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Supplemental Material

Highly Sensitive and Stable Identification of Graphene Layers via The Topological Edge States and Graphene Regulation Enhancing

Photonic Spin Hall Effect[†]

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I. MANUFACTURING OF THE LAYERED STRUCTURE $\Downarrow \Downarrow \Downarrow \Downarrow \Downarrow \Downarrow \Downarrow \Downarrow \Downarrow$ (a) The laser lithography (b) After the lithography SiO₂ Photoresist The pattern transfers to Remove the photoresis the oxide in buffered gydrofluoric acid Graphene CaF The bulk etching of silicon in KOH (f) Insert the dielectrics into (e)

[10].

Furthermore, various methods exist for fabricating the layered structure (LS), including femtosecond laser drilling [4] and physical vapor deposition (PVD) [5-7]. Among these PVD techniques, presents distinct advantages for electromagnetic thin film fabrication compared to the others [8]. Thin film deposition techniques have shown the ability to accurately regulate both optical properties and thickness [9]. Fig. S2 provides a schematic of a basic PVD system. The process comprises four stages [10]: (i) a toms from the source material are vaporized by a high-energy source, like an ions or electron beam; (ii) the vapor is conveyed to the substrate; (iii) the metal atoms react with a relevant reactive gas (such as nitrogen, methane, or oxygen) during this process; and (iv) the resulting coating is applied to the substrate's surface.

II. THE CALCULATION FORMULA OF THE FRESNEL REFLECTION COEFFICIENTS OF P AND S WAVE

For the graphene, Si and CaF₂ layers, their respective transfer matrices re described as follows [11]:

Fig. S1. The illustrations depicting the fabrication process flow for the proposed LS. The specific materials are clearly indicated on the right side of the process flow. (a) The laser lithography. (b) After the lithography. (c) The pattern transfers to the oxide in buffered gydrofluoric acid. (d) Remove the photoresist.(e) The bulk etching of silicon in KOH. (f) Insert the dielectrics into the grooves.

In the manufacturing process, the etching technique is utilized to create the layered structure (LS). A silicon wafer serves as the substrate, and wet anisotropic etching is employed to form vertical grooves of varying thicknesses based on the scale of the two materials in the proposed design [1]. Specifically, this wet etching is conducted with a 44 wt% potassium hydroxide (KOH) solution at a temperature of 85 °C, with a thermally grown SiO₂ layer acting as a hard mask throughout the etching process [2]. Once the grooves meet the design specifications from our theoretical analysis, materials can be deposited into the corresponding areas [1,2]. Theoretically, as the height and width of the substrate increase freely, it can be considered an ideal structure, aligning with our theoretical predictions. The detailed fabrication process flow for the proposed LS is illustrated in Figs. S1(a)-(f). As shown in Ref. [3], this method produced an array of silicon nanowires with a height of 51 µm and a diameter of 550 nm, resulting in an aspect ratio of 93.

Power supply Fig. S2. The schematic diagram representation of a fundamental PVD system



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$$\boldsymbol{M}_{j} = \begin{pmatrix} \cos(k_{jz}d_{j}) & -\frac{i}{\eta_{j}}\sin(k_{jz}d_{j}) \\ -i\eta_{j}\sin(k_{jz}d_{j}) & \cos(k_{jz}d_{j}) \end{pmatrix},$$
(S1)

here, *j* represents various dielectrics, denoted as A, B, C, D, and G. The component of the wave vector along the +*z*-axis is given by $k_{jz}=\omega/cn_j \sin\theta_j$, where *c* denotes the speed of EWs in a vacuum. For *s*-waves, the EWs conductivity η_j is expressed as $\eta_j = (\varepsilon_0/\mu_0)^{1/2}n_j \cos\theta_j$. For *p*-waves, $\eta_j = (\varepsilon_0/\mu_0)^{1/2}n_j/\cos\theta_j$ [11]. ε_0 and μ_0 are vacuum permittivity and permeability. The transfer matrix for the structure G(SiCaF₂)^N(SiCaF₂)^N is [11]:

$$\mathbf{M} = \mathbf{M}_{G} (\mathbf{M}_{A} \mathbf{M}_{B})^{N} (\mathbf{M}_{C} \mathbf{M}_{D})^{N} = \begin{bmatrix} m_{11} & m_{12} \\ m_{21} & m_{22} \end{bmatrix}, \quad (S2)$$

where, m_{11} , m_{12} , m_{21} , and m_{22} are elements of the overall transfer matrix. The period N=4. The reflection coefficient r and the transmission coefficient t can be obtained using the following equations [11]:

$$r = \frac{(m_{11} + m_{12}\eta_0)\eta_0 - (m_{21} + m_{22}\eta_0)}{(m_{11} + m_{12}\eta_0)\eta_0 + (m_{21} + m_{22}\eta_0)},$$
(S3)

$$t = \frac{2\eta_0}{(m_{11} + m_{12}\eta_0)\eta_0 + (m_{21} + m_{22}\eta_0)}.$$
 (S4)

The Fresnel reflection coefficients of p and s waves are severally expressed as r^p and r^s , and the Fresnel transmission coefficients of p and s waves are represented as t^p and t^s . The corresponding reflectance and transmittance are given by $R^{p}=|r^p|^2$, $T^{p}=|t^p|^2$, $R^{s}=|r^{s}|^2$ and $T^{s}=|t^{s}|^2$ [11]. The p-waves aborptivity A^p and s-waves absorptivity A^s can be calculated as followed [11]:

$$A^p = 1 - T^p - R^p, \tag{S5}$$

$$A^s = 1 - T^s - R^s. \tag{S6}$$



Fig. S3. The absorptivity curves of *p*-waves correspond to the LS without monolayer graphene and different graphene μ_c .

The absorption A^p of *p*-waves is calculated according to (S5) in the case of the LS without monolayer graphene and different graphene chemical potential $\mu_{\rm C}$, respectively. Fig. S3 shows that when the LS does not have the monolayer graphene, its A^p is close to 0, because the overall LS is consist of lossless dielectrics Si and CaF₂. At this time, corresponding to Fig. 2 and Fig. 3(a) and (b), topological edge states are formed at the interface of LS₁ and LS₂, and the transmission peak of high quality factor is generated. When the graphene layer is loaded on the surface of LS, there are obvious A^p curves in Fig. S3, which change with the variation of $\mu_{\rm C}$. This is due to the introduction of graphene, LS has a loss, and a part of the propagating electromagnetic waves is absorbed. However, because the thickness of graphene (0.34 nm for monolayer) is too thin compared to the whole LS thickness of up to 3145.6 μ m, the influence on the bands of LS₁ and LS₂ in Fig. 2(a) can be ignored, and topological edge states still occur. The frequency point of topological edge states transmission peak is fixed. As can be seen from the electric field energy distribution diagram in Fig. 5(d), the electric field is localized in the graphene layer at the LS surface, which is the cause of graphene absorption. At the interface of LS_1 and LS_2 , the electric field is still localized, forming topological edge states. This also explains that in Figs.4 (a)-(d), combined with the transmission of topological edge states and the absorption of graphene, the reflection coefficient r^p of p-waves can reach the minimum value at a fixed frequency, and the r^p value is very small when $\mu_{\rm C} = 0.1 \, {\rm eV}$.



Fig. S4. (a) The p-wave transmissivity of the LS corresponding to the absence of graphene and the loading of multilayer graphene, and (b) the enlarged diagram.

Fig.S4 discusses the influence of the graphene layers number on the topological characteristics of the LS. As shown in Fig. S4(a), when there is no graphene layer, the topological edge states transmissivity peak T^p of the *p*-wave is generated at *f*=198.493 GHz. When the number of loaded graphene layers ranges from 1 to 25, T^p gradually decreases due to the enhanced intrinsic absorption of graphene. Fig. S4(a) is an enlarged image. Since the graphene layers is thin enough relative to the overall structure and has almost no influence on the topological edge states, the frequency point of T^p is fixed at 198.493 GHz and remains unchanged.



Fig. S5. (a) The transmittance curves of the *s*-waves and *p*-waves simulated by the transfer matrix method (T^s and T^p) and the finite element algorithm (T^{s} ' and $T^{p'}$), and (b) the enlarged diagram.

To validate the accuracy of the simulation results, we employed both the Transfer Matrix Method (TMM) [11] and the Finite Element Method (FEM) [12] to simulate the transmission spectra of the s-wave and p-wave under the topological edge states shown in Fig. 3(a). As illustrated in Fig. S5, the s-wave transmission peak (T^s) and p-wave transmission peak (T^p) obtained from the TMM occur at frequencies of f=198.687 GHz and f=198.493 GHz, respectively. Meanwhile, the s-wave transmission peak $(T^{s'})$ and p-wave transmission peak (T^{p}) obtained from the FEM appear at frequencies of f=198.55 GHz and f=198.36 GHz, respectively. The frequency difference between T^s and $T^{s'}$ is 0.137 GHz, and 0.133 GHz is the frequency difference between T^p and T^p ', indicating that the shifts produced by both algorithms frequency are approximately the same. However, the peak values for both the s-wave and p-wave obtained from the TMM and FEM are equal, i.e., $T^{s}=T^{s}$ = 0.921 and $T^{p}=T^{p}$ = 0.923. This phenomenon can be attributed to the electromagnetic waves being incident at an angle of 10°. Because the transfer matrix method is an analytic solution algorithm, and the finite element algorithm is a numerical solution algorithm, there will be truncation error when the network is divided. In addition, differences between the two algorithms in handling boundary conditions, phase, and numerical errors cause the transmission curve to exhibit the same frequency shift while maintaining the same peak values [13,14]. The ratio between the frequency difference of 0.137

GHz and 0.133 GHz and the corresponding operating frequency point is only 0.069 % and 0.067 % (as shown in Fig. S5(a)), indicating that the difference of algorithms has little effect on the performance of the whole structure and does not affect the corresponding conclusion. Therefore, based on the mutual verification of the two scientific and authoritative algorithms, the correctness of the simulation results of this work is guaranteed.

IV. FRABRICATION OF MULTILAYER GRAPHENE ON LS SUBSTRATE

First, graphene is produced via rapid thermal chemical vapor deposition (RT-CVD) with a copper foil (35 µm thick, 99.85% purity) serving as the substrate. After the growth process, the graphene film on the copper foil is affixed to a thermal release tape (TRT, Jinsung Chemical Co.) using a laminator. The graphene/copper/TRT structure is then submerged in an etching chamber filled with a sulfuric acid etching solution and hydrogen peroxide. Once the copper foil has been completely etched away, the graphene adhered to the TRT is washed with deionized water. The graphene film on the TRT is subsequently placed in a laminator with a 3145.6 µm-thick LS to create a TRT/graphene/LS composite. The TRT can be easily removed by applying heat to the TRT/graphene/LS composite, as the TRT loses its adhesion at elevated temperatures [15]. This process can be repeated multiple times to achieve additional layers of graphene on the LS.

V. THE METHOD TO MODULATE THE GRAPHENE CHEMICAL POTENTIAL



Fig. S6. Top view of the external bias voltage applied to graphene..



Multilayer graphene transfer on LS substrate (c)





(O₂ plasma etching)

Cr/Au electrode deposition (thermal evaporation)

Spin coating ion gel

Fig. S7. Fabrication process of graphene-based LS. (a) Multilayer graphene transfer on LS substrate. (b) Graphene patternin (O_2 plasma etching. (c) Cr/Au electrode deposition (thermal evaporation). (d) Spin coating ion gel.

To induce a bias voltage in the graphene, this method patterns a rectangular gold (Au) electrode around the graphene layer, along with a independent ground Au electrode on the LS substrate. A thin layer of ion gel, composed of the ionic liquid 1-ethyl-3-methylimidazolium bis (trifluoromet hylsulfonly)imide, [1-ethyl-3methylimidazolium][Trifluoromethanesulfonimide], poly fluoride-co-hexafluoropropylene), (vinvlidene and polyvinylidene fluoride-hexafluoropropylene copolymer, [1ethyl-3-methylimidazolium][Trifluoromethanesulfonimide], is applied atop the graphene and Au electrodes to adjust the graphene layer $\mu_{\rm C}$ [16].

About fabrication, multilayer graphene is dry-transferred on a 3145.6-µm-thick LS substrate with thermal release tape. Then, the graphene layer is etched using O₂ plasma ashing setup (100 W, 45 s). The pattern can be precisely defined by placing a shadow mask on the top of the graphene layer and a plane-type magnet on the backside of the PET substrate while etching. To make an electrode, the Cr/Au layer is deposited using a thermal evaporator (Cr/Au: 10 nm/50 nm) with a shadow mask. Because LS substrate is fragile to hightemperature, the whole device IS maintained at 5 °C during the deposition process. The ion gel solution is prepared by mixing polyvinylidene fluoride-hexafluoropropylene copolymer, ionic liquid [1-ethyl-3-

methylimidazolium][Trifluoromethanesulfonimide], acetone, and methyl ethyl ketone with a weight ratio of 1:4:10:10. The solution is stirred for more than 1 week at 53 °C. Then, the iongel layer is spin-coated on the graphene and electrode pattern at 4000 rpm, and the entire device is cured in a vacuum oven at 70 °C for 12 hours [15].

REFERENCES

- [1] S. Guo, C. Hu, H. Zhang, Journal of Optics, 2020, 22, 105101.
- [2] W. M. J. Green, J. Scheuer, G. DeRose, *Applied physics letters*, 2004, 85, 3669-3671.
- [3] L. Xiuling, Current Opinion in Solid State and Materials Science, 2012, 16, 71-81
- [4] R. B. Gowda, P. Sharan, K. Saara, Results in Optics, 2023, 10, 100352.
- [5] X. Zhao, Y. C. Shin, Applied Physics A, 2011, 104, 713-719.
- [6] J. J. J. Kaakkunen, M. Silvennoinen, K. Paivasaari, and P. Vahimaa, *Physics Procedia*, 2011, **12**, 89-93.
- [7] Q. Xie, X. Li, L. Jiang, B. Xia, X. Yan, W. Zhao, and Y. Lu, *Applied Physics A*, 2018, **122**, 1-8.
- [8] L. Grineviciute, C. Babayigit, D. Gailevičius, E. Bor, M. Turduev, V. Purlys, and K. Staliunas, *Applied Surface Science*, 2019, 481, 353-359.
- [9] S. Wilbrandt, O. Stenzel, and N. Kaiser, Journal of Physics D: Applied Physics, 2007, 40, 1435.
- [10] A. S. H. Makhlouf, Woodhead Publishing, 2011, 3-23.
- [11] L. Qi, Z. Yang, F. Lan, X. Gao, and Z. Shi., Physics of Plasmas, 2010, 17, 0425016.
- [12] G. Meunier, The finite element method for electromagnetic modeling.
- [13] A. F. Koenderink, W. L. Vos. JOSA B. 2005, 22, 1075-1084.
- [14] I. Haddouche, L. Cherbi, Optics Communications, 2017, 382, 132-137.
- [15] M. Lim, H. D. Kim, H. C. Shim, Nano Energy, 2024, 131, 110189.