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Tunable terahertz plasmons in graphite thin films

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21 **Tunable terahertz plasmons are essential for reconfigurable photonics,**
22 **which have been demonstrated in graphene through gating, though with**
23 **relatively weak responses. Here, we demonstrate strong terahertz plasmons in**
24 **graphite thin films via infrared spectroscopy, with dramatic tunability by even a**
25 **moderate temperature change or an in-situ bias voltage. Meanwhile, through**
26 **magneto-plasmon studies, we reveal that massive electrons and massless Dirac**
27 **holes make comparable contributions to the plasmon response. Our study not**
28 **only sets up a platform for further exploration of two-component plasmas, but**
29 **also opens an avenue for terahertz modulation through electrical bias or**
30 **all-optical means.**

31

32 Tunable terahertz photonic devices are indispensable in terahertz applications in
33 sensing, imaging, waveguiding etc. [1-5]. Various tunable plasmonic materials
34 operating in the terahertz range have been explored, including graphene [6, 7],
35 phase-changing compounds [8-11] and carbon nanotubes [12-14]. In particular, with
36 good gate tunability, graphene is very promising for terahertz plasmonic applications
37 [15, 16]. However, due to relatively weak response to light [17], graphene has to be
38 combined with metallic structures to realized feasible modulation [18-24]. An
39 extension to graphite thin films beyond monolayer is a natural avenue to achieve
40 stronger and more intrinsic plasmonic response. Though the gate electrical field is
41 screened in graphite thin films, the thermal carrier density depends strongly on
42 temperature [25, 26], which promises sensitive tuning of plasmons by temperature.

43 Compared to phase-changing compounds, which typically exhibit switchable
44 plasmons around the critical temperature [9], the plasmons in graphite are expected to
45 be tuned continuously in a broad temperature range. Thermo-tuning of plasmons, in
46 particular with ultrafast lasers to excite carriers [27-29], promises applications in
47 all-optical modulation and tunable plasmonic metamaterials.

48 Moreover, graphite is a semimetal where massive electrons and massless Dirac
49 holes coexist, residing around K-point and H-point of the Brillouin zone, respectively
50 [30-32], and forming a two-component plasma [33-35]. Depending on their relative
51 oscillation phase, the collective oscillations of electrons and holes can be categorized
52 into optical and acoustic modes [34, 36]. The interrogation of plasmons in graphite
53 thin films can possibly gain insight into the many-body interaction in the
54 two-component plasma, which involves both regular and massless Dirac fermions.

55 In our work, we perform a systematic study of the graphite plasmon in the
56 terahertz regime using far-field infrared spectroscopy. Strong temperature and bias
57 voltage dependence of the plasmon has been revealed. The magnetic field effect on
58 the plasmon differentiates the contributions of massive and massless fermions to the
59 collective oscillation and comparable Drude weights from both components are
60 inferred.

61

62 A schematic of the far-infrared transmission measurement is shown in Fig. 1(a).
63 Extinction spectra $1-T/T_0$ characterizes the electromagnetic responses of graphite thin
64 films or microstructure arrays. A typical exfoliated graphite thin film on Si substrate is

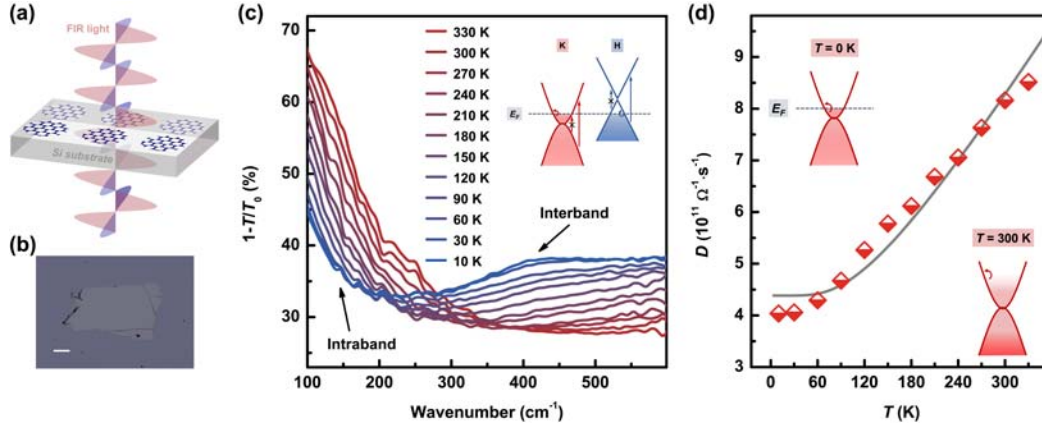
65 displayed in Fig. 1(b). Details of the fabrication and measurement procedures are
 66 given in Supplemental Material [37]. The extinction spectra of a typical unpatterned
 67 graphite film with the thickness of ~ 20 nm are shown in Fig. 1(c). At liquid helium
 68 cryogenic temperatures, the free carrier Drude response is still quite pronounced even
 69 without thermal excitation, and a step-like feature, indicating the onset of interband
 70 transitions, is observed around 400 cm^{-1} . These behaviors are consistent with the band
 71 structure of graphite [31], in which the hole pocket with a linear dispersion near the H
 72 point coexists with an electron pocket with a parabolic dispersion near K point [30],
 73 and the Fermi level determines the onset of interband transitions, as illustrated in the
 74 inset of Fig. 1(c).

75 Due to the semimetal nature and the Fermi energy in a close proximity of the
 76 band touching in graphite, the Drude response and interband excitations are sensitive
 77 to temperature. Thermal excitation can efficiently increase the free carrier density,
 78 resulting in an enhancement of the Drude response and suppression of interband
 79 transitions due to Pauli blocking. Such blocking prohibits the optical transitions from
 80 empty states in the valence band or into filling states in the conduction band, which
 81 have been created by thermal excitation. We plot the extracted Drude weight (see
 82 Supplementary Material [37] for details) as a function of temperature in Fig. 1(d)
 83 which is consistent with the prediction for 2D electron gas with touched valence and
 84 conduction bands:

$$D = Ak_B T \ln \left(2 \cosh \left(\frac{\mu}{2k_B T} \right) \right) \quad (1)$$

85 where D is the Drude weight, A is a fitting coefficient, T is the temperature, k_B is

86 Boltzmann constant, and μ is the chemical potential. For simplicity, we treat
 87 $\mu = E_F(T = 0 \text{ K})$ as a temperature-independent parameter, which is an accurate
 88 exercise for parabolic bands (see Supplementary Material [37] for details) [40, 41].
 89 The fitting value of the chemical potential is 21 meV, in good agreement with the
 90 literature [31].



91
 92 FIG. 1. Characterization of the exfoliated graphite thin film. (a) An illustration of the
 93 far-infrared spectroscopy scheme. (b) A typical optical image of the exfoliated
 94 graphite thin film on Si substrate, scale bar 100 μm . (c) Temperature-dependent
 95 far-infrared spectra of a graphite film with thickness of $\sim 20 \text{ nm}$. The inset shows the
 96 electron (K point) and hole (H point) pocket in the Brillouin zone at zero temperature.
 97 The arrows (with a cross) indicate intraband and (Pauli-blocked) interband transitions,
 98 and the gray dashed line represents the Fermi energy. (d) Drude weight in (c) as a
 99 function of temperature. The curve is the fitting based on Eq. (1). The inserted
 100 sketches illustrate the carrier distributions at zero and room temperature, and the
 101 arrows represent the intraband transitions.

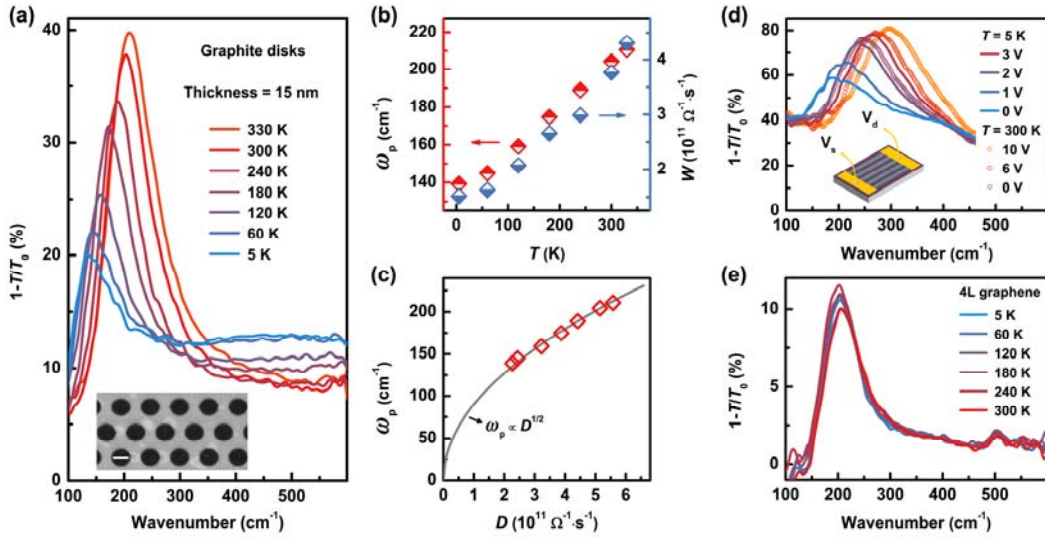
102

103 The temperature dependence of the Drude weight for graphite films is inherited
104 by plasmons. We patterned a continuous thin film with thickness of 15 nm into an
105 array of disks with diameter of 2.3 μm , as shown by the scanning electron microscope
106 (SEM) image in the inset of Fig. 2(a). With the temperature increasing from 5 to 330
107 K, the plasmon frequency increases 60%, and the intensity increases to 2 times of the
108 original accordingly, as demonstrated in Fig. 2(a). The extinction of the plasmon in
109 graphite films is generally much larger than that in graphene, and the linewidth is
110 comparable (see Supplementary Material [37]), which are desirable for real
111 applications. In addition, the field confinement factor is ~ 15 at 5 K, much better than
112 that even for the thinnest noble metal structures in IR frequencies [42]. Figure 2(b)
113 shows the plasmon frequency and spectrum weight (procedures to extract the weight
114 are in Supplementary Material [37]) as functions of temperature. In the low frequency
115 (long wavelength) regime, the two-dimensional (2D) plasmon frequency scales as
116 \sqrt{D} , where D is the Drude weight [6, 15, 43]. Such a scaling works very well for the
117 plasmon in the graphite thin film, as shown in Fig. 2(c), where we plot the plasmon
118 frequency in Fig. 2(b) as a function of Drude weight D at each temperature obtained
119 from a graphite film with a similar thickness.

120 The temperature dependence is so sensitive that even the Joule heating of an
121 electrical device based on the graphite thin film can largely in-situ modulate the
122 plasmonic response. As demonstrated in Fig. 2(d), a current passing through a
123 graphite ribbon array induces a pronounced shift of the plasmon frequency and an
124 enhancement of the intensity, with the device both at low and room temperature

125 environments. This promises reconfigurable metasurfaces through in-situ bias-tuning.

126 In addition to the graphite thin film, for comparison, we also performed a
 127 temperature dependent measurement of a 4-layer graphene disk array on SiO₂/Si
 128 substrate (mid-IR spectrum of the sample is in Supplementary Material [37]). As
 129 shown in Fig. 2(e), the plasmon frequency and intensity are almost independent of the
 130 temperature, since few-layer graphene tends to be doped to a high Fermi level
 131 (equivalent to thousands of Kelvin) by the surrounding media, and according to Eq.
 132 (1), the Drude weight is almost a constant within our temperature range. The
 133 temperature insensitivity of highly doped graphene is in marked contrast to that of
 134 graphite thin films with a much lower Fermi level.



135
 136 FIG. 2. The temperature-dependent plasmon. (a) Temperature-dependent far-infrared
 137 spectra of a graphite disk array with the diameter of 2.3 μm and the period of 3.75 μm .
 138 The inset shows the SEM image, scale bar 2 μm . The graphite film thickness is ~ 15
 139 nm. (b) Plasmon frequency and weight W extracted from (a) as functions of
 140 temperature T . (c) Plasmon frequency as a function of the Drude weight D extracted

141 from a graphite film with a similar thickness as that of the disk array. (d) Far-infrared
142 spectra of a biased graphite ribbon array with the ribbon width of 4 μm and thickness
143 of ~ 20 nm at 5 K and room temperature. The inset is an illustration of a typical device.
144 (e) Temperature-dependent far-infrared spectra of a 4-layer graphene disk array with
145 disk diameter of 1 μm .

146
147 By shrinking the structure size, the plasmon can be pushed to higher frequencies.
148 A series of graphite micro-rectangle arrays from a large graphite film with thickness
149 of 15 nm was fabricated. The inset of Fig. 3(a) shows the typical morphology of the
150 graphite rectangle array. As shown in Fig. 3(a), with the decreasing of the side length
151 from 3 to 0.5 μm , the plasmon blueshifts, and its intensity decreases. At the same time,
152 the fitted linewidth of the plasmon gradually increases from 90 cm^{-1} to 203 cm^{-1} , as
153 displayed in Fig. 3(b). The plasmon broadening and intensity reduction are
154 compelling evidences of the increasing Landau damping, since the interband
155 transition channels start to dominate beyond ~ 400 cm^{-1} , making the annihilation of
156 higher energy plasmons into electron-hole pairs efficient. Nevertheless, as a two
157 dimensional film, the plasmon dispersion still follows the standard $\omega_p \propto \sqrt{q}$ scaling
158 law [43], as verified in Fig. 3(c), where $q = \pi/L$ (L is the rectangle side length) is
159 the plasmon wave vector, without considering the plasmon phase shift at boundaries
160 for simplicity [45]. The plasmon dispersion, broadening and intensity can also be
161 confirmed by the loss function [46, 47] $-\text{Im}(1/\epsilon)$, as displayed as a pseudocolor
162 map in Fig. 3(c). The calculation details are presented in the Supplementary Material

[37]. As we can see from the map, the plasmon broadens with increasing frequency and beyond certain frequency (wave vector), the plasmon peak is almost completely smeared out. Such cut-off frequency depends on the temperature and higher temperature can sustain higher cut-off frequency due to the reduced interband Landau damping originated from the band-filling effect [26, 50], as exemplified by the simulation of a graphite rectangle array and the response of graphite split rings in Supplementary Material [37].

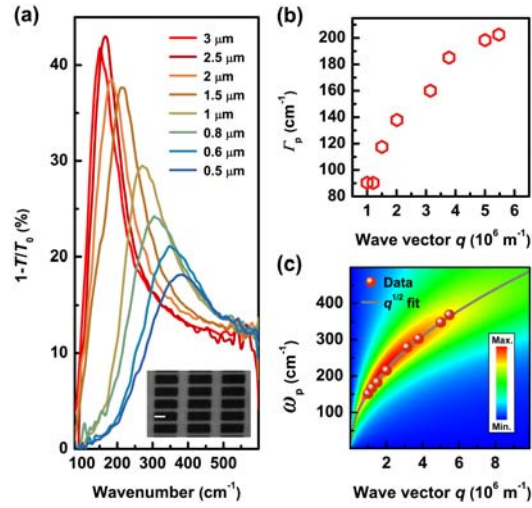


FIG. 3. Plasmon dispersion. (a) Extinction spectra of rectangle arrays with decreasing side length. The thickness of the graphite film is about 15 nm. The inset shows an SEM image of a typical rectangle array with a scale bar of 1 μm . (b) Plasmon linewidth as a function of the wave vector q . (c) Plasmon frequency in (a) as a function of the wave vector. The grey curve is a $q^{1/2}$ scaling. The loss function is also shown as a background.

The response of graphite plasmon to an external magnetic field is of potential interest due to the small cyclotron mass [54]. Moreover, owing to very different

180 nature of carriers in the H- and K-pockets, the free carriers in graphite can be treated
 181 as a two-component plasma and a magnetic field can quantify their contributions to
 182 the collective excitation. In a disk with a single component carrier, the
 183 magneto-plasmon resonances have two modes, the bulk mode ω^+ and the edge mode
 184 ω^- [55]:

$$185 \quad \omega^\pm = \sqrt{\omega_0^2 + \left(\frac{\omega_c}{2}\right)^2} \pm \frac{\omega_c}{2} \quad (2)$$

186 where ω_0 is the plasmon frequency at zero magnetic field, ω_c is the cyclotron
 187 frequency. For massive carriers, $\omega_c = eB/m_c$, with m_c as the cyclotron mass. In
 188 graphite disks, since Dirac holes at H-point exhibit much larger cyclotron frequency
 189 than massive electrons at K-point, based on Eq. (2), the originally coupled
 190 electron-hole plasma will be decoupled even at a moderate magnetic field. Therefore,
 191 we expect to see magneto-plasmon modes contributed solely by one of the carrier
 192 components.

193 We performed magneto-transmission measurements with the magnetic field
 194 perpendicular to the graphite disk array (Faraday configuration). The extinction
 195 spectrum without an external magnetic field is shown in Fig. 4(a), with a peak
 196 frequency of $\sim 135 \text{ cm}^{-1}$ at 10 K. The measured relative transmission spectra with the
 197 magnetic field ranging from 0 T to 17.5 T are presented in Fig. 4(b). The sample was
 198 always at the liquid-helium temperature, so the thermal excitation is minimal. The
 199 possible excitonic effect can be ignored due to the only moderate magnetic field in
 200 our study [56]. With the increase of the field, the frequency and magnitude of the dip
 201 indicated by the orange arrow dramatically increase. Other weaker dips are higher

202 energy Landau level transitions of electrons, as detailed in Supplementary Material
 203 [37]. The features of massless Dirac holes at H-point [57, 58], which typically have
 204 much higher frequencies and scale as \sqrt{B} , are not clearly observed. We plot the
 205 extracted frequency of the major dip as a function of the magnetic field (B not less
 206 than 4T) in Fig. 4(c) and fit it with Eq. (2). The fitted cyclotron mass $m_c \approx 0.039m_0$
 207 (m_0 is the free electron mass) is consistent with that in the literature for K-pocket
 208 electrons [58]. As expected, the fitted plasmon frequency $\omega_0 \approx 93 \text{ cm}^{-1}$ is smaller
 209 than the measured total frequency (135 cm^{-1}) of the two-component carriers at zero
 210 field. These behaviors clearly suggest that the electrons and holes are decoupled under
 211 B -field due to the unparallelled cyclotron frequency (See Fig. 4(c) for the cyclotron
 212 frequencies of holes and electrons, and the magneto-transmission spectra of an
 213 unpatterned graphite thin film shown in Supplementary Material [37].), and the
 214 observed magneto-plasmon mode is from a single component, i.e., electrons in the
 215 K-pocket. More quantitatively, the total Drude weight $D_{total} = D_e + D_h$, with D_e
 216 and D_h as Drude weights from electrons in K-pocket and holes in H-pocket at zero
 217 field, respectively. Because the plasmon frequency $\omega \propto \sqrt{D}$, we have $\omega_{total}^2 = \omega_e^2 +$
 218 ω_h^2 . With the aforementioned ω_{total} and $\omega_e = \omega_0$, we get $\omega_h \approx 97 \text{ cm}^{-1}$.
 219 Therefore, the Drude weights of electrons and holes in graphite are comparable,
 220 which is consistent with previous results [59, 60]. The lower branch ω^- of the split
 221 plasmon (edge mode [55]) in Eq. (2) is not observed in our experiment, presumably
 222 due to its resonance frequency beyond our lower measurement limit (about 80 cm^{-1})
 223 after both branches are well-split. Here we plot it based on Eq. (2) with the obtained

parameters, as shown by the orange dashed line in Fig. 4(c).

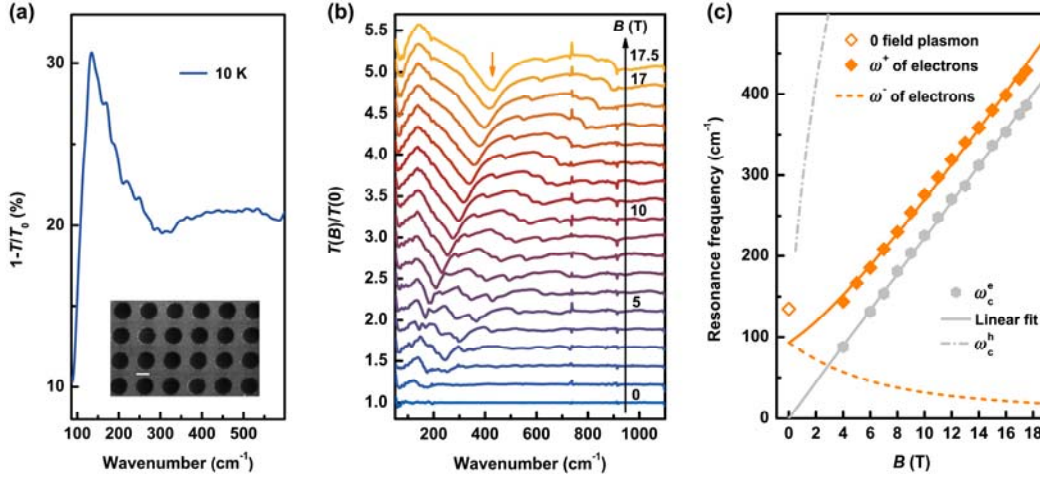


FIG. 4. Magneto-plasmon. (a) Extinction spectra of a disk array with the diameter of 3 μm at 10 K without a magnetic field. The inset shows the SEM image, scale bar 2 μm . The graphite film thickness is ~ 20 nm. (b) Relative transmission spectra $T(B)/T(0)$ in a Faraday configuration for the sample in (a). For clarity, spectra are shifted vertically. (c) Extracted frequency (orange data points) of the dip indicated by the orange arrow in (b) as a function of the magnetic field. The orange solid line is the fitting curve for the bulk mode based on Eq. (2), and the edge mode is also plotted as an orange dashed line. Zero field plasmon frequency determined in (a) is also shown for comparison. The light gray dashed line is the cyclotron frequency of holes in H-pocket, which is plotted based on the documented Fermi velocity of the Dirac cone [58]. The light gray data points are the cyclotron frequencies of electrons in K-pocket, extracted from the spectra in Supplementary Material [37]. The light gray solid line is the linear fitting.

In conclusion, we have systemically studied the tunable plasmon in graphite thin

241 films through infrared spectroscopy. In addition to strong optical response, graphite
 242 plasmon also manifests pronounced temperature and bias voltage dependence, which
 243 is in sharp contrast to the plasmon in graphene. The response of the plasmon mode in
 244 a magnetic field suggests that electrons and holes contribute similar Drude weights. In
 245 addition to the temperature and a magnetic field, plasmons in graphite thin films can
 246 be potentially tuned by intercalation as well [61], which may uncover rich physics and
 247 unleash huge potential for applications.

248

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