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Graphene intracavity spaser absorption spectroscopy☆

Yu.E. Lozovik [a,b,c,d,e], I.A. Nechepurenko [a,c], A.V. Dorofeenko [a,b,c,*]

[a] *Dukhov Research Institute of Automatics (VNIIA), 22 Suschevskaya, Moscow 127055, Russia*
[b] *Institute for Theoretical and Applied Electromagnetics RAS, 13 Izhorskaya, Moscow 125412, Russia*
[c] *Moscow Institute of Physics and Technology, 9 Institutskiy pereulok, Dolgoprudny 141700, Moscow Region, Russia*
[d] *Institute for Spectroscopy RAS, 5 Fizicheskaya, Troitsk 142190, Russia*
[e] *Moscow National Research Nuclear University MEPhI, 31 Kashirskoe Highway, Moscow 115409, Russia*

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Abstract

We propose an intracavity plasmon absorption spectroscopy method based on graphene active plasmonics. It is shown that the plasmonic cavity contribution to the sensitivity is proportional to the quality factor Q of the graphene plasmonic cavity and reaches two orders of magnitude. The addition of gain medium into the cavity increases the sensitivity of method. Maximum sensitivity is reached in the vicinity of the plasmon generation threshold. The gain contribution to the sensitivity is proportional to $Q^{1/2}$. The giant amplification of sensitivity in the graphene plasmon generator is associated with a huge path length, limited only by the decoherence processes. An analytical estimation of the sensitivity to loss caused by analyzed particles (molecules, nanoparticles, etc.) normalized by the single pass plasmon scheme is derived. Usage of graphene nanoflakes as plasmonic cavity allows a high spatial resolution to be reached, in addition to high sensitivity.

Keywords: Graphene; Plasmonics; Spaser; Spectroscopy

1. Introduction

Methods of intracavity laser spectroscopy [1–4] are widely used in chemical, biological and physical research [5–8]. The idea of these methods is the study of sample objects (atoms, molecules, clusters) directly in the laser cavity, which leads to one of the most sensitive laser spectroscopy methods – intracavity laser absorption spectroscopy [9–11].

With the development of plasmonics [12–21], a variety of spectroscopy methods have been suggested [22–24]. After the invention of plasmonic generators (spasers) [25–29], intracavity *spaser* spectroscopy has become feasible [30,31]. In such systems based on plasmons of noble metals, spaser sensitivity enhancement is essentially lower than sensitivity enhancement in conventional lasers. This is due to high plasmon absorption, which causes a noise in the cavity [31,32]. It is the cavity noise that limits the sensitivity of both the laser [33] and spaser [31] intracavity spectroscopy methods, which is maximal near the lasing threshold.

A significant increase in the sensitivity of spaser spectroscopy can be expected in systems that support low-loss plasmons. One of the most promising systems in this

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* Corresponding author.
E-mail address: alexandor7@gmail.com (A.V. Dorofeenko).

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respect is graphene [34–39]. Graphene is an extremely thin 2D material [40,41] which has high carrier mobility [42]. This material supports plasmons [34,37,43–48], and control of electron Fermi level achievable by doping, for example, by the use of a gate electrode [49], makes it applicable from optical to THz region. The latter frequency region is of high interest as it contains vibrational transitions of molecules. The use of graphene opens up opportunities to create extremely fast and compact optical and THz devices [50,51]. In near-IR graphene plasmons, the localization factor reaches higher values while losses are lower than in metal plasmons [44].

On the one hand, the high plasmon localization is very good for sensing. On the other hand, localization means extremely large values of the wave numbers and, thus, difficulties with the excitation of such plasmons by the incident wave. As a result, most of the proposed graphene plasmon sensors use hybrid systems including graphene and metal [52,53]. In such systems, a plasmon has lower wave numbers than in a pure graphene and can be excited by the Kretschmann method. This problem can be resolved by direct plasmon excitation with a gain medium. Such systems belong to active graphene plasmonics [54]. The most pronounced device in this field is graphene plasmon generator (graphene spaser), which has recently been proposed and investigated [55–60].

Let us note that the constraints arising in spaser realization were repeatedly discussed in the literature. In the case of nano-sized metal spasers with semiconductor gain, one needs enormous pumping current [61]. In the case of CW optical pumping, extreme heating of metallic nanoparticle should appear [62]. However, this difficulty was overcome by using pulsed optical pumping or distributed plasmonic geometry. This has lead to numerous spaser realizations [63–67]. A graphene spaser has even lower losses, which reduces the aforementioned constraints.

In this paper, we propose a novel graphene intracavity spaser absorption spectroscopy (GICSAS) method based on graphene spaser. A detailed analysis of the passive cavity and gain contributions in such a scheme is performed. It is shown that the sensitivity enhancement depends on the plasmon cavity quality factor as $Q^{3/2}$. It includes the $\sim Q$ contribution of passive cavity and $\sim\sqrt{Q}$ contribution of surface plasmon generation effect.

2. Geometry and model

Consider a nanoflake of graphene near the amplifying medium (quantum dots) with the population inversion induced by incoherent pumping (Fig. 1) as a model system illustrating the operation of the GICSAS method. The active medium excites the plasmonic mode, and this process dominates over the far-field radiation due to strong plasmon localization [68,69].

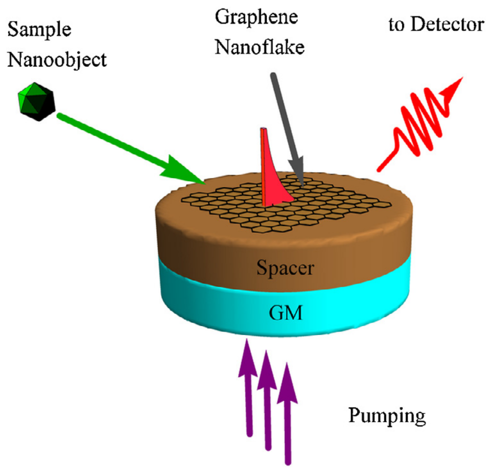

Fig. 1. Schematic picture of graphene spaser based sensor.

A graphene nanoflake-based spaser was considered in detail in Ref. [57], where a comprehensive modeling of the graphene nanoflake spaser with the account of a set of plasmonic modes has been carried out. This modeling has shown a single-mode spasing regime in a 50 nm graphene nanoflake. Therefore, we confine ourselves by a single-mode model. This model should take spontaneous emission noise into account, because it is crucial for the estimation of the maximal method sensitivity, as will be shown below. To describe generation, we implement a simple model that is conventionally used in the case of intracavity laser spectroscopy [70]. This model takes into account spontaneous emission [71] by considering the field/matter interaction to be proportional to $n+1$:

$$\dot{n} + \gamma n = \Omega(n+1)D, \tag{1}$$

$$\dot{D} + (D - D_0)/T_1 = -\Omega(n+1)D. \tag{2}$$

Here n is the plasmon number, D is the gain medium population inversion, D_0 is the pumping parameter of the gain medium, $\gamma = 1.56 \times 10^{12}\ \mathrm{s}^{-1}$ is the plasmon mode damping factor, $T_1 = 10^{-11}$ s is the longitudinal relaxation time of the gain medium and $\Omega = 5.3 \times 10^9\ \mathrm{s}^{-1}$ is the interaction constant of the gain medium with plasmonic mode, which is controlled by the gain medium parameters and system geometry (see paper [72] for the relation of the model parameters to the physical ones).