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1       **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

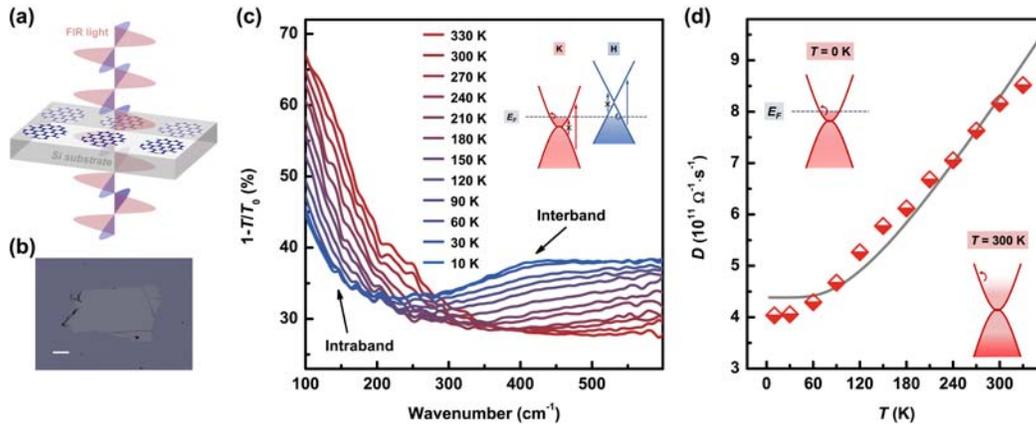
65 displayed in Fig. 1(b). Details of the fabrication and measurement procedures are  
66 given in Supplemental Material [37]. The extinction spectra of a typical unpatterned  
67 graphite film with the thickness of  $\sim 20$  nm are shown in Fig. 1(c). At liquid helium  
68 cryogenic temperatures, 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  $400 \text{ cm}^{-1}$ . 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  $\mu$  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  $\mu\text{m}$ . (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.

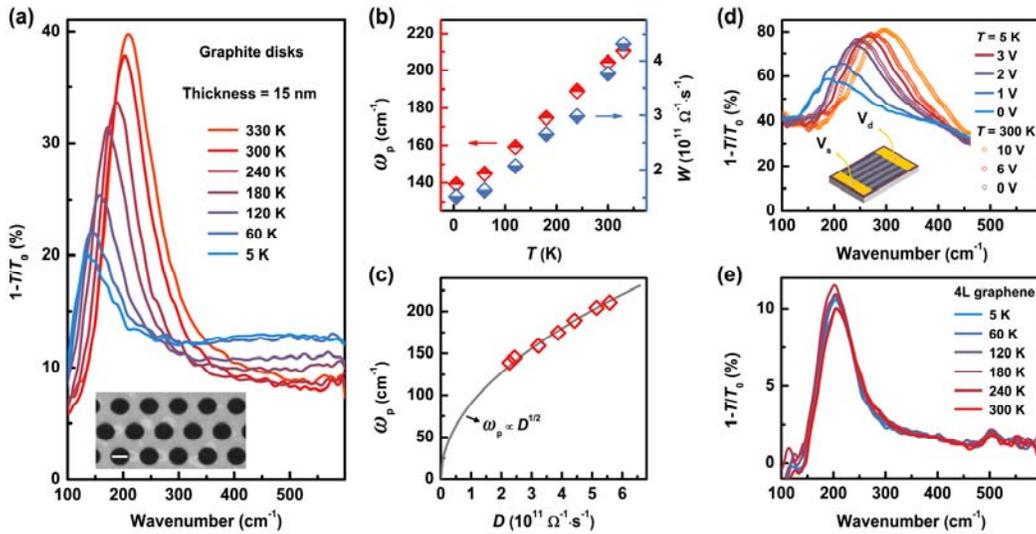
102

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  $\mu\text{m}$ , 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  $\sim 15$  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 SiO<sub>2</sub>/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.



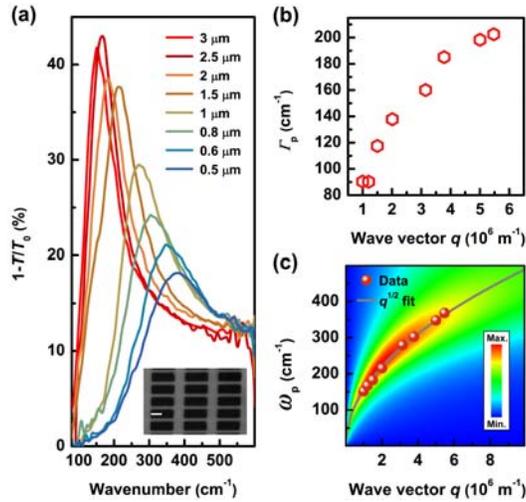
135  
 136 FIG. 2. The temperature-dependent plasmon. (a) Temperature-dependent far-infrared  
 137 spectra of a graphite disk array with the diameter of 2.3  $\mu\text{m}$  and the period of 3.75  $\mu\text{m}$ .  
 138 The inset shows the SEM image, scale bar 2  $\mu\text{m}$ . The graphite film thickness is  $\sim$ 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  $\mu\text{m}$  and thickness  
143 of  $\sim 20$  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  $\mu\text{m}$ .

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  $\mu\text{m}$ , the plasmon blueshifts, and its intensity decreases. At the same time,  
152 the fitted linewidth of the plasmon gradually increases from 90  $\text{cm}^{-1}$  to 203  $\text{cm}^{-1}$ , 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  $\sim 400$   $\text{cm}^{-1}$ , 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

163 [37]. As we can see from the map, the plasmon broadens with increasing frequency  
 164 and beyond certain frequency (wave vector), the plasmon peak is almost completely  
 165 smeared out. Such cut-off frequency depends on the temperature and higher  
 166 temperature can sustain higher cut-off frequency due to the reduced interband Landau  
 167 damping originated from the band-filling effect [26, 50], as exemplified by the  
 168 simulation of a graphite rectangle array and the response of graphite split rings in  
 169 Supplementary Material [37].



170  
 171 FIG. 3. Plasmon dispersion. (a) Extinction spectra of rectangle arrays with decreasing  
 172 side length. The thickness of the graphite film is about 15 nm. The inset shows an  
 173 SEM image of a typical rectangle array with a scale bar of 1  $\mu\text{m}$ . (b) Plasmon  
 174 linewidth as a function of the wave vector  $q$ . (c) Plasmon frequency in (a) as a  
 175 function of the wave vector. The grey curve is a  $q^{1/2}$  scaling. The loss function is also  
 176 shown as a background.

177

178 The response of graphite plasmon to an external magnetic field is of potential  
 179 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  $\omega^+$  and the edge mode  
184  $\omega^-$  [55]:

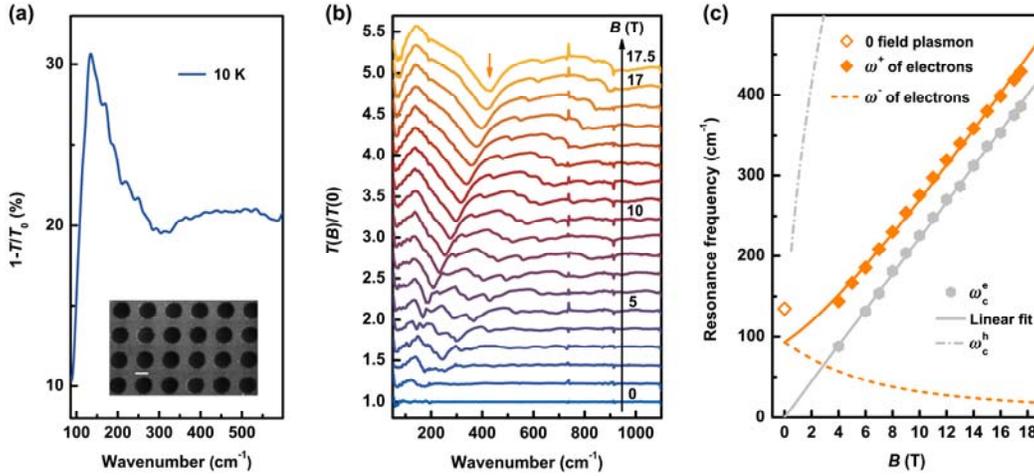
$$\omega^\pm = \sqrt{\omega_0^2 + \left(\frac{\omega_c}{2}\right)^2} \pm \frac{\omega_c}{2} \quad (2)$$

186 where  $\omega_0$  is the plasmon frequency at zero magnetic field,  $\omega_c$  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 ( $135 \text{ cm}^{-1}$ ) 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  $\omega_h^2$ . With the aforementioned  $\omega_{total}$  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  $\omega^-$  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  $80 \text{ cm}^{-1}$ )  
 223 after both branches are well-split. Here we plot it based on Eq. (2) with the obtained

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



225

226 FIG. 4. Magneto-plasmon. (a) Extinction spectra of a disk array with the diameter of 3  
 227  $\mu\text{m}$  at 10 K without a magnetic field. The inset shows the SEM image, scale bar 2  $\mu\text{m}$ .

228 The graphite film thickness is  $\sim 20$  nm. (b) Relative transmission spectra  $T(B)/T(0)$  in  
 229 a Faraday configuration for the sample in (a). For clarity, spectra are shifted vertically.

230 (c) Extracted frequency (orange data points) of the dip indicated by the orange arrow  
 231 in (b) as a function of the magnetic field. The orange solid line is the fitting curve for  
 232 the bulk mode based on Eq. (2), and the edge mode is also plotted as an orange

233 dashed line. Zero field plasmon frequency determined in (a) is also shown for  
 234 comparison. The light gray dashed line is the cyclotron frequency of holes in

235 H-pocket, which is plotted based on the documented Fermi velocity of the Dirac cone  
 236 [58]. The light gray data points are the cyclotron frequencies of electrons in K-pocket,

237 extracted from the spectra in Supplementary Material [37]. The light gray solid line is  
 238 the linear fitting.

239

240

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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