

# Supplementary Materials

## Origin of Giant Rashba Effect in Graphene on Pt/SiC

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### ARPES and spin-ARPES data analysis

Fig. S1(a) shows the ARPES intensity map of the Pt intercalated graphene on the SiC substrate. The energy distribution curve (EDC) profiles taken from the ARPES intensity map are shown in Fig. S1(c, d, e) to visualize the shape-changing of the spectra with increasing  $\pi$  states binding energy (or corresponding photoemission angle/momentum). It is observed that in addition to the main peak (marked as  $\pi$ ), there are low-intensity peaks on the left and right sides. One of them corresponds to the branch of the initial Dirac cone that was present before Pt intercalation (marked as  $\pi'$ ), and the second one can be associated with Pt d states in accordance with Ref. [13]. As a result, the shape of the spectrum is more complex, and we took these shape features into account when analyzing the spin-resolved ARPES data.

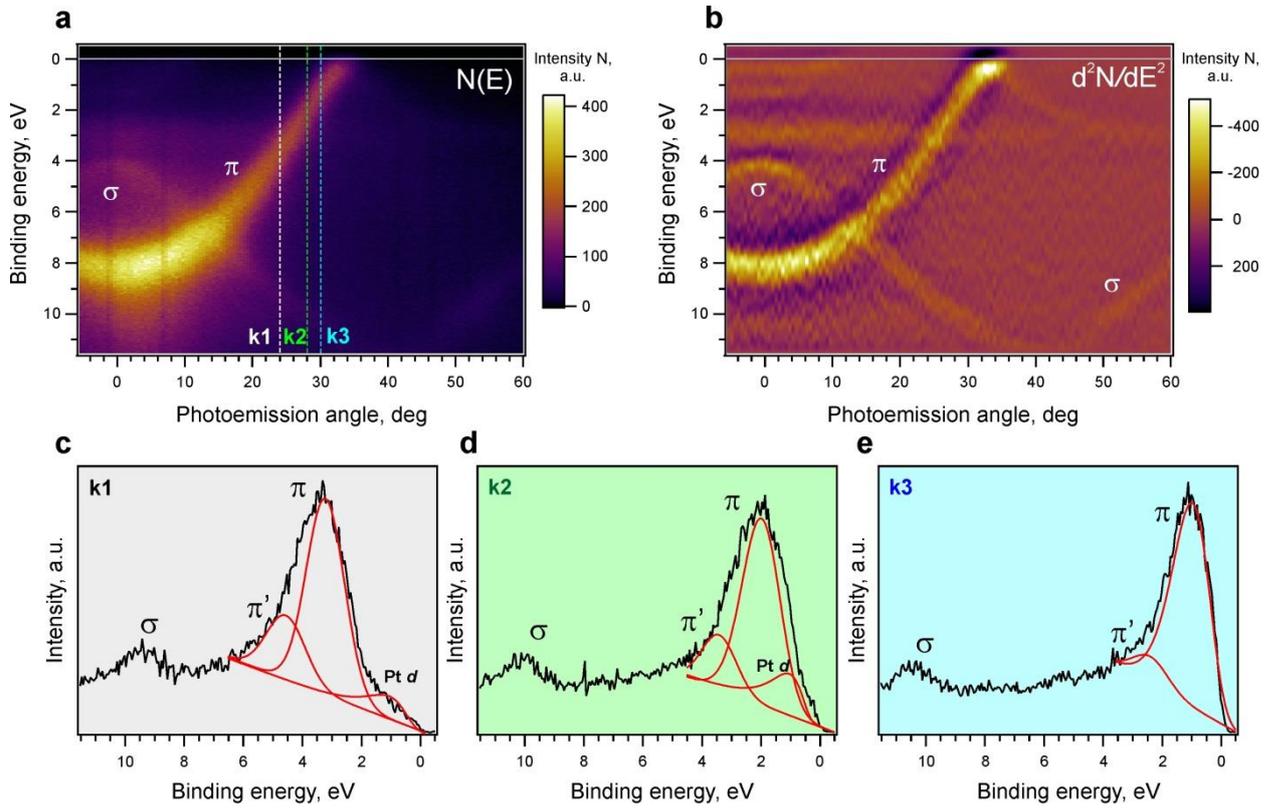


Figure S1. (a) – ARPES intensity map of the Pt intercalated graphene measured along the  $\Gamma K$  direction of the surface Brillouin zone. (b) – The same ARPES intensity map presented as the second derivative by energy. (c, d, e) – EDC profiles taken from the ARPES intensity map on (a) at different values of emission angles ( $k_{||}$ ).

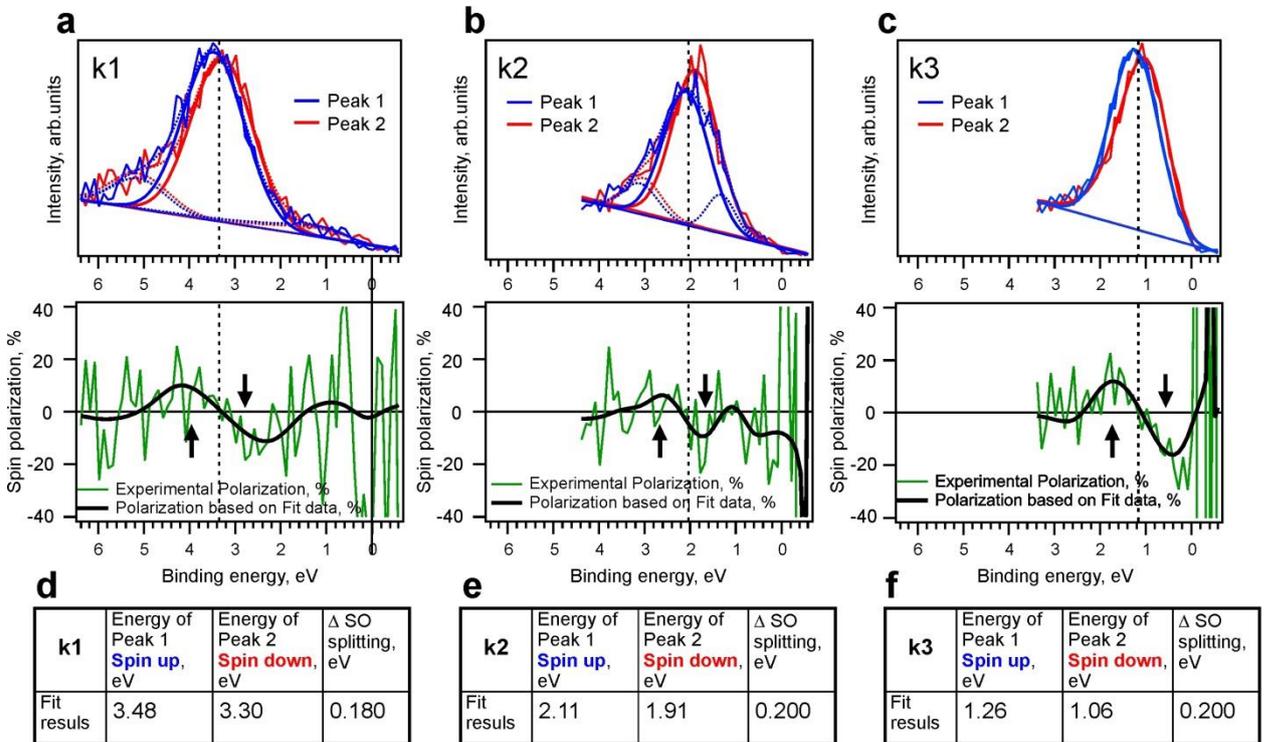


Figure S2.(a, b, c) - Spin-resolved ARPES spectra of the Pt intercalated graphene measured at different values of emission angles ( $k_{||}$ ) along the  $\Gamma K$  direction of the surface Brillouin zone –

*the spectra presented with the best-fit results. The original spin polarization curves and curves, calculated inversely from the fit data, are presented below the spin-resolved spectra. Blue and red color denote opposite projections of electron spin. (d, e, f) – Tables with the binding energies of  $\pi$  states and the spin splittings derived from the fitting procedure.*

In Fig. S2, the spin-ARPES spectra measured at different emission angles ( $k_{II}$ ) are presented. To determine the spin splitting of graphene  $\pi$  states, we fitted the original spin-resolved data with several Gaussian peaks and linear background. The best-fit results for different  $k_{II}$  profiles are also presented in Fig. S2. The original polarization curves are shown below the spin-resolved data. Additionally, we calculated the polarization curves inversely from the fit spectra by the following expression:

$$p^{fit} = \frac{I_{up}^{fit} - I_{down}^{fit}}{I_{up}^{fit} + I_{down}^{fit}},$$

where  $I_{up}^{fit}$  and  $I_{down}^{fit}$  are the best-fit curves for the corresponding original spectra. It is worth noting that original polarization curves have a strong deviation near the Fermi energy since it is produced by division of small values ( $I_L - I_R$ ) and ( $I_L + I_R$ ) due to a low intensity of photoemission signal near the Fermi energy (division by nearly zero gives a large uncertainty).

Fig. S2 (d-f) shows the values of the energy position of the spin-up and spin-down  $\pi$  states obtained as a result of the fitting procedure. The difference between these values gives us a spin splitting value of 180-200 meV. The obtained spin splittings agree with those determined from the peak maxima of the spin-ARPES spectra because of the high intensity of  $\pi$  states compared with additional states.