Intrinsic Terahertz Plasmons and Magnetoplasmons in Large Scale Monolayer Graphene

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Abstract

We show that in graphene epitaxially grown on SiC the Drude absorption is transformed into a strong terahertz plasmonic peak due to natural nanoscale inhomogeneities, such as substrate terraces and wrinkles. The excitation of the plasmon modifies dramatically the magneto-optical response and in particular the Faraday rotation. This makes graphene a unique playground for plasmon-controlled magneto-optical phenomena thanks to a cyclotron mass 2 orders of magnitude smaller than in conventional plasmonic materials such as noble metals.

Reference

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Intrinsic Terahertz Plasmons and Magnetoplasmons in Large Scale Monolayer Graphene

I. Crassee,*,† M. Orlita,‡⊥ M. Potemski,‡ A. L. Walter,#,▽ M. Ostler,§ Th. Seyller,§ I. Gaponenko,† J. Chen,∥¶ and A. B. Kuzmenko†

†Département de Physique de la Matière Condensée, Université de Genève, 1211 Genève, Switzerland
‡Grenoble High Magnetic Field Laboratory, CNRS-UJF-UPS-INSA F-38042 Grenoble Cedex 09, France
§Lehrstuhl für Technische Physik, Universität Erlangen-Nürnberg, 91058 Erlangen, Germany
⊥Faculty of Mathematics and Physics, Charles University, Ke Karlovu 5, 121 16 Praha 2, Czech Republic
▽Centro de Fisica de Materiales (CSIC-UPV/EHU) and Donostia International Physics Center (DIPC), 20018 Donostia-San Sebastian, Spain
∥E.O. Lawrence Berkeley Laboratory, Advanced Light Source, MS6-2100, Berkeley, California 94720, United States
¶Centro de Fisica de Materiales (CSIC-UPV/EHU) and Donostia International Physics Center (DIPC), 20018 Donostia-San Sebastian, Spain
#E.O. Lawrence Berkeley Laboratory, Advanced Light Source, MS6-2100, Berkeley, California 94720, United States
▽Department of Molecular Physics, Fritz-Haber-Institut der Max-Planck-Gesellschaft, Faradayweg 4-6, 14195 Berlin, Germany

Supporting Information

ABSTRACT: We show that in graphene epitaxially grown on SiC the Drude absorption is transformed into a strong terahertz plasmonic peak due to natural nanoscale inhomogeneities, such as substrate terraces and wrinkles. The excitation of the plasmon modes dramatically the magneto-optical response and in particular the Faraday rotation. This makes graphene a unique playground for plasmon-controlled magneto-optical phenomena thanks to a cyclotron mass 2 orders of magnitude smaller than in conventional plasmonic materials such as noble metals.

KEYWORDS: Graphene, terahertz, magneto-optics, magnetoplasmons, Faraday rotation

Graphene attracts a lot of attention as a novel optoelectronic and plasmonic material for applications ranging from the terahertz to the visible.1–14 One expects that plasmon waves in graphene can be squeezed into a much smaller volume1,11 than in noble metals routinely used in plasmonics and, importantly, can be manipulated by external gate voltage. A Dirac-like linear electronic dispersion and zero bandgap in graphene make its electromagnetic response rather unusual compared to other known two-dimensional conductors, such as 2D electron gases (2DEGs) in semiconductor heterostructures,15 even though the basic description of the propagating plasma modes is essentially the same.1,2

In order to observe plasmonic absorption optically one generally has to break the translational invariance of the system. Typical ways to couple plasmons to electromagnetic radiation in two-dimensional systems are placing an external grid in the vicinity of the sample16 and making stripe- or dotlike periodic structures inside the system.17–19 In graphene, this coupling has recently been achieved by patterning it in the shape of ribbons10 and by using a metallic atomic force microscopy (AFM) tip in scattering-type scanning near field optical microscopy.12 However, no measurement of graphene plasmons in magnetic field was reported so far.

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was one graphene layer and that the backside of the SiC substrate was graphene free. After hydrogenation, graphene becomes strongly p-doped; the Fermi level, \( \varepsilon_F = -0.34 \) eV, is below the Dirac point, which corresponds to a hole concentration \( n = \varepsilon_F/(\pi \hbar v_F^2) \approx 8 \times 10^{12} \text{ cm}^{-2} \). All magneto-optical measurements were done in the Faraday geometry (magnetic field and propagation of light normal to the sample) using a Fourier transform infrared spectrometer connected to a split-coil superconducting magnet. The illuminated area of several square millimeters was fully covered by graphene.

Figures 1a,b shows the optical transmission and Faraday rotation spectra measured at 5 K in fields \( B \) up to 7 T. The diagonal conductivity \( \sigma_{xx}(\omega) \), normalized to the universal conductivity \( \sigma_0 = e^2/4\hbar^2 \), shown in Figure 1c is obtained from the optical transmission, as described in the Supporting Information. At zero magnetic field, we observe a strong maximum centered around 6.5 meV (1.6 THz) instead of the normally expected Drude peak at zero frequency due to free carriers. As we demonstrate below, the deviation from the Drude behavior is associated with the presence of a confinement potential acting on free carriers and the corresponding plasmonic absorption. A similar resonance was observed recently in graphene microribbons in the polarization perpendicular to the ribbons.

When a magnetic field is applied, the plasma resonance splits into two modes, one of which increases and the other decreases with \( B \) (Figure 1c). In order to get further insight into the origin of these modes we extracted the Hall conductivity \( \sigma_{xy}(\omega) \) from the Faraday rotation spectra (Figure 1d) and obtained the optical conductivity for left and right circularly polarized radiation, \( \sigma_{\pm}(\omega) = \sigma_{xx} \pm i \sigma_{xy} \) (Figure 1e,f), as described in the Supporting Information. One can clearly see that each of the modes is excited only in one circular polarization. The peak positions in \( \sigma_{\pm}(\omega) \) and \( \sigma_{xx}(\omega) \) we denote \( \omega_+ \) and \( \omega_- \) respectively, since the former increases and the latter decreases with magnetic field. The inset of Figure 1f shows the field dependence of \( \omega_+ \) and \( \omega_- \). The field-induced splitting of the plasmon peak resembles strikingly the appearance of collective resonances observed previously in disk-shaped quantum dots of two-dimensional electron gases based on GaAs heterostructures and in bound 2D electrons on the surface of liquid helium. In both cases, the upper and lower branches were attributed to the so-called bulk and edge magnetoplasmons, respectively, with the frequencies

\[
\omega_{\pm} = \frac{\omega_0^2}{4} \pm \frac{|\omega_0|}{2}
\]

where \( \omega_0 \) is the plasmon frequency at zero field, \( \omega_0 = \pm eB/mc \) is the cyclotron frequency, defined as positive for electrons and negative for holes, \( m \) is the cyclotron mass, and \( c \) is the speed of light. At high fields \( (|\omega_0| \gg \omega_0) \), the upper branch becomes essentially the usual cyclotron resonance with a linear dependence on magnetic field, while the lower branch represents a collective mode confined to the edges of a graphene layer, as determined by Raman spectroscopy.

In order to clarify the origin of the confinement that causes the plasmonic resonance, we performed vibrating cantilever AFM imaging, allowing us to extract topographic and phase information. In Figure 2a, a topographical height image of a 10 \( \times \) 10 \( \mu m^2 \) area of the sample is presented. The dominating structures are the terraces due to the miscut angle of SiC. Their irregular shape as compared to morphologies observed earlier is related to the specific graphitization temperature used in our work. Importantly, the terraces are oriented in the same direction across the entire sample. Figure 2b presents the map of the oscillation phase of the cantilever on the same area. The dark spots in the phase correspond to regions without graphene, as was determined by Raman spectroscopy. Closer inspection of the AFM images also reveals numerous wrinkles such as the ones indicated by the arrows in Figure 2c,d. The wrinkles are formed due to the relaxation of strain in graphene during the cooling down after the graphitization. Figure 2e shows height profiles for the lines marked in Figure 2a. Profiles 1 to 3 correspond to steps in the SiC substrate, while traces 4 to
plasmon structure and its splitting in magnetic Lorentz formula for the optical conductivity. Doping levels makes this interpretation less straightforward. Silicon face of SiC, the presence of many layers with different polarizations.

Polarized optical transmission (Figure 2f) provides a hint that the terahertz resonance peak is related to the morphological structures seen by AFM. A significant anisotropy is found, which correlates with the orientation of the terraces. In particular, the absorption maximum is at about 1.7 times higher energy for the electric field perpendicular to the terraces than for the parallel orientation. Note that the peak position remains at finite energy for every polarization. The excitation of the plasmon parallel to the terraces is likely due to the rough shape of the SiC step edges. One cannot exclude that the wrinkles, which are randomly oriented, might also play a role in confining the carriers. Because of the anisotropy, the curves and the plasmon energy in Figure 1 are effective averages over all polarizations.

It is worthwhile to notice that the terahertz transmission of multilayer graphene epitaxially grown on the carbon side of SiC also reveals an absorption peak, although at a somewhat lower energy, with a strong polarization dependence as shown in the Supporting Information. Although it might have a similar energy, with a strong polarization dependence as shown in the Supporting Information. Although it might have a similar energy, with a strong polarization dependence as shown in the Supporting Information. Although it might have a similar energy, with a strong polarization dependence as shown in the Supporting Information. Although it might have a similar energy, with a strong polarization dependence as shown in the Supporting Information. Although it might have a similar energy, with a strong polarization dependence as shown in the Supporting Information. Although it might have a similar energy, with a strong polarization dependence as shown in the Supporting Information. Although it might have a similar energy, with a strong polarization dependence as shown in the Supporting Information. Although it might have a similar energy, with a strong polarization dependence as shown in the Supporting Information.
than the expected value of $2|e|\ell = 0.68$ eV. The difference might be related to a noncomplete coverage of the substrate by graphene (see Figure 2b), however can also be due to the presence of the background $n_0$. The broadening, $h\gamma'$ of the peak is about 10–12 meV, which is more than twice smaller than observed in a similar sample by another group.\(^3\) It might be larger than the intrinsic electron scattering, since the spectral feature is additionally broadened by the distribution of sizes and shapes of the homogeneous regions. A more detailed discussion of the spectral weight, background, and scattering is given in the Supporting Information.

The Dirac-like charge carriers in graphene at high doping are expected to show a classical cyclotron resonance with a linear field dependence,\(^2\) which perfectly agrees with our data. The cyclotron mass depends on doping according to the relation $m = |e|\ell / \sqrt{\epsilon_0}$. Using $|e|\ell = 0.34$ eV, and $m = 0.055 m_0$, we find that $v_F = 1.04 \times 10^6 \text{m/s}$, which matches remarkably well the Fermi velocity obtained by other methods.\(^3\)

The plasmon frequency, $h\omega_0 = 6.5$ meV, contains important information about the intrinsic properties of the electron gas and the confinement causing the plasmon excitation. It is instructive to compare the value of $\omega_0$ found in this work with the resonant frequency for the reference case of isolated disk-shaped quantum dots, for which the effective medium model predicts:\(^17,35,32\)

$$\omega_0^2 = \frac{3\pi^2 n e^2}{2mdk}$$

where $k = (1 + \epsilon_{\text{SiC}})/2 \approx 5$ is the average dielectric constant of the surrounding media and $d$ is the dot diameter. For $d = 1 \mu$m, which roughly corresponds to the mean size of homogeneous regions in our sample, and for the same charge density $n = 8 \times 10^{12} \text{cm}^{-2}$, the expected plasmon frequency would be 15.2 meV. This is more than twice the experimentally observed value. Lacking a rigorous quantitative model for the present sample, we ascribe this difference to the fact that the defect lines that separate homogeneous graphene regions are very narrow, and that the electromagnetic coupling between neighboring regions therefore plays an important role. Such a coupling causes a redshift\(^32,5\) of the plasmon energy as compared to the case of noncoupled particles, which is commonly observed in plasmonic nanostructures.

Important for terahertz applications is the question to what extent the presence of a plasmon affects the Faraday rotation in graphene. The simulations in Figure 4a demonstrate that the Faraday angle is at maximum close to the magnetoplasmon resonance and therefore can be controlled not only by magnetic field but also by $\omega_0$. Here we compare the calculated Faraday rotation of homogeneous graphene ($\omega_0 = 0$) and the rotation in the presence of plasmons such as the one observed in our sample (6.5 meV) and for higher plasmon energies (20 and 40 meV). A way to increase the plasmon frequency is to decrease the size of the homogeneous regions, which could be done, for example, by varying the miscut angle of the substrate. Notice that a route to increase the Faraday rotation for a given plasmon frequency is to use samples with reduced electronic scattering as demonstrated in Figure 4b. Rotations above 0.1 radians by just one atomic layer at a modest field of 1 T do not seem to be out of experimental reach.

Similarly, in more conventional plasmonic materials the Faraday and Kerr angles are enhanced close to the plasma resonance.\(^24,39\) For example, in a recent work a Kerr rotation on the order of $10^{-4}$ to $10^{-5}$ radians was detected in an array of Au disks.\(^24\) Thus, graphene shows more than 2 orders of magnitude larger rotation, which is a direct consequence of a much smaller cyclotron mass even though the carrier density per unit cell is also much lower than in noble metals. The measurements presented in this Letter were performed at low temperature to maximally resolve the magnetoplasmonic spectral structures. We do believe, though, that the magnetoplasmonic phenomena described here will persist up to room temperature, based on our temperature dependent experimental study of the cyclotron resonance in epitaxial graphene on the carbon face of SiC.\(^40\)

In conclusion, we found that morphological defects on the nanoscale such as atomic steps in SiC and wrinkles in epitaxial graphene produce a remarkably strong plasmon resonance. This resonance has essentially the same origin as the plasmon peak observed in two-dimensional electron gases and in nanostructured graphene. The important difference, however, is that the confinement potential in epitaxial graphene is natural and does not require special lithographic patterning, which risks reducing the carrier mobility. Instead, one can think of controlling the plasmon frequency by varying the preparation of the substrate and the graphitization process. The presence of the plasmon dramatically changes the cyclotron resonance and Faraday rotation. Graphene appears to be a unique material, where one finds simultaneously a small effective mass giving rise to strong magneto-optical effects and excellent plasmonic

**Figure 3.** The magnetic field dependence of the plasmon energy $h\omega_0$, cyclotron resonance energy $h\omega_c$, and magnetoplasmon energies $h\omega_{mp}$. The dotted line is a linear fit to the cyclotron energy.

**Figure 4.** A simulation of the effect of the plasmon energy and the scattering on the Faraday rotation. (a) Faraday rotation for $h\gamma = 11$ meV for different values of $h\omega_0 = 0, 6.5, 20, \text{and } 40$ meV. (b) Faraday rotation for $h\omega_0 = 20$ meV and different values of $h\gamma = 11$ and 5.5 meV. In both cases the magnetic field is 1 T.
properties. This combination opens pathways toward plasmon-controlled terahertz magneto-optics.

** ASSOCIATED CONTENT **

* Supporting Information

Details of the extraction of the magneto-optical conductivity, effective medium approach, magnetic field dependence of some fitting parameters, and terahertz transmission spectra of multilayer graphene at the C-side. This material is available free of charge via the Internet at http://pubs.acs.org.

** AUTHOR INFORMATION **

Corresponding Author

*E-mail: Iris.Crassee@unige.ch.

Notes

The authors declare no competing financial interest.

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