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# Anisotropic Acoustic Plasmons in Black Phosphorus

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**ABSTRACT**

Graphene separated a few nanometers away from a metal surface can support ‘acoustic plasmons’, which exhibit extreme plasmon confinement an order of magnitude higher than that of conventional graphene plasmons. Here, we investigate acoustic plasmons supported in a monolayer and multilayers of black phosphorus (BP) placed shortly above a conducting plate. In the presence of a conducting plate, the acoustic plasmon dispersion for the armchair direction is found to exhibit the characteristic linear scaling in the mid- and far-infrared regime while it largely deviates from that in the long-wavelength limit and near-infrared regime. For the zigzag direction, such scaling behavior is not evident due to relatively tighter plasmon confinement. Further, we demonstrate a new design for an acoustic plasmon resonator that exhibits higher plasmon confinement and resonance efficiency than BP ribbon resonators in the mid-infrared and longer wavelength regime. Theoretical framework and new resonator design studied here provide a practical route toward the experimental verification of the acoustic plasmons in BP and open up the possibility to develop novel plasmonic and optoelectronic devices that can leverage its strong in-plane anisotropy and thickness-dependent band gap.

**KEYWORDS.** black phosphorus, acoustic plasmon, gap plasmon, surface plasmon polaritons, anisotropy, two-dimensional material.

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3 Two dimensional (2D) materials<sup>1,2</sup> have attracted enormous interest due to their unique  
4 properties such as ultrahigh charge carrier mobility,<sup>3,4</sup> anomalous quantum Hall effect,<sup>5</sup> and  
5 strong light-matter interaction.<sup>6,7</sup> Among a variety of such exciting properties, strong light-matter  
6 interactions in 2D materials are particularly intriguing considering the extreme size mismatch  
7 between their atomic-scale thicknesses and wavelengths of free-space light,  $\lambda_0$ . Moreover, this  
8 feature plays a central role in many potential applications of 2D materials such as optical  
9 modulators,<sup>8,9</sup> metasurfaces,<sup>10-13</sup> biosensors,<sup>14,15</sup> and photodetectors.<sup>16,17</sup> For a particular set of 2D  
10 materials including graphene, light-matter interactions can be even more intense because of the  
11 excitation of surface plasmons.<sup>18-20</sup> Compared to conventional surface plasmons in noble metals,  
12 the plasmons in 2D materials exhibit tighter confinement ( $\sim\lambda_0/100$ )<sup>21-23</sup> as well as tunability by  
13 extrinsic doping.<sup>20,24</sup> Many researchers have demonstrated that such features allow for the  
14 development of nanoscale photonic and optoelectronic devices that have novel functionality and  
15 superior performance inaccessible with conventional materials.<sup>15,25,26</sup>

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33 Recent work on graphene indicates that the plasmon wavelength, and accordingly the  
34 confinement of 2D plasmons, can be further reduced in the presence of a conducting plate  
35 adjacent to the graphene.<sup>27-30</sup> As in the case of spatially separated double-layer graphene,<sup>31,32</sup> the  
36 hybridization of two plasmons in a graphene sheet and its mirror image leads to the formation of  
37 two plasmon branches: less confined ‘optical’ and highly confined ‘acoustic’ plasmons,  
38 depending on whether charges in two layers oscillate in-phase or out-of-phase. In the double-  
39 layer case with a gap much smaller than plasmon wavelength, the acoustic mode with out-of-  
40 phase charge oscillation becomes a dark mode due to the cancellation between dipole momenta  
41 in two layers, while the optical mode that has a net dipole momentum is optically active.<sup>33</sup> In the  
42 present case of a 2D layer on metal, however, the acoustic mode becomes optically active in the  
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3 absence of a second 2D layer to cancel the dipole momentum, while the optical mode is  
4 prohibited since it mandates the tangential electric fields to be non-zero at the surface of the  
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6 conducting plate. Due to the out-of-phase charge oscillations, the vertical electric fields of  
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8 acoustic plasmons are largely confined within the nanometric gap with a conducting plate, which  
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10 gives extreme plasmon confinement defined by the gap size. In contrast to conventional  
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12 graphene plasmons or a 2D electron gas that has a parabolic dispersion, interestingly, acoustic  
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14 plasmons exhibit a linear dispersion at small frequencies.<sup>28-30</sup>

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19 Recently, black phosphorus (BP) has been extensively studied as a novel anisotropic  
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21 plasmonic material.<sup>34-39</sup> In contrast to other 2D plasmonic materials, the inherent in-plane  
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23 anisotropy of BP renders the plasmon dispersion dependent on the propagation direction.<sup>34</sup> These  
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25 anisotropic plasmons are expected to enable the development of novel polarization-dependent  
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27 optoelectronic devices such as optical modulators,<sup>40,41</sup> tunable polarization rotators,<sup>37,42</sup> and  
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29 polarization-sensitive photodetectors.<sup>43,44</sup> One possible way to maximize the light-matter  
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31 interaction in BP for such applications is to leverage the extreme confinement of acoustic  
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33 plasmons. In this regard, it is imperative to understand how the in-plane anisotropy of BP is  
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35 manifested through the acoustic plasmon dispersion and how these plasmons enhance the light-  
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37 matter interactions in BP. For practical applications, in addition, new resonator configurations  
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39 for acoustic plasmons that require minimal post-processing after the BP deposition should be  
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41 investigated due to its instability in the ambient environment.<sup>45,46</sup>

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47 In this work, we theoretically investigate the dispersion of acoustic plasmons in freestanding  
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49 BP placed adjacent to a conducting plate. Using both analytical and numerical approaches, we  
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51 study how the in-plane anisotropy of BP is reflected in the plasmon dispersion and how the  
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53 acoustic plasmons scale with frequency  $\omega$  and gap size  $g$  with a conducting plate. The effect of  
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3 doping and BP thickness is examined as well. Further, we propose a practically viable and highly  
4 efficient design for an acoustic plasmon resonator, for which we use a modified Fabry-Perot (F-  
5 P) resonance model to describe the resonant behavior.  
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## 11 RESULTS AND DISCUSSION

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14 **Theory.** In the geometrical configuration considered here, a BP layer is placed above a  
15 conducting plate, and the distance between them is denoted by  $g$  (see Figure 1a). We consider  
16 surface plasmons propagating in the positive  $x$  direction. In order to study the influence of BP  
17 anisotropy on plasmon propagation, one of the two principal lattice axes, i.e., ‘armchair’ (AC)  
18 and ‘zigzag’ (ZZ), is aligned along the  $x$  direction. Figure 1b shows typical electric field  
19 distributions of conventional BP surface plasmons propagating along the AC direction for  $\lambda_0 =$   
20  $25 \mu\text{m}$  for comparison. Here, we used five layers of BP (thickness  $t = 2.675 \text{ nm}$ ) and assumed a  
21 damping constant of  $\eta = 10 \text{ meV}$  and an electron density of  $n = 1 \times 10^{13} \text{ cm}^{-2}$ . Although no  
22 reliable experimental values for the damping constant have been reported so far, previous  
23 research on graphene, which has similar damping pathways, indicates that a damping constant of  
24  $10 \text{ meV}$  is within an experimentally feasible range,<sup>20</sup> and accordingly, this value has been widely  
25 used in BP studies.<sup>34,37</sup> From now on, we will use the same condition for BP unless mentioned  
26 otherwise. The conductivities we used for numerical simulations are summarized in the  
27 supplementary material. Throughout the paper, we will mostly focus on the case of five layers  
28 due to their experimental feasibility and reproducibility.<sup>47</sup> However, we will still investigate the  
29 cases with a different number of layers including a monolayer for completeness. Conventional  
30 BP plasmons exhibit a symmetric field profile with the plasmon wavelength  $\lambda_c = 1200 \text{ nm}$ , which  
31 gives the vertical confinement of  $\lambda_c/2\pi = 191 \text{ nm}$ . Figures 1c and 1d show the field distributions  
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3 in the presence of a conducting plate for  $g = 5$  nm and  $\lambda_0 = 25$   $\mu$ m. As in the case of graphene,<sup>28-</sup>  
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<sup>30</sup> the electric field is constant across the gap region due to the out-of-phase charge oscillation between the BP layer and the conducting plate, which clearly shows that the observed mode is an acoustic plasmon. For the given  $\lambda_0$ , the vertical confinement of the acoustic plasmon within the gap is  $\lambda_0/5,000$ , which is around 38 times higher as compared to conventional plasmons propagating in the AC direction and 3 times higher than that of plasmons propagating in the ZZ direction. Contrary to the graphene case, the acoustic plasmon wavelength  $\lambda_{ac}$  largely differs for the two orthogonal directions (260 nm and 56 nm for AC and ZZ direction), which implicates a strong in-plane anisotropy in the plasmon dispersion for the BP case.

Before numerical investigation, we derive an analytical expression for the plasmon dispersion. The details for the mathematical derivation can be found in the supplementary material. Here, we define the dimensionless momentum  $q$  as  $k/k_0$  with the plasmon wavenumber in the  $x$  direction  $k$  and the free-space wavenumber  $k_0 \equiv \omega/c$ , with  $c$  being the speed of light in free-space. Thus,  $\text{Re}(q)$  directly gives the ratio of  $\lambda_0/\lambda_{ac}$ . For  $\text{Re}(q)\hat{g} \ll 1$ , the plasmon dispersion for a BP layer on a freestanding plane in a vacuum is given as follows.

$$q = \frac{i}{4\alpha} + \sqrt{\left(\frac{i}{4\alpha}\right)^2 + \frac{i}{2\alpha\hat{g}}}, \quad (1)$$

with the dimensionless conductivity  $\alpha \equiv (2\pi\sigma)/c$  and the dimensionless gap height  $\hat{g} \equiv k_0g$ . The in-plane anisotropy of BP is accounted for by using the anisotropic conductivity  $\sigma$  (in Gaussian units).<sup>34</sup>

$$\sigma = \sigma_{AC} \cos^2 \theta + \sigma_{ZZ} \sin^2 \theta. \quad (2)$$

Here  $\theta$  is the angle of propagation direction with the AC axis. We show that at low frequencies satisfying  $\omega \ll 4\sqrt{D/g}$  with the anisotropic Drude weight  $D = D_{AC} \cos^2\theta + D_{ZZ} \sin^2\theta$ , the plasmon dispersion in Eq. (1) is further simplified to

$$q = \frac{1}{\sqrt{-2i\alpha\hat{g}}}. \quad (3)$$

Under the additional assumption of  $\eta/\hbar \ll \omega$ , Eq. (3) becomes  $c/\sqrt{4gD}$  so that  $q$  becomes constant in  $\omega$  and scales with  $g$  as  $g^{-1/2}$ . Thus, the plasmon dispersion in Eq. (3) clearly shows the characteristic features of acoustic plasmons. Note that the constant  $q$  in  $\omega$  represents linear scaling with  $\omega$  in terms of  $k$  since  $k = q\omega/c$ . The linear scaling regime is accordingly given by the intersection of three inequalities; (1)  $\omega < 2\sqrt{D/t}$  ( $\equiv \omega_{pl}$ ), (2)  $\text{Re}(q)\hat{g} \ll 1$ , (3)  $\eta/\hbar \ll \omega \ll 4\sqrt{D/g}$ . The inequality (1) comes from the condition for the existence of plasmons,  $\text{Re}(\epsilon_{BP}) < 0$ , with the effective permittivity of BP,  $\epsilon_{BP} = 1 + i(4\pi\sigma)/(\omega t)$ , where the first (second) term denotes dielectric (Drude) response.<sup>48</sup> In the limit of zero thickness, the former is negligible, and its contribution increases with thickness. Similarly, a reduced doping will also enhance the relative dielectric contribution. Lastly, let us consider the two cases outside of linear scaling regime. At  $0 \leq \omega \lesssim \eta/\hbar$ , the plasmon dispersion in Eq. (3) scales with  $\omega$  as  $\omega^{-1/2}$  due to the increase in  $\text{Re}(\sigma)$ . At high frequencies where  $1 \lesssim \text{Re}(q)\hat{g}$ , on the other hand, the plasmon dispersion asymptotically approaches that of conventional plasmons without a conducting plate.

**Acoustic Plasmon Dispersion.** Plasmon dispersion curves for five layers of BP from numerical simulations along with those from Eq. (1) in the case of  $g = 5$  nm are shown in Figure 2a. In addition, the plasmon dispersion for the case of conventional plasmons without a conducting plate is plotted alongside for comparison. As shown in the figure, generally,  $\text{Re}(q)$  for the AC direction is smaller at a given  $\omega$  because of a larger  $\sigma$ . In the case of conventional



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3 plasmons without a conducting plate,  $\text{Re}(q)$  for the AC direction follows the classical parabolic  
4 scaling behavior (which corresponds to linear scaling with  $\omega$  in terms of  $q$ ), while for the ZZ  
5 direction, it largely deviates from that. This is attributed to the fact that  $\lambda_c$  becomes comparable  
6 to  $t$  as  $\omega$  approaches  $\omega_{\text{pl}} = (0.175 \text{ eV})/\hbar$ . The AC case shows no such tendency since  $\omega_{\text{pl}}$   
7 corresponds to higher frequency,  $(0.593 \text{ eV})/\hbar$ . In the presence of a conducting plate, the  
8 plasmon dispersion for the AC direction is nearly constant in  $\omega$  at most of frequencies showing  
9 the characteristic scaling behavior of acoustic plasmons except for the near-infrared (IR) regime  
10 and very small frequencies satisfying  $0 \leq \omega \lesssim \eta/\hbar$ . In the near-IR regime,  $\lambda_{\text{ac}}$  is comparable to  $g$   
11 so that the plasmon dispersion follows the conventional case. For  $0 \leq \omega \lesssim \eta/\hbar$ ,  $\text{Re}(q)$  increases  
12 with decreasing  $\omega$  as  $\omega^{-1/2}$  as expected from Eq. (3), which is the same for the ZZ direction.  
13 Physically, such divergent behaviors come from overdamped oscillations, as the real part of  
14 conductivity becomes very large at such low frequencies. In contrast to the AC case, however,  
15 the plasmon dispersion for the ZZ direction shows no linear scaling behavior owing to larger  
16  $\text{Re}(q)$  and smaller  $\omega_{\text{pl}}$ . As  $\omega \rightarrow (0.175 \text{ eV})/\hbar$ , it follows that for the case without a conducting  
17 plate. The larger  $\text{Re}(q)$  for the ZZ direction also results in a significant discrepancy between the  
18 numerical and the analytical results for the acoustic dispersion as well, while the two results for  
19 the AC direction are in a good agreement because of a smaller  $\text{Re}(q)$ .

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43 Figure 2b shows the figure of merit (FOM),  $\text{Re}(q)/\text{Im}(q)$ , for the plasmon dispersions given  
44 in Figure 2a. At small frequencies, the FOM increases almost linearly with  $\omega$  and the FOMs for  
45 different crystal axes have the similar value, as expected from the analytical results (see  
46 supplementary material). At higher frequencies, however, it starts to deviate from this trend  
47 before rolling down with increasing  $\omega$  as intraband Landau damping sets in. Particularly for the  
48 ZZ direction, FOM becomes zero at  $\omega = (0.175 \text{ eV})/\hbar$ . In contrast, the AC case is found to be  
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3 less damped and persists up to the near-IR regime due to lower plasmon confinement and larger  
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5  $\omega_{\text{pl}} = (0.593 \text{ eV})/\hbar$ . The numerical results also agree with the analytical results in that the FOM  
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7 for acoustic plasmons is always larger than those for conventional plasmons when  $\text{Re}(q)\hat{g} \ll 1$ .  
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9 In Figures 2c and 2d, we examined the effect of  $g$  on  $\text{Re}(q)$  and FOM given  $\omega = 0.025 \text{ eV}$ . For a  
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11 small  $g$ , where  $\text{Re}(q)\hat{g} \ll 1$ ,  $\text{Re}(q)$  scales with  $g$  as  $g^{-1/2}$  since it follows Eq. (1). For a large  $g$ , the  
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13 analytical expression deviates from the numerical results and  $\text{Re}(q)$  asymptotically approaches  
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15 the conventional case. Figure 2d shows the decrease in the FOM as the plasmon nature changes  
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17 from acoustic to conventional.  
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22 **Doping and Number of Layers Dependence.** From a practical viewpoint, it is important to  
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24 consider the effect of the electron density,  $n$ , as well as the number of layers,  $N$ , on the acoustic  
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26 plasmon dispersion, since many potential applications require an active tuning of the optical  
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28 properties of 2D materials. The plasmon dispersion at different  $n$  is shown in Figure 3a. With  
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30 increasing  $n$ ,  $\text{Re}(q)$  decreases at a fixed  $\omega$  due to the increase in  $D$  and accordingly  $\sigma$ .<sup>34</sup> In  
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32 addition, the increase in  $D$  broadens the plasmon-supporting band limited by  $\omega_{\text{pl}}$ . In the ZZ case,  
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34 in particular,  $\omega_{\text{pl}}$  is located within the frequency range of interest leading to substantial change in  
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36 the dispersion with  $n$ . In the AC case,  $\omega_{\text{pl}}$  is in the near-IR regime so that the plasmon dispersion  
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38 is less sensitive to  $n$ . For both directions, the FOM increases with increasing  $n$  at a given  $\omega$   
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40 because of lower plasmon confinement (Figure 3b). At low frequencies, however, the FOM does  
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42 not change appreciably with  $n$ .  
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47 Shown in Figure 3c is how the acoustic plasmon dispersion changes with increasing  $N$  for  $g$   
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49  $= 5 \text{ nm}$ . Here, we fixed  $n$  to be  $1 \times 10^{13} \text{ cm}^{-2}$ . For larger  $N$ , more sub-bands contribute to the  
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51 optical absorption, thereby increasing  $D$ .<sup>34</sup> For the AC direction, this leads to a slight decrease in  
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53  $\text{Re}(q)$  as  $N$  increases. The increase in  $\text{Re}(q)$  for  $N = 20$  in the near-IR regime is attributed to the  
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3 decrease in  $\omega_{\text{pl}}$ . The change in  $\omega_{\text{pl}}$  leads to the significant changes in the plasmon dispersion for  
4 the ZZ direction as well. With decreasing  $N$ , the FOMs have a larger value for a broader  
5 frequency range (Figure 3d). Note that the results in Figure 3 indicate the larger asymmetry in  
6 the dispersion for small  $n$  and larger  $N$ , which agrees well with the expectation from the  
7 anisotropy in  $\sigma$  (Figure S1b).  
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12 **Modal Reflection of Acoustic Plasmons.** In addition to the plasmon dispersion, the modal  
13 reflection of plasmons plays an important role in predicting the behavior of plasmonic  
14 resonators.<sup>49</sup> In this regard, we investigated the reflectance and reflection phase picked up by  
15 acoustic plasmons at the open edge of a BP/free-space/conducting plate system. Here, we focus  
16 on two different types of edge termination; semi-infinite BP/semi-infinite conducting plate  
17 (SBSC) and infinite BP sheet/semi-infinite conducting plate (IBSC), which are schematically  
18 illustrated in Figures 4a and 4b. Figures 4c and 4d show the electric field distributions along the  
19 AC direction after reflection at the SBSC and IBSC edges. In the SBSC case, acoustic plasmons  
20 are almost totally reflected, while in the IBSC case, they are coupled to conventional BP  
21 plasmons in the metal-free region.  
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38 Figures 4e and 4f show the reflectance and reflection phase picked up by plasmons after  
39 reflection at the SBSC and IBSC edges for different  $g$  of 2, 5, and 10 nm. In the SBSC case, the  
40 reflectance of acoustic plasmons is always close to unity due to the absence of a waveguide  
41 mode in free-space, similar to the graphene ribbon case.<sup>49</sup> For the ZZ direction where  $\lambda_{\text{ac}}$  easily  
42 becomes comparable to  $t$ , the reflectance is smaller due to more efficient coupling to photonic  
43 radiation modes at the edge. In the long wavelength limit where acoustic plasmons are more  
44 confined within the gap, the reflection phase approaches that for the metal gap plasmon case,  $-\pi$ ,  
45 due to the similarity in the mode profile.<sup>50,51</sup> With increasing  $\omega$ , the reflection phase converges to  
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3 a value around  $-0.5\pi$ . For the  $ZZ$  direction, the reflection phase converges to this value more  
4 rapidly as  $\omega \rightarrow (0.175 \text{ eV})/\hbar$ . Note that the non-trivial reflection phase obtained here is  
5 somewhat different from that for the monolayer graphene case,  $-3/4\pi$ ,<sup>49</sup> because of the larger  
6 thickness of five layers of BP. However, our numerical results show that  $-3/4\pi$  is recovered for a  
7 monolayer of BP.  
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12 In contrast to the SBSC case, the reflectance for the IBSC case abruptly decreases with  
13 increasing  $\omega$  so that at high frequencies, the reflection becomes negligible (Figure 4f). This is  
14 because of the small difference in  $q$  between acoustic and conventional plasmons, which also  
15 accounts for the smaller reflectance for the cases of propagation in the  $ZZ$  direction and larger  
16 gaps. The reflection phase scales with  $\omega$  similarly to the SBSC case, except that it has larger  
17 values at most frequencies and converges more rapidly. Note that we neglect the reflection  
18 phases for the  $ZZ$  direction at around  $\omega = \omega_{\text{pl}}$ , since the numerical simulations fails to give  
19 reliable values for the reflection phase because of a negligibly small  $\lambda_{\text{ac}}$ .  
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33 **Optical Responses of Acoustic Plasmon Resonators.** From our numerical results for the  
34 plasmon dispersion and the reflection phase, we estimate the resonant frequencies of two  
35 different types of acoustic plasmon resonators, having either periodic ribbons or a continuous BP  
36 sheet on a periodic array of conducting plates (gold). These two configurations are considered  
37 due to their experimental feasibility. The other feasible design, BP ribbons on a continuous  
38 conducting plate, is excluded due to its small far-field signal compared to the other  
39 configurations (see Figure S4). We also focus only on the AC case to compare the resonant  
40 behaviors of different configurations (for the  $ZZ$  direction, see supplementary material). Figure  
41 5a shows the far-field extinction spectra for the case of BP(ribbon)/metal(ribbon) with  $g = 5 \text{ nm}$   
42 as a function of inverse conducting plate width ( $1/w$ ). We set the distance between two  
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3 neighboring gold plates to be  $w$  as well. The far-field extinction is defined by  $1-T$  with  $T$  being  
4 the far-field transmittance normalized to that without a resonator. The red dashed lines show the  
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6 the estimated resonant frequencies from the plasmon dispersion in Figure 2a and the reflection phase  
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8 shown in Figure 4e using the F-P equation, which is  
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$$2k_{ac}w + 2\Phi_r = 2m\pi, \quad (4)$$

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11 where  $k_{ac}$ ,  $\Phi_r$  and  $m$  represent the wavenumber of acoustic plasmons, the reflection phase at the  
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13 edge and the order of the F-P resonance, respectively. As shown in the figure, the estimated  
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15 resonant frequencies for different F-P resonance orders agree very well with full numerical  
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17 results. Figure 5b shows extinction spectra for the case of BP(sheet)/metal(ribbon). In contrast to  
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19 the previous case, the estimated resonant frequencies using the reflection phase in Figure 4f  
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21 significantly deviates from full numerical results. In the BP(sheet)/metal(ribbon) case, the unit  
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23 cell of the plasmon resonator should be considered as a combination of two F-P resonators  
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25 formed within the gap and intermediate region. The modified F-P model gives the resonant  
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27 condition as (see supplementary material for details),  
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$$2k_{ac}w + 2k_cw = 2l\pi. \quad (5)$$

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36 Here,  $k_c$  represents the wavenumber of conventional plasmons, and  $l$  is the order of F-P  
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38 resonances. As shown in the figure, the resonance frequency estimated from the new model is a  
39  
40 decent match with numerical results. We emphasize that in the modified F-P model, the zeroth-  
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42 order F-P resonance is not allowed due to the absence of the phase term in Eq. (5) and  
43  
44 accordingly the first occurring mode corresponds to the second-order F-P resonance. As a result,  
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46 the plasmon resonances can occur at higher frequencies than the case of  
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48 BP(ribbon)/metal(ribbon).  
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3 **Comparison of Resonator Designs.** Based on the resonance model developed above, we  
4 investigate the extinction intensities of two different acoustic plasmon resonators together with a  
5 conventional BP ribbon resonator. Figures 6a-c illustrate optical coupling routes in three  
6 conventional BP ribbon resonator. Figures 6a-c illustrate optical coupling routes in three  
7 different designs. In the BP(ribbon) and BP(ribbon)/metal(ribbon) cases, incident waves are  
8 coupled to conventional surface plasmons or acoustic plasmons directly after scattering at the  
9 edge of resonator units. In the BP(sheet)/metal(ribbon) case, however, incident waves can launch  
10 both conventional and acoustic plasmons. Because of small reflection at the resonator edge, two  
11 plasmons are efficiently coupled to each other during propagation within the resonator, which  
12 can be considered as an indirect coupling of incident waves to plasmons. The electric field  
13 enhancement maps at the first occurring resonance ( $\omega_R = 0.083 \text{ eV}/\hbar$ ) of three different designs  
14 are shown in Figures 6d-f. The BP(sheet)/metal(ribbon) case shows the largest field  
15 enhancement on resonance than those of the other configurations. Figures 6e and 6f clearly show  
16 that the resonances indeed result from the acoustic (out-of-phase) mode as can be seen from  
17 highly confined and vertically constant electric fields within the gap. In the double ribbon case  
18 where the charges in two layers oscillate in-phase, however, the electric fields inside the gap are  
19 appreciably weak confirming that the optical (in-phase) mode is an active mode (see  
20 supplementary materials).  
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42 Figure 6g shows the extinction spectra for three different configurations where the first-  
43 occurring resonances coincide at  $\omega_R = (0.083 \text{ eV})/\hbar$ . Among three designs, the  
44 BP(sheet)/metal(ribbon) design shows the largest extinction intensity on resonance while the  
45 BP(ribbon)/metal(ribbon) case exhibits the smallest intensity. In addition, the higher order modes  
46 of the BP(sheet)/metal(ribbon) resonator have larger extinction intensities than those of the  
47 BP(ribbon) resonator, which indicates that the background transmission in the  
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3 BP(sheet)/metal(ribbon) resonator remains relatively smaller for a given  $w$ . We found out that  
4 the trend in Figure 6g between three designs holds in mid-IR and longer wavelength regimes,  
5 which covers most of the frequency range of interest (Figure 6h). With the same BP material  
6 parameters, the BP(sheet)/metal(ribbon) case exhibits the largest extinction on the first-occurring  
7 resonance of up to 60% for  $\omega_R < 0.142 \text{ eV}/\hbar$ . Using the plasmon dispersion and the extinction  
8 results in Figure 6h, we calculate the efficiency of coupling from free-space waves to plasmons,  
9  $\kappa$ , for different designs as given in the inset (for the details, see supplementary material). The  
10 inset shows that for the BP(sheet)/metal(ribbon) resonator, the coupling from free-space waves  
11 to acoustic plasmons is several times more efficient than the BP(ribbon) case. The strong  
12 coupling efficiency for the BP(sheet)/metal(ribbon) case is attributed to the fact that the  
13 metal(ribbon) array is highly efficient in focusing the free-space light into a slit mode between  
14 the conducting plate units, which facilitates coupling to the plasmon mode. The presence of the  
15 metal(ribbon) array also helps to reduce the background transmission for a large  $w$ , which  
16 explains the larger extinction intensities for higher order modes compared to the BP(ribbon)  
17 case. The similarity in the scaling behavior of the coupling efficiency between the BP(ribbon)  
18 and the BP(sheet)/metal(ribbon) indicates that the focused light in the slit mode in the  
19 BP(sheet)/metal(ribbon) case mostly excites acoustic plasmons via conventional plasmons as  
20 suggested in Figure 6c. At higher frequencies, however, the extinction intensity for the  
21 BP(ribbon) case is larger due to the significant increase in the other contributor to extinction  
22 intensity, i.e., a cavity quality factor, which is determined by the propagation loss of plasmons  
23 and the reflectance at resonator edges (for definition, see supplementary materials). In contrast to  
24 the indirect coupling to acoustic plasmons, the direct coupling from free-space waves to acoustic  
25 plasmons is inefficient due to the larger difference in wavenumber between them as can be seen  
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3 in the BP(ribbon)/metal(ribbon) case. In addition to higher extinction intensity, the  
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5 BP(sheet)/metal(ribbon) resonators are more suitable for practical implementation compared to  
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7 the other designs since potentially, no patterning steps are required after the deposition of BP,  
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9 thereby minimizing process-induced damages to BP.  
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## 17 CONCLUSION

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19 In conclusion, we have investigated the anisotropic dispersion for the acoustic plasmons in a  
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21 freestanding BP layer coupled to a conducting plate. The dispersion for the acoustic plasmons  
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23 was found to scale linearly with  $\omega$  in the mid- and far-IR regimes except in the long wavelength  
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25 limit. At high frequencies, where  $\lambda_{ac}$  becomes comparable to  $g$ , it approaches the dispersion of  
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27 conventional BP plasmons without a conducting plate. Due to larger confinement and narrower  
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29 plasmon-supporting band, the ZZ case largely deviates from the linear scaling behavior. The  
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31 analytical results confirmed the numerical results and clearly showed that the linear scaling  
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33 regime becomes broader for smaller gap size and number of layers, and higher carrier density.  
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35 Further, we numerically demonstrated different types of acoustic plasmon resonators including  
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37 BP(ribbon)/metal(ribbon) and BP(sheet)/metal(ribbon) configurations. Among feasible design  
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39 options, the BP(sheet)/metal(ribbon) resonator exhibited the largest extinction intensity than the  
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41 other possible configurations considered due to higher coupling efficiency. We developed a  
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43 modified F-P resonance model to account for the resonant behavior of such a plasmon resonator.  
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45 Importantly, our new resonator design can be realized using a continuous sheet of BP without  
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47 nano-patterning, which can introduce defects and edge roughness in BP. While an experimental  
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49 realization of acoustic plasmon resonances in BP is not trivial, recent advances<sup>47,52</sup> in the growth  
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3 of high-quality large-area BP samples show promising routes for the verification of our  
4 theoretical predictions. Also, our findings on acoustic plasmons in BP help to develop novel  
5 optoelectronic devices using optical anisotropy and extreme field confinement such as  
6 metasurfaces,<sup>10,12,13</sup> biosensors,<sup>15,30</sup> optical modulators,<sup>40,41</sup> molecular trapping,<sup>53-55</sup> and  
7 photodetectors.<sup>43,44</sup>  
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## 19 **METHODS**

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21 We used COMSOL Multiphysics with the RF Module for numerical simulations. In order to  
22 calculate the dispersion relation for acoustic plasmons in BP along with the reflection phase and  
23 amplitude, a port placed inside the simulation domain was used to solve for the eigenmode,  
24 launch the mode, and measure the reflection from the terminal interface. Perfectly matched  
25 layers (PMLs) were used at all simulation boundaries to increase accuracy. The electric field  
26 distributions in Figures 1 and 4 were also calculated in the same configuration. For Figures 5 and  
27 6, we used a plane wave with transverse magnetic polarization to obtain the extinction spectra of  
28 two acoustic plasmon resonators. Perfect electrical conductor (PEC) boundary conditions were  
29 used at both boundaries to simulate a periodic structure and reduce the computation time through  
30 symmetry. In most cases, the conducting plate was assumed to be 50 nm-thick gold with a  
31 dielectric function obtained elsewhere.<sup>56</sup> The Drude model is used to approximate the  
32 conductivity of multilayer BP (see supplemental material for details). For numerical calculations,  
33 BP is modeled as a slab with thickness  $t$  and effective permittivity  $\epsilon_{BP}$  as defined in the text.<sup>48</sup>  
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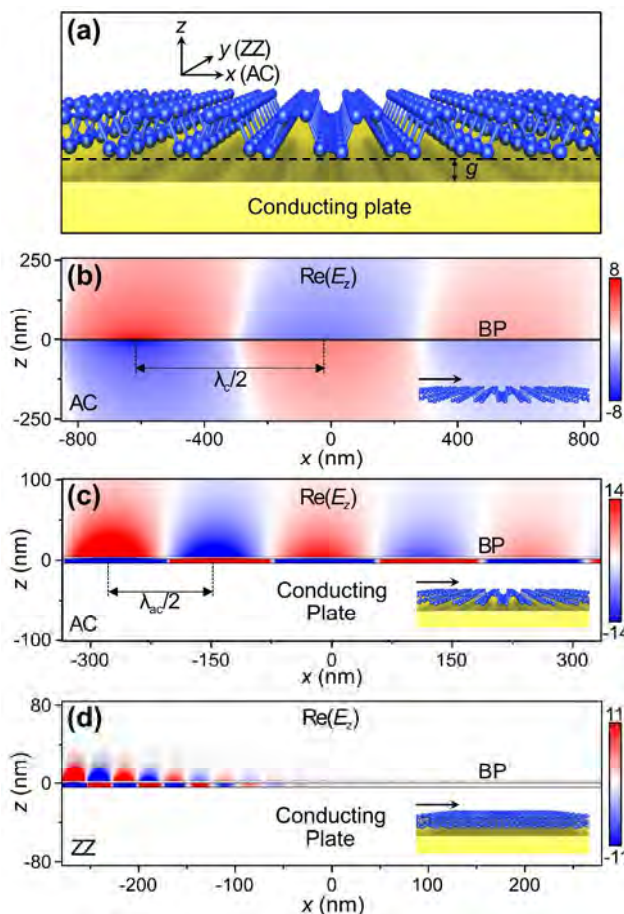
## 54 **AUTHOR CONTRIBUTIONS.**

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3 I.-H.L., L.M.M., and S.-H.O. conceived the idea. I.-H.L. and D.A.M. performed numerical  
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5 simulations using COMSOL. I.-H.L., L.M.M., K.K., and T.L contributed to theoretical analysis.  
6  
7 K.K. and T.L. calculated material parameters. All authors analyzed the data and wrote the paper  
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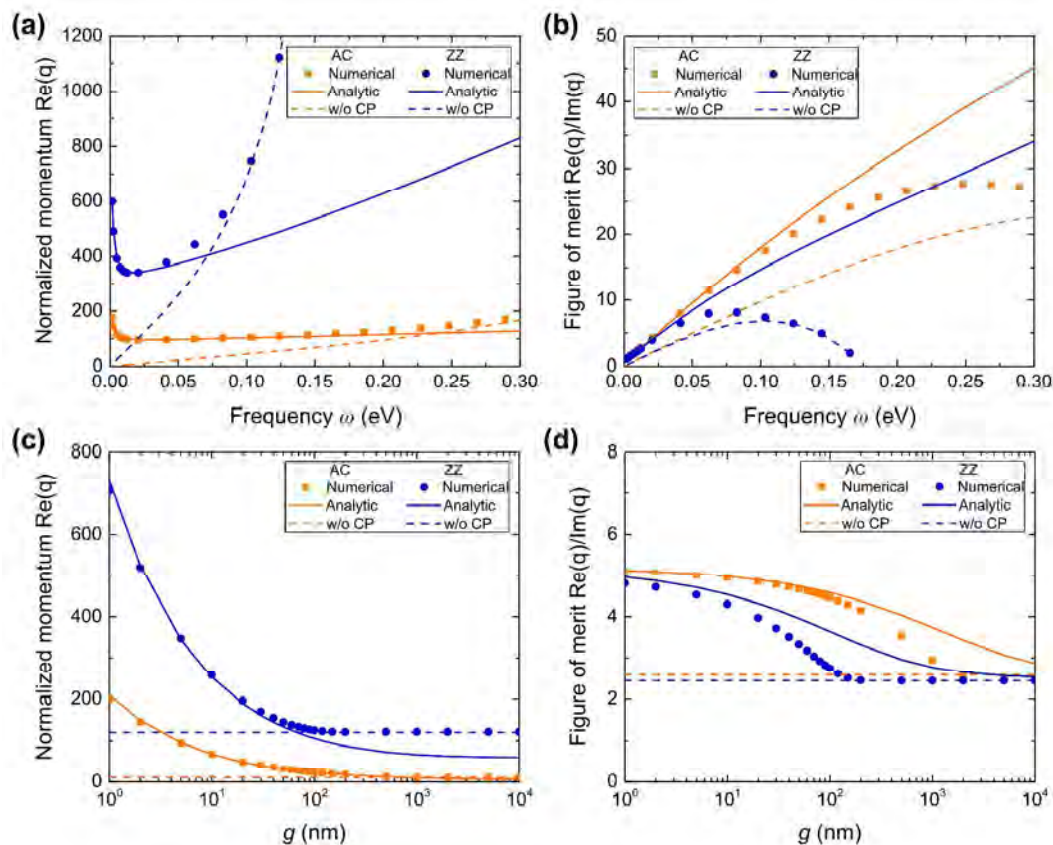
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20  
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22  
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24  
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26  
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28  
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31 the University of Minnesota.  
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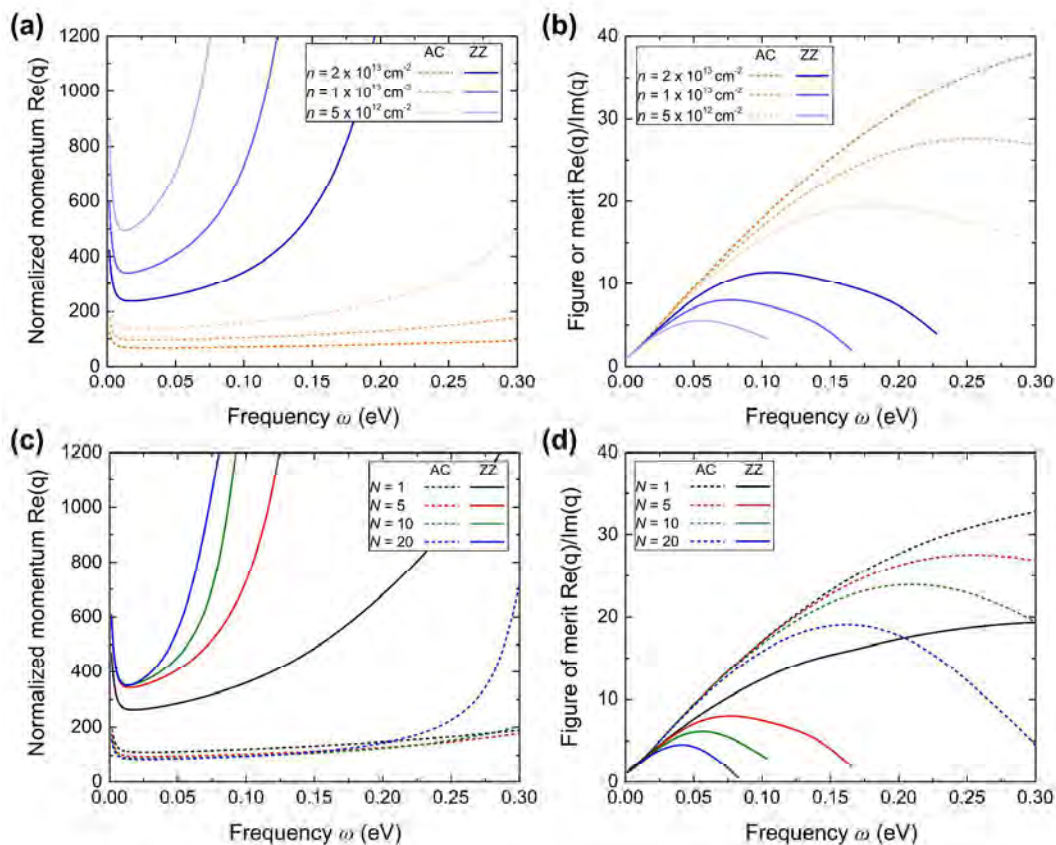
38 **Notes.** The authors declare no competing financial interests.  
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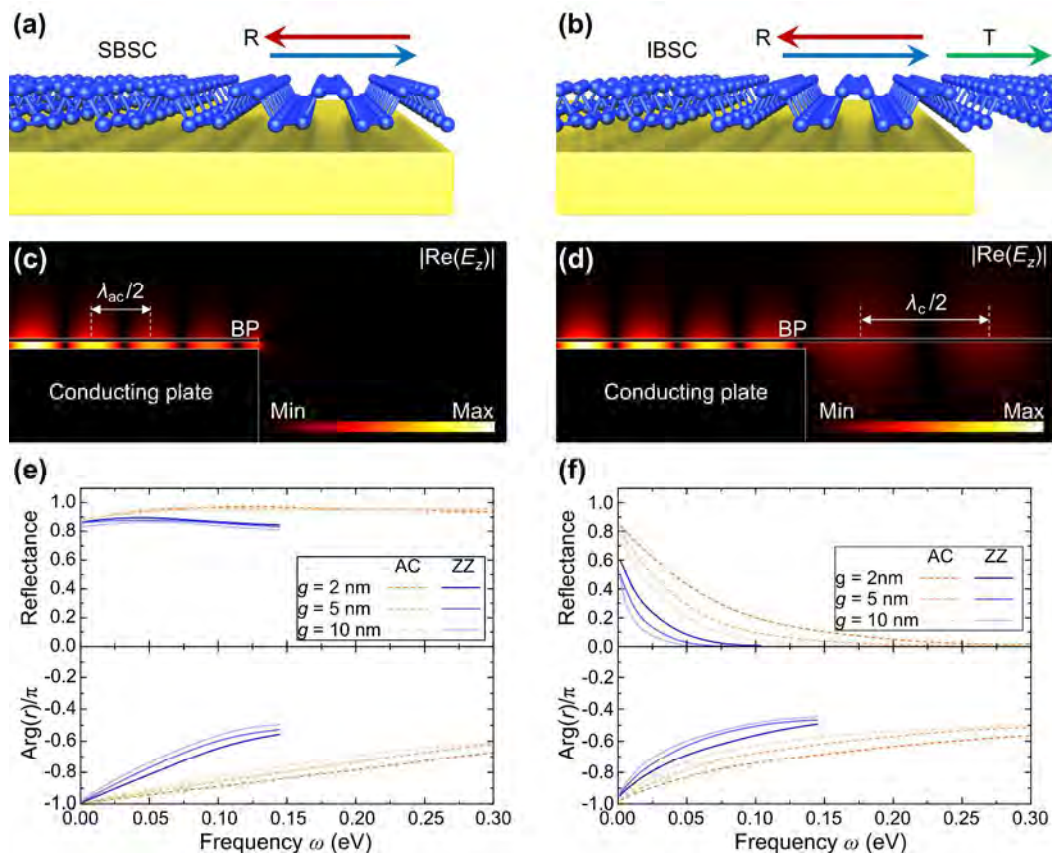
**Figure 1.** (a) The geometrical configuration supporting acoustic plasmons. The  $z$ -component of the electric field of (b) conventional plasmons propagating in the AC direction with the plasmon wavelength,  $\lambda_c$ , and acoustic plasmons propagating in (c) the AC and (d) ZZ direction with the plasmon wavelength  $\lambda_{ac}$  are shown for the free-space wavelength of 25  $\mu\text{m}$ . In (c) and (d), the gap between the BP and the conducting plate was 5 nm. The insets in (b)-(d) show the geometrical configurations considered and the arrows represent the propagation direction of plasmons (the positive  $x$  direction in all cases).



**Figure 2.** (a) The plasmon dispersion and (b) figure of merit (FOM) along the AC and ZZ directions. Here, we assumed  $g = 5$  nm. (c) The momentum and (d) FOM as a function of  $g$  along the AC and ZZ directions. In all cases, the dispersions given in terms of the real part of the dimensionless momentum,  $q \equiv k/k_0$  and FOM is defined as  $\text{Re}(q)/\text{Im}(q)$ . Numerical and analytical results are plotted together, and the numerical results for plasmons without a conducting plate (CP) are given for comparison.

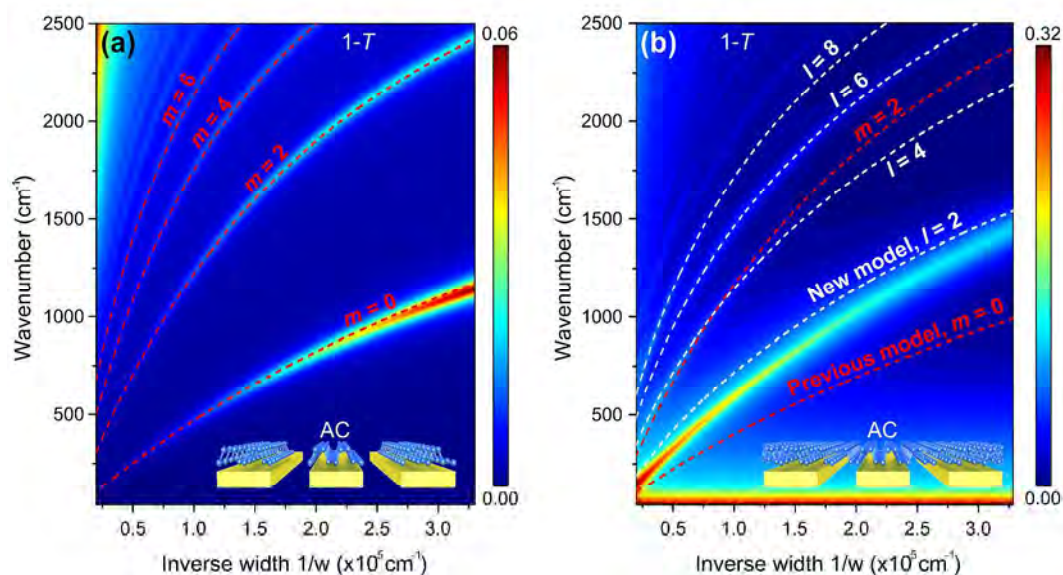


**Figure 3.** Effect of electron density,  $n$ , on (a) plasmon dispersion and (b) FOM.  $n$  is varied from  $5 \times 10^{12}$ ,  $1 \times 10^{13}$  to  $2 \times 10^{13} \text{ cm}^{-2}$ . The effect of the number of layers,  $N$ , on (c) plasmon dispersion and (d) FOM. Here, 1, 5, 10, and 20 layers are considered.

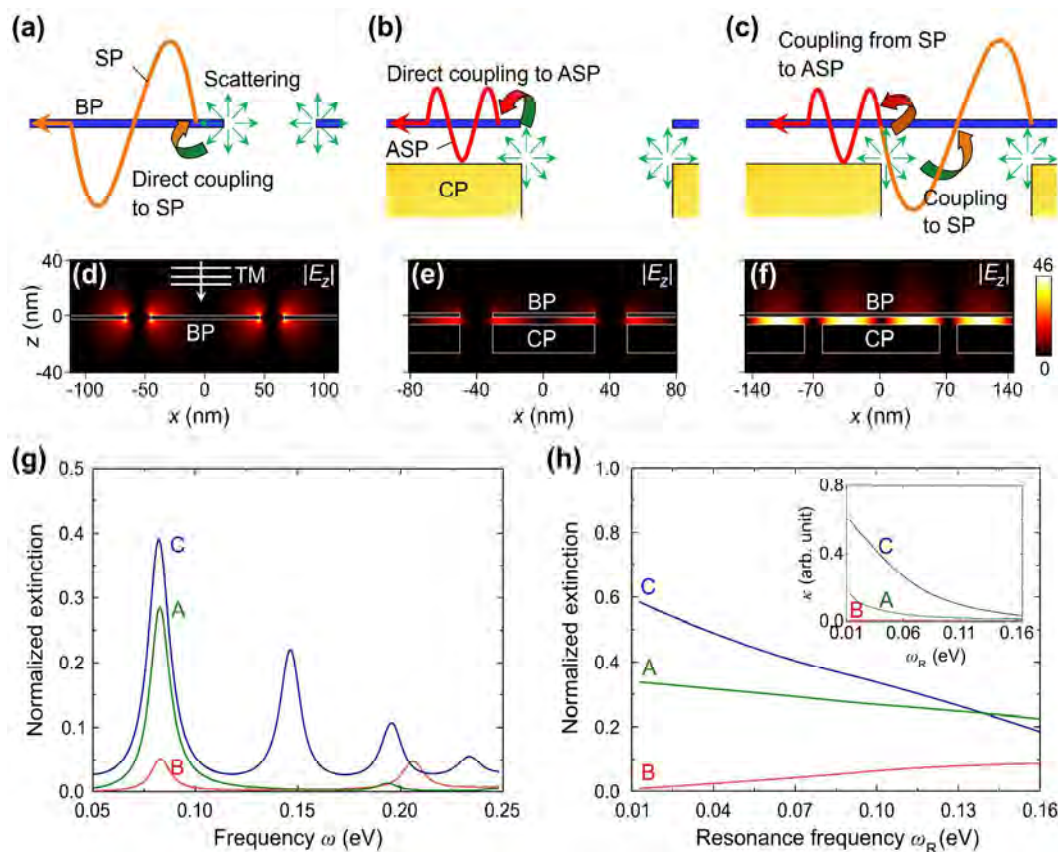


**Figure 4.** Schematic illustration of reflection at two different types of edge termination with (a) semi-infinite and (b) infinite BP sheet over the edge of a semi-infinite conducting plate (SBSC and IBSC cases, respectively). Electrical field distribution after reflection for (c) SBSC and (d) IBSC cases for AC direction,  $g = 5$  nm, and  $\lambda_0 = 25$   $\mu\text{m}$ . Reflection amplitude and phase of an acoustic plasmon after reflection at the edge for (e) SBSC and (f) IBSC cases for different  $g$  and crystal axes. Here,  $r$  denotes the reflection coefficient for acoustic plasmons.





**Figure 5.** Extinction spectra of BP acoustic plasmon resonators with (a) BP(ribbon)/metal(ribbon) and (b) BP(sheet)/metal(ribbon) for the AC direction as a function of inverse conducting plate width ( $1/w$ ) under illumination by a normally incident plane wave with transverse magnetic polarization. Red dashed lines are the estimated resonant frequencies for different orders of interference ( $m$ ) using the conventional Fabry-Perot resonance equation. White dashed lines are the estimation from the modified Fabry-Perot resonance equation for different orders ( $l$ ).



**Figure 6.** Schematic illustration of coupling routes to surface plasmons (SP) or acoustic surface plasmons (ASP) for (a) BP(ribbon) ('A'), (b) BP(ribbon)/CP(ribbon) ('B'), and (c) BP(sheet)/CP(ribbon) ('C') resonators where 'CP' means a conducting plate. The  $z$  electric field enhancement on resonance at  $\lambda_0 = 15 \mu\text{m}$  ( $\omega = 0.083 \text{ eV}/\hbar$ ) for (d) A, (e) B, and (f) C resonators. (g) Extinction spectra for three different geometries where the first-occurring resonance coincide at  $\lambda_0 = 15 \mu\text{m}$ . In (d)-(g), the widths of a resonator unit were 92, 61, and 128 nm for A, B, and C resonators, respectively. (h) Normalized extinction intensities on resonance as a function of resonance frequency for three different geometries. Inset shows corresponding coupling efficiency  $\kappa$ . In (d)-(h), both the spacing between two resonator units and the thickness of a conducting plate were fixed to be 20 nm for optimized performance. The resonators were illuminated by a normally incident plane wave with transverse magnetic (TM) polarization.



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## 7 Anisotropic Acoustic Plasmons in Black Phosphorus

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10 and Sang-Hyun Oh  
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