Theoretical estimation of the increase in the photoelectric effect cross section of the X-ray interaction with charged graphene

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Abstract. Theoretical perspective discussions about negatively charged graphene slabs and their radiation properties are presented. It is shown that additional electrons in graphene, injected in a charging process, can enhance the value of the total cross section of photon radiation interactions, especially for X-rays and lower energies. A detailed discussion about the photoelectric effect on electrons trapped in quantum wells in graphene supplements the investigations on light carbon radiation shields. It is concluded that hundreds of parallel graphene slabs would give a significant contribution to modern radiation protection.

1 Introduction

The total cross section of medium- and high-energy photon radiation (namely, gamma and X-rays) interactions with matter is composed of four main components based on the photoelectric effect, Compton scattering, e^-e^+ pair production in the nuclear field, and analogical pair production in the quasi-free electron field (triplet production) [1]. All of them, except the pair production in the nuclear field, are strictly connected to the total number of electrons per atom, not the nucleus itself. Recent theoretical investigations have discussed that an increase in the number of electrons (*e.g.*, by charging) can increase the total cross section value of photon radiation. Graphene, which has great electric properties, can be especially dedicated to that effect [2].

In the first approximation, the calculations of that enhanced cross section can be made under the assumption that graphene (carbon) is neutral with additional electron gas corresponding to the negative charge [2]. That approach clearly proves that additional electrons will increase the cross section of Compton scattering and the cross section of triplet production, which shows that their formulae are based on free electrons. However, in the second approximation the charged graphene is a more complicated structure and some additional compounds for lower energies, namely from the photoelectric effect, should be taken into consideration.

2 Charged graphene

Detailed calculations of charged material properties are usually provided using the density functional theory (DFT) where the *ab initio* self-consistent electronic structure calculations are used [3]. Especially, some of the charged-graphene properties were numerically calculated a few years ago [4, 5].

Let us define the *graphene slab* as a thin and flat material (d width) composed of many single parallel graphene monolayers¹ with the layer density (*i.e.* number of layers per μm) as w_G . The first question is: what is the maximal surface charge density, Q_{max} (called the *charge* within this paper)? The answer can be found in the cited paper: for the graphene slab with a standard distance between single layers (vacuum spacing) of $s = 20 \text{ \AA} = 2 \text{ nm}$, the maximal charge per layer equals $Q_1 = 6.457 \cdot 10^{-6} \text{ C/cm}^2$ [4]. Assuming that the typical width of the graphene slab is $d \approx 100 \mu\text{m}$,

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¹ This definition is not equivalent to graphite where each layer is connected by metallic bonding [6] and located much closer to each other (the distance between layers in graphite is approx. 0.33 nm, which is 6 times smaller than that between graphene layers in the graphene slab).

Fig. 1. A scheme of two parallel graphene layers (distant from each other by s) with the plane-averaged potential for electrons, dipping at $s/2$ and forming a quantum well [5].

one can find there approximately 50000 layers ($w_G \approx 500 \mu\text{m}^{-1}$), which results in the theoretically maximal possible charge as $Q_{\text{max}(1)} = 0.323 \text{ C/cm}^2$ per slab. This is equivalent to $q_1 \approx 0.01$ additional electrons per atom. Providing the same calculations for quite different assumptions and methods [5], one can find that the maximum possible charge is almost one order of magnitude greater ($Q_2 = 6.1 \cdot 10^{-5} \text{ C/cm}^2$ per single layer and $Q_{\text{max}(2)} = 3.05 \text{ C/cm}^2$ per whole slab; this is equivalent to $q_2 \approx 0.1$ additional electrons per atom).

The important point from the charged graphene numerical calculations is the fact that between two parallel layers a quantum well for additional electrons exists [5], see fig. 1. More than that, when the distance between layers (vacuum spacing, s) increases, the number of electrons inside the well also increases. However, for $s = 2 \text{ nm}$ and Q_2 , only $\epsilon \approx 1.34\%$ of additional electrons per atom are trapped inside the quantum well [5]. One must note, that in the cited paper the authors claimed that the Fermi energy for that quantum well and for the standard atomic well are the same. Using that claim we can now assume that the work function values are approximately the same, and equal $W \approx 4.26 \text{ eV}$ [5, 7]. This means that the photoelectric effect can occur for the electrons trapped in the quantum well between charged graphene layers. Thus the cross section for that specific effect can be found for low photon energies, especially for X-rays.

3 Cross section

Assuming, that some small number of electrons (ϵq) are trapped between layers, one can present the total cross section per atom as

$$\sigma_{\text{total}}(q') = \sigma_{\text{ph}}(Z') + \sigma_{\text{ph}}(\epsilon q) + q' \sigma_C + \sigma_{\text{pair}} + q' \sigma_{\text{trip}}, \quad (1)$$

where $q' = Z + q$ and q represents the additional charge (number of electrons) per atom, while $Z = 6$ for graphene (carbon). Additionally, $Z' = Z + (1 - \epsilon)q$, which represents electrons located in an atomic well. One can use also the notion of $q = Qn^{-1}$, where n is the concentration of atoms in a single layer ($n \approx 3.82 \cdot 10^{15} \text{ cm}^{-2}$ for graphene). The rest of the symbols in eq. (1) corresponds to the effects: photoelectric σ_{ph} , Compton σ_C (on a single electron), e^-e^+ pair creation σ_{pair} and triplet creation σ_{trip} (on a single electron) [1, 2]. One must note, however, that the photoelectric effect on the charged carbon atom (Z') and that on trapped electrons (ϵq) are treated independently.

The cross section of the photoelectric effect is usually described by the Pratt-Scofield equation for photon energies $> 0.5 \text{ MeV}$ [1]. For less precise values one can use the older Hall equation [8]. For lower energies, especially for X-rays, the photoelectric cross section is described by the relativistic Sauter equation [8], which depends on the electron work

Fig. 2. Excess relative ratio (R) of the total cross section of the single layer charged graphene to the total cross section of non-charged graphene, for two surface charge densities, $Q_1 < Q_2$.

function:

$$\sigma_{\text{ph}}(Z) = \frac{3}{2} \varphi_0 \alpha^4 \left(Z \frac{E_e}{E_x} \right)^5 (\gamma^2 - 1)^{3/2} \left[\frac{4}{3} + \frac{\gamma(\gamma - 2)}{\gamma + 1} \left(1 - \frac{1}{2\gamma(\gamma^2 - 1)^{1/2}} \ln \frac{\gamma + (\gamma^2 - 1)^{1/2}}{\gamma - (\gamma^2 - 1)^{1/2}} \right) \right], \quad (2)$$

where

$$\gamma = (1 - \beta^2)^{-1/2} = \frac{E_x - E_B + E_e}{E_e}, \quad (3)$$

and $E_x = h\nu$ is the photon energy given in eV, $E_e = m_e c^2 \approx 5.11 \cdot 10^5$ eV is the electron rest mass energy, E_B is the binding energy of the electron (assumed to be equivalent to the work function, $E_B \approx W \approx 4.26$ eV), $\alpha = e^2/(\hbar c) \approx 1/137$ (fine-structure constant), and φ_0 is the Thomson cross section ($\varphi_0 = 8\pi e^4/(3E_e^2) \approx 0.66526$ barn). In the case of $\sigma_{\text{ph}}(Z')$ and $\sigma_{\text{ph}}(\epsilon q)$ components in eq. (1), one shall assume to substitute $Z \rightarrow Z'$ and $Z \rightarrow \epsilon q$ in eq. (2), respectively.

Using the Beer-Lambert-Bouguer law, one can now calculate the linear attenuation coefficient for the photon interaction on the graphene slab as

$$\mu = \sigma_{\text{total}} n w_G, \quad (4)$$

which can be directly used in radiation protection calculations.

4 Results

4.1 Total cross section

The process of charging increases the value of total cross section in a wide range of energies. Figure 2 presents the excess relative ratio of the total cross section of charged graphene to the total cross section of non-charged graphene, namely $R = \sigma_{\text{total}}(q')/\sigma_{\text{total}}(Z) - 1$, for the single graphene layer. One can clearly see that R decreases with the energy increase. However, for photon energies $E_x < 10$ keV the value of R is constant (plateau) and equals $R \approx 0.0087$ for Q_1 and $R \approx 0.0847$ for Q_2 . Generally, for $Q < 10^{-4}$ C/cm² the relation $R(Q)$ in this plateau can be approximated by the empirical formula: $R_{\text{plateau}} = 1514.2 \cdot \exp(1.0083 \cdot \ln Q)$.

Additionally, for photon energies $150 \text{ keV} < E_x < 1.5 \text{ MeV}$, the value of R meets the second plateau and equals there $R \approx 0.0018$ for Q_1 and $R \approx 0.0166$ for Q_2 . Generally, the relation $R(Q)$ in the second plateau can be approximated by the linear relation: $R_{\text{plateau}} = 272.5Q$, but for $Q > 0.02$ C/cm² the second plateau disappears.

Fig. 3. The ratio (P) of the radiation intensities between charged and non-charged single graphene slabs ($d = 100 \mu\text{m}$) for two surface charge densities, $Q_{\max(1)} < Q_{\max(2)}$ (see eq. (6)).

4.2 Photoelectric effect

For the electrons located in the quantum well outside atoms (ϵq), the cross section calculated by eq. (2) is very small and equals $1.8 \cdot 10^{-18}$ barn/atom for Q_1 and $1.3 \cdot 10^{-13}$ barn/atom for Q_2 , for $E_x = 1 \text{ keV}$ for both. Those values decrease when the energy increases.

The presented values of the cross section are too small to be significant from a radiation protection point of view. However, for a hypothetical $Q = 0.01 \text{ C/cm}^2$ (in the single layer) it would be easily detectable for 1 keV ($\sigma = 0.016 \text{ b/atom}$) and for a hypothetical $Q > 0.5 \text{ C/cm}^2$ becomes significant for the radiation protection of X-rays.

Values of $\sigma_{\text{ph}}(\epsilon q)$ can be approximated by the empirical equation (obtained by regression methods as presented in [1]), for $1 \text{ keV} < E_x < 0.4 \text{ MeV}$:

$$\sigma_{\text{ph}}(\epsilon q) \approx 6.18263 \cdot 10^{11} (\epsilon q)^5 E_x^{-3.4437}, \quad (5)$$

where the photon energy E_x is given in eV and q is given in e/atom. Equation (5) is faster and simpler than eq. (2) and differences between their results are less than 10% (for $2 \text{ keV} < E_x < 300 \text{ keV}$).

4.3 Linear attenuation coefficient

The typical physical quantity which can be measured in the mentioned problem is the radiation intensity, I , which is reduced after radiation passes through matter. The easiest way to test the radiation properties of charged graphene is to measure the intensity of passing radiation before ($I_{\text{non-charged}}$) and after (I_{charged}) charging and calculate its ratio, P . Thus, using the Beer-Lambert-Bouguer law, one can find that

$$P = \frac{I_{\text{charged}}}{I_{\text{non-charged}}} = e^{d(\mu_{\text{non-charged}} - \mu_{\text{charged}})}, \quad (6)$$

where d is the thickness of the slab, and μ corresponds to the linear attenuation coefficient of non-charged and charged slabs of graphene, respectively (see eq. (4)). The situation where $P < 1$ means the reduction of I_{charged} after charging and the success of the mentioned effect detection.

However, the results given by eq. (6) for a single graphene slab ($d = 100 \mu\text{m}$) indicate that differences between X-rays intensity are undetectable, $P > 0.9999$ for $E_x > 7 \text{ keV}$, both for $Q_{\max(1)}$ and $Q_{\max(2)}$, see fig. 3. For much smaller energies ($< 1.5 \text{ keV}$ for $Q_{\max(2)}$ and $< 0.7 \text{ keV}$ for $Q_{\max(1)}$) the P value drops below 0.99, which creates theoretical chances for direct detection of the radiation intensity, I .

The results presented above were calculated for a single graphene slab with $d = 100 \mu\text{m}$ only. That is why the expected results are so weak. To enhance the intensity decrease, P , one must use many parallel slabs. For example for 10^5 slabs the P value is less than 0.99 when $E_x < 100 \text{ keV}$ for $Q_{\max(2)}$ or $E_x < 20 \text{ keV}$ for $Q_{\max(1)}$.

5 Discussion

The charging process of graphene changes its radiation properties. More electrons improve the cross section of ionizing radiation interactions, because most effects responsible for those interactions are based on electrons. Detailed theoretical considerations about the Compton effect and triplet production in charged graphene have been previously described [2] but the presented paper discusses the photoelectric effect and its implications in the same context.

The photoelectric effect therefore can be treated in two separate ways: as an effect on electrons trapped in the atomic well (their number equals $Z + (1 - \epsilon)q$) and as an effect on electrons trapped in the quantum well created between graphene layers (their number equals ϵq). The Sauter equation was used to calculate precise values of the cross section for both cases, which is necessary for the calculation of the linear attenuation coefficients. Additionally, the empirical equation for the cross section of the photoelectric effect on non-atomic bound electrons (ϵq) was formulated as eq. (5). This would be useful for some quick but less precise calculations.

The presented results were formulated for a single graphene slab only. Figure 2 contains the relative ratio of charged to non-charged cross section values. One can clearly see that the biggest differences (and the strongest effects) are located in the low-energy region (below 100 keV). It is caused mostly by the photoelectric effect and, in some part, by the Compton scattering. This is why a reduction of radiation intensity is given for low and very low energies, as shown in fig. 3.

Generally speaking the described effect of the intensity reduction is very small (fig. 3). It would be much stronger for higher values of Q , however, it is questionable whether it is possible from a physical and technical point of view. The maximum number of additional electrons per atom ($q_2 = 0.1 \text{ e/atom}$ for Q_2) seems to be quite a small number; however, larger values would change the internal graphene structure and potentially destroy the whole slab (for example, for a hypothetical 0.5 C/cm^2 one has approximately 817 additional electrons per atom which would immediately tear apart the structure). Another way is to charge few hundreds or thousands of parallel graphene slabs to enhance the proposed effect. This is a main field of future experimental investigations [2].

One must note, that the term *slab* corresponds to the many single monolayers of graphene, where additional electrons can be injected between them. This is not equivalent to graphite, where layers are connected by metallic bonding [6]. This form of graphene (*e.g.* graphene flakes as slabs) can be created in experimental way, *e.g.* in the Institute of Electronic Materials Technology (ITME), Poland.

The proposed effect can be also enhanced when the negatively charged slab is compared not to a neutral one, but to a positively charged slab. When one assumes that the absolute values of positive and negative charges are the same, the total differences of charges are double, *e.g.* $|2Q_{\max(1)}|$ and $|2Q_{\max(2)}|$. For that assumption the result of $P < 0.99$ from the section 4.3 above would change into: for single slab $< 1.8 \text{ keV}$ for $Q_{\max(2)}$ or $< 0.9 \text{ keV}$ for $Q_{\max(1)}$, and for 10^5 slabs: $< 600 \text{ keV}$ for $Q_{\max(2)}$ or $< 25 \text{ keV}$ for $Q_{\max(1)}$. This result once again shows that the radiation properties of charged slabs strongly depend on the absolute value of the charge.

Of course graphene is not the only material which can be useful for the proposed effect demonstration. Many other materials, which can be charged (*e.g.* steel, graphite, etc.), can be used as well; however, the expected effects would be smaller because of the relatively better electric properties of graphene.

One should add that the enhanced cross section values of photon interactions also can be measured in an indirect way. The measurement of photoelectrons and/or the measurement of photons in Compton spectroscopy are the easiest ways to check the enhancement of the total cross section. It has been proved also that the Raman active mode of graphene is changing due to the charging process [4]. Nevertheless, the experimental investigations of the proposed effect will be the main subject of future studies.

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