absorption in a thin metallic slab. Equation (3) implies that the absorption is maximal and reaches 50% at a thickness \(l_1 = l_\alpha\) (transmission and reflection are then each 25%). We note another peculiarity: the different dependence of \(l_\alpha\) and, thereby, of all the coefficients \(T, R,\) and \(A\) (Fig. 1) on the angle of incidence for different polarizations. Thus, for TM-polarization \(l_{\alpha,\text{TM}} = 2\epsilon\sigma_0/(\sigma \cos \theta)\) increases with increasing angle of incidence, while, on the other hand, for TE-polarization \(l_{\alpha,\text{TE}} = (2\epsilon\sigma_0 \cos \theta)/\sigma\) decreases.

This behavior is explained by the fact that, in the case of a TE-wave, the electric field is always in the plane of the slab, so it induces tangential currents, while the "effective" thickness of the slab increases in inverse proportion to \(\cos \theta\). Thus, for the TE-wave these effects begin to play a role at smaller slab thicknesses compared to the case of normal incidence. In the case of a TM-wave, the angle between the normal and the electric field vector \(E\) becomes important: for \(\theta \neq 0\) the tangential component of the vector \(E\) is less than for \(\theta = 0\), so that the induced tangential current is smaller.

With an appropriate substitution these expressions for \(T, R,\) and \(A\) are easily generalized to the case where the processes are not taking place in free space but in a waveguide. Thus, for example, it is easy to show that for a rectangular waveguide, expressions for \(T, R,\) and \(A\) can be obtained with the substitution \(\cos \theta \rightarrow \sqrt{1 - (\pi a/ka)^2} - (\pi m/\pi k)^2)^{1/2}\) [6], where \(a\) and \(b\) are the linear dimensions of the waveguide and \(n\) and \(m\) are integers corresponding to the waveguide modes (for a TM-wave both of these indices must be nonzero and for TE-polarization, at least one index must be nonzero). The validity of this substitution is evident since the modes of a rectangular waveguide are combinations of plane waves with an angle of inclination relative to the axis of the waveguide. Similarly to the situation in free space, a reduction in the thickness required for maximum absorption owing to inclined incidence of a TE-wave in a waveguide occurs as the cutoff frequency is approached.

Thus, thin films can be used, on one hand, as concentrators of energy within an extremely small volume (absorption of half the incident radiation for nanometer thicknesses) and, on the other, as efficient polarizers. In particular, for a thickness and angle of incidence that satisfy the inequality

\[
l_{\alpha,\text{TE}} \ll l_1 \ll l_{\alpha,\text{TM}}, \tag{4}
\]

the film reflects a TE-polarized wave and transmits a TM-wave.

In deriving Eqs. (3) we have used the conditions at both boundaries of the film. The film itself is characterized by a bulk specific conductivity \(\sigma\). Since the thickness of these films is considerably smaller than the thickness of the skin layer, in Eqs. (3) and afterward the bulk conductivity shows up together with the thickness of the slab in the form of the product \(\sigma l_1\). This is essentially a surface conductivity. In the present limit \((k_zl_1 \ll 1)\) it is simplest to treat the film as infinitely thin and, instead of using the exact boundary conditions, to use effective boundary conditions, according to which the tangential components of the electric fields on different sides of the slab are equal and there is a jump in the tangential magnetic

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component proportional to the surface current induced in the film (i.e., by the surface conductivity $\sigma_l$). This approach leads to exactly the same Eq. (3). In addition, the description based on surface currents and surface conductivity also holds in the case where the thickness of the slab is less than the electron mean free path and the concept of a specific bulk conductivity loses all meaning [12].

**Features of transmission and absorption in metallic films on substrates.** As noted above, the maximum absorption in a separately positioned thin metallic film is 50%. The absorption can be increased by combining additional elements with a thin film. One way of increasing the absorption in a film is to optimize the film thickness. Thus, it has been shown [8] that for a system consisting of a thin film on a dielectric substrate, the expressions for the reflectivity and transmission for a TE-wave are given by

$$R_{TE} = \frac{(1 - \alpha_{TE})(1 + B_{TE} + \alpha_{TE})\varepsilon_+ - (1 + \alpha_{TE})(1 + B_{TE} - \alpha_{TE})\varepsilon_-}{(1 + \alpha_{TE})(1 + B_{TE} + \alpha_{TE})\varepsilon_- - (1 - \alpha_{TE})(1 + B_{TE} - \alpha_{TE})\varepsilon_+}^2, \quad (5)$$

$$T_{TE} = \left|\frac{4\alpha_{TE}[(1 + \alpha_{TE})(1 + B_{TE} + \alpha_{TE})\varepsilon_- - (1 - \alpha_{TE})(1 + B_{TE} - \alpha_{TE})\varepsilon_+]}{(1 + \alpha_{TM})(1 + B_{TM} + \alpha_{TM})\varepsilon_- - (1 - \alpha_{TM})(1 + B_{TM} - \alpha_{TM})\varepsilon_+}\right|^2, \quad (6)$$

where $\alpha_{TE} = k_z/k_{0e}$, $B_{TE} = \eta/(\varepsilon_0 \cos \theta)$, $\varepsilon_\pm = \exp(\pm ik_z l)$, $l$ is the substrate thickness, $\eta$ is the surface conductivity, given by $\eta = \sigma l$, for a thin film with a thickness exceeding the electron mean free path, $\sigma$ is the conductivity, and $l_1$ is the film thickness.

For a TM-polarized wave,

$$R_{TM} = \frac{(1 - \alpha_{TM})(1 + B_{TM} + \alpha_{TM})\varepsilon_- - (1 + \alpha_{TM})(1 + B_{TM} - \alpha_{TM})\varepsilon_+}{(1 + \alpha_{TM})(1 + B_{TM} + \alpha_{TM})\varepsilon_- - (1 - \alpha_{TM})(1 + B_{TM} - \alpha_{TM})\varepsilon_+}^2, \quad (6)$$

$$T_{TM} = \left|\frac{4\alpha_{TM}[(1 + \alpha_{TM})(1 + B_{TM} + \alpha_{TM})\varepsilon_- - (1 - \alpha_{TM})(1 + B_{TM} - \alpha_{TM})\varepsilon_+]}{(1 + \alpha_{TM})(1 + B_{TM} + \alpha_{TM})\varepsilon_- - (1 - \alpha_{TM})(1 + B_{TM} - \alpha_{TM})\varepsilon_+}\right|^2, \quad (6)$$

where $\alpha_{TM} = k_0\varepsilon/k_z$ and $B_{TM} = (\eta \cos \theta)/(\varepsilon_0 c)$.

It is important to note that Eqs. (5) and (6) have been obtained for a system geometry in which the substrate is facing the incident radiation. It has been shown theoretically and experimentally [6] that in this geometry, absorption in the system can exceed 50%. In a way similar to the above problem, the effect of a thin film with inclined incidence increases for a TE-wave ($B_{TE} \sim 1/\cos \theta$) and decreases for a TM-polarized wave ($B_{TM} \sim \cos \theta$). An analysis of Eqs. (5) and (6) shows that there is a range of thicknesses for which the absorption in the film takes on extreme values. These values occur where the derivative of the absorption with respect to thickness goes to zero. It is easy to show that this condition leads to the simple equality $\sin(2k_z l) = 0$, which for the smallest nonzero thickness corresponds to a quarter wave plate. In order for the extremum to correspond to a maximum, the following condition must hold for the conductivity:

$$B_i > \alpha_i - 1, \quad i = TM \text{ or } TE. \quad (7)$$
Under these conditions, the absorption
\[ A_i = 4 \alpha_i^2 B_i/(1 + \alpha_i^2 + B_i)^2. \] (8)

Thus, Eq. (8) implies the existence of an optimum \( \alpha \):
\[ \alpha_i^2 = 1 + B_i. \] (9)

Then the maximum absorption in the slab is
\[ A_{\text{max}} = B_i/(1 + B_i). \] (10)

In Eqs. (7), (9), and (10) the parameters are the dielectric constant, the angle of incidence, etc. These parameters can be chosen for a TE-polarized wave so as to make the absorption >50% (maximum absorption without a substrate), and it is possible to approach complete absorption (an almost black electromagnetic hole for \( B_i >> 1 \)). In the following we use the standard form of the conductivity [12] for graphene. We have made experimental measurements in a waveguide exposed to a TE_{10} wave in the \( K_a \) band. It follows from the general form of the surface conductivity of graphene that in this band, the conductivity is essentially real (the imaginary part is many orders of magnitude smaller).

**Results and Discussion.** To confirm the theoretical results we have made an experimental study of the electromagnetic properties of multilayer (sandwich) graphene/polymer (polymethyl methacrylate PMMA) structures on quartz substrates at microwave frequencies. The synthesis of these structures is discussed in detail elsewhere [8]. We dwell only on the main steps. In the first step, graphene produced by chemical vapor deposition (CVD) at a temperature of 1000°C in a methane atmosphere on a copper substrate (thickness 25 mm, 99.8% pure) was coated with a ~600 nm-thick PMMA layer by spin coating. The thickness of the polymer layer was monitored with a VeeCo Dektak6M profilometer. The copper substrate was then dissolved in iron chloride (FeCl_3). The resulting “freestanding” graphene film coated with the polymer was repeatedly washed in distilled water and transferred to a quartz substrate of specified thickness (7.2 × 3.4 × 0.53 mm substrates were used). Repeating this procedure layer by layer yielded sandwich structures of graphene layers separated by the polymer. Samples with from 1 to 6 graphene/PMMA layers were studied. In order to further monitor the properties of the graphene, a 10 × 10 mm freestanding film was transferred to a separate quartz substrate (radius 55 mm, thickness 0.53 mm) and the surface layer of PMMA was then dissolved in acetone. The quality of the graphene was monitored by scanning microscopy (SEMLEO 1455 Vand) and Raman scattering spectroscopy, and the optical absorption spectra were examined. These data showed that the test films consist predominantly of monolayer graphene.

The electromagnetic response of the samples at frequencies of 26–37.5 GHz was studied using a panoramic standing wave and attenuation measurement system (R2-408 R VSWR and Transmission Loss Meter, Elmika, Vilnius, Lithuania) intended for measuring the moduli of the reflection and transmission coefficients (the ratios of the amplitudes of the incident wave to the amplitude of the reflected wave (\( S_{11} \)) and to the transmitted wave (\( S_{21} \)), VSWR, and attenuation of the waveguide structures. In accordance with the measurement techniques (described in detail in [9] and [13]), a sample in the form of a thin film on a quartz substrate was placed in a cross section (7.2 × 3.4 mm) of the waveguide perpendicular to the direction of propagation of the electromagnetic wave (Fig. 2). For testing the theoretical predictions of the effect of substrate thickness on the electromagnetic response of these samples, the thickness of the substrates was varied by placing additional plane-parallel epoxy (EPIKOTETM Resin 828) slabs of thickness 0.7, 0.9, 1.0, 1.2, or 1.5 mm in the waveguide next to the quartz slab (Fig. 2).

There were several reasons for choosing this method of simulating different thicknesses of the substrate. Using additional polymer slabs of different thicknesses makes it easy to change the resultant thickness of the substrate (the thickness of the epoxy + 0.53 mm quartz), which, in turn, makes it much easier to do the experiment by reducing the required number of samples of multilayer structures with specified geometric parameters. At microwave frequencies epoxy, like quartz [8], is a dielectric with low absorption [14]. It is, however, much easier to make these slabs out of polymer materials than quartz.

Figure 3 shows the experimental transmission (\( T = S_{21}^2 \)), reflectivity (\( R = S_{11}^2 \)), and absorption (\( A = 1 - T - R \)) for samples containing up to six graphene/PMMA layers at a frequency of 31 GHz as functions of the substrate thickness. These curves show that the maximum absorption (~80%) in this frequency range, which corresponds to the reflection minimum (1–2%), is observed for a substrate thickness of 1.25 mm. These experimental data are in good agreement with the above theoretical predictions (Fig. 4).

The extremely high absorption of microwaves by graphene should be noted. Even a single graphene layer absorbs ~30% of the incident wave. The optical absorption, which is determined by interband transitions, is considerably lower (\( \pi \alpha \sim 2.3\% \)) [15]. The absorption in the sandwich structure is exclusively caused by graphene. The imaginary part of the
Fig. 2. Illustrating a method for making measurements at microwave frequencies and experimental graphene/PMMA sandwich samples. The electromagnetic radiation propagates along the $z$ axis.

Fig. 3. Reflectivity (a), transmission (b), and absorption (c) as functions of substrate thickness; the number of graphene layers is 1 (1), 2 (2), 3 (3), 4 (4), 5 (5), and 6 (6).

Fig. 4. Theoretical dependence of absorption on substrate thickness; the number of graphene layers is 1 (1), 2 (2), 3 (3), 4 (4), 5 (5), and 6 (6).
dielectric constants of quartz and epoxy resin satisfy the relation \( k_\varepsilon \ll 1 \) in this frequency range; this in turn determines the low level of absorption in these materials. A PMMA layer with a width of \( \sim 600 \) nm is optically thin in this range and does not contribute to the overall level of absorption.

**Conclusions.** The feasibility of adjusting the electromagnetic response of thin graphene films deposited on a dielectric substrate over a wide range has been demonstrated experimentally and theoretically. It has been shown that the level of transmission, reflection, and absorption can be controlled by the substrate thickness, angle of incidence, and polarization of the incident wave. The effects described here can be used to create detectors, sensors, and efficient polarizers over a wide spectral range. Given the possible absorption of macroscopically large amounts of energy in microscopic volumes, this effect may be useable in a wide variety of applications in power engineering.

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**REFERENCES**

1. D. H. Priest and R. C. Talcott, *IRE Trans. Electron Devices*, 8, 243–251 (1961).
2. L. K. Ang, Y. Y. Lau, R. A. Kishen, and R. M. Gilgenbach, *IEEE Trans. Plasma Sci.*, 26 (3), 290–295 (1998).
3. A. Valfells, L. K. Ang, Y. Y. Lau, and R. M. Gilgenbach, *Phys. Plasmas*, 7 (2), 750–757 (2000).
4. H. Bosman, Y. Y. Lau, and R. M. Gilgenbach, *Appl. Phys. Lett.*, 82, 1353–1355 (2003).
5. V. G. Andreev, V. A. Vdovin, and P. S. Voronov, *Pis’ma Zh. Tekh. Fiz.*, 29, 68–73 (2003).
6. K. Batrakov, P. Kuzhir, S. Maksimenko, A. Paddubskaya, S. Voronovich, T. Kaplas, and Yu. Svirko, *Appl. Phys. Lett.*, 103, 0731171–0731174 (2013).
7. P. Kuzhir, A. Paddubskaya, S. Maksimenko, T. Kaplas, and Yu. Svirko, *Nanoscale Res. Lett.*, 8 (1), 60–66 (2013).
8. K. Batrakov, P. Kuzhir, S. Maksimenko, A. Paddubskaya, S. Voronovich, Ph. Lambin, T. Kaplas, and Yu. Svirko, *Sci. Rep.*, 4, 071911–071914 (2014).
9. P. Kuzhir, N. Volynets, S. Maksimenko, T. Kaplas, and Yu. Svirko, *J. Nanosci. Nanotechnol.*, 13 (8), 58464–58467 (2013).
10. S. Voronovich, A. Paddubskaya, K. Batrakov, P. Kuzhir, S. Maksimenko, T. Kaplas, and Yu. Svirko, *Appl. Sci.*, 4, 255–264 (2014).
11. L. D. Landau and E. M. Lifshitz, *Electrodynamics of Continuous Media*, 2nd edn., *Theoretical Physics*, Vol. VIII [in Russian], Nauka, Moscow (1982).
12. S. A. Mikhailov, in: S. Yamashita, Y. Saito, and J. H. Choi (Eds.), *Carbon Nanotubes and Graphene for Photonic Applications*, Ch. 7, Woodhead Publishing (2013), pp. 171–219.
13. B. Chung. *Progres. Electromag. Res*. 75, 239–252 (2007).
14. A. Paddubskaya, D. Bychanok, A. Plyushch, P. Kuzhir, A. Nemilentsau, S. Maksimenko, S. Bellucci, L. Coderoni, F. Micciulla, I. Sacco, and G. Rinaldi, *J. Nanoelectron. Optoelectron.*, 7, 81–86 (2012).
15. R. Nair, P. Blake, A. Grigorenko, K. Novoselov, T. Booth, T. Stauber, N. Peres, and A. Geim, *Science*, 320, 1308 (2008).