Title
Experimental evidence of plasmarons and effective fine structure constant in electron-doped graphene/h-BN heterostructure

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Electron-electron interaction is fundamental in condensed matter physics and can lead to composite quasiparticles called plasmarons, which strongly renormalize the dispersion and carry information of electron-electron coupling strength as defined by the effective fine structure constant \( \alpha_{ee} \). Although h-BN with unique dielectric properties has been widely used as an important substrate for graphene, so far there is no experimental report of plasmarons in graphene/h-BN yet. Here, we report direct experimental observation of plasmaron dispersion in graphene/h-BN heterostructures through angle-resolved photoemission spectroscopy (ARPES) measurements upon in situ electron doping. Characteristic diamond-shaped dispersion is observed near the Dirac cone in both 0° (aligned) and 13.5° (twisted) graphene/h-BN, and the electron-electron interaction strength \( \alpha_{ee} \) is extracted to be \( \alpha_{ee} \approx 0.9 \pm 0.1 \), highlighting the important role of electron-electron interaction. Our results suggest graphene/h-BN as an ideal platform for investigating strong electron-electron interaction with weak dielectric screening, and lays fundamental physics for gate-tunable nano-electronics and nano-plasmonics.

**INTRODUCTION**

Electron-electron interaction is ubiquitous in solids and plays an important role in condensed matter physics. In graphene, the strength of the electron-electron interaction is quantified by the ratio of the Coulomb potential \( U = e^2k_F/e \) to the kinetic energy \( K = \hbar v_F k_F \), where \( v_F \) and \( k_F \) are the Fermi velocity and Fermi momentum respectively, and \( e \) is the dielectric constant. This ratio defines a fundamental constant \( \alpha_{ee} = U/K = e^2/\epsilon \hbar c \), which is in analogy to the fine structure constant \( \alpha = e^2/\hbar c = 1/137 \) in quantum electrodynamics, with the speed of light \( c \) replaced by the Fermi velocity \( v_F \) and the effective dielectric screening of the environment taken into account by \( \epsilon \). Because of this analogy, \( \alpha_{ee} \) is called the effective fine structure constant. The effective fine structure constant \( \alpha_{ee} \) reflects the relative strength of electron-electron interaction and determines many fundamental physical properties of graphene, e.g., optical absorption and transport properties. Moreover, by tuning the carrier concentration and electron-electron correlation, superconductivity has been reported in twisted bilayer graphene placed on h-BN substrate. Revealing the electron-electron interaction strength and extracting the effective fine structure constant are fundamentally important.

Electron-electron interaction can significantly affect the electronic dispersion by reshaping the graphene Dirac cone dispersion with a modified Fermi velocity. Moreover, collective excitations of electron gas can form plasmons, and the interaction between plasmons and charges leads to composite quasiparticles called plasmarons, which strongly modify the electronic dispersion with newly generated plasmaron bands.
Emergence of plasmaron bands in 0° aligned graphene/h-BN upon electron doping

Signatures of plasmaron bands first become observable at a carrier concentration of \(8.1 \times 10^{12} \text{ cm}^{-2}\) as two twisted bands near the Dirac point (marked by red arrows in Fig. 2d, e). The twisted bands become more obvious and increase in size with doping, and they eventually evolve into a clear diamond-shaped dispersion at even higher electron doping (Fig. 2f–h), indicating the emergence of plasmaron bands. This is different from graphene on SiC substrate where no diamond-shaped dispersion is observed\(^{13}\). In addition to the diamond-shaped dispersion near the Dirac point energy, moiré replica bands are also clearly observed on both sides of the graphene bands near \(E_F\) (Fig. 2e–h). Such moiré superlattice replicas are also observed as replica pockets in the Fermi surface map shown in Fig. 2i. To summarize the evolution of the electronic structure upon electron doping, the Dirac point energy \(E_D\) and plasmaron crossing energy \(E_{pm}\) (defined as the crossing energy between graphene Dirac cone and the plasmaron bands, indicated by red arrows in Fig. 2d–h) are both plotted in Fig. 2j. Plasmaron bands are clearly observed at intermediate to high electron doping with Dirac point energy \(E_D\) shifted from \(-0.28\) eV to \(-0.64\) eV (blue symbols in Fig. 2j). Such shift in \(E_D\) corresponds to carrier concentration ranging from \(8.1 \times 10^{12} \text{ cm}^{-2}\) to \(5.8 \times 10^{13} \text{ cm}^{-2}\). With increasing electron doping, the energy separation between \(E_D\) and \(E_{pm}\) becomes larger. We note that the moiré superlattice period is determined by the lattice mismatch between graphene and h-BN\(^{24}\) and is doping independent, and as a result, the separation between the moiré superlattice replica and the original Dirac cone is expected to be doping independent. Therefore, the increasing energy and momentum separation between the plasmaron bands and Dirac cone upon electron doping (see Supplementary Fig. 3 for more details) confirms that it is not caused by overlapping of graphene Dirac cone and moiré superlattice bands, but instead band renormalization induced by plasmon-charge interaction. In addition, the ARPS dispersions remain quite sharp, and indeed they are sharper than those at lower carrier concentration. This is in agreement with previous report on graphene/h-BN at a lower doping (with the Dirac point at \(-0.3\) eV\(^{8}\), where the decrease of scattering rate is attributed to the increase of long-range impurity screening from the higher electron density. Here we show that at an even higher doping (with the Dirac point at \(-0.64\) eV) and in the presence of plasmarons, the electron scattering rate still remains low, which is useful for gate-tunable nano-electronics and plasmonics.

**Plasmaron bands observed in a 13.5° twisted graphene/h-BN upon electron doping**

Since the moiré superlattice period of graphene/h-BN strongly depends on the twist angle\(^{15}\), to check whether the plasmaron features depend on the twist angle, we show in Fig. 3 ARPES results on a 13.5° twisted graphene/h-BN heterostructure. The moiré superlattice period decreases from \(\lambda \approx 14\) nm at 0° to 1.05 nm at 13.5°, and therefore the superlattice replica Dirac cone is much farther away from the graphene Dirac cone. Figure 3a shows...
the exfoliated monolayer graphene, which was then transferred onto a h-BN flake (shown in Fig. 3b) with a designed twist angle of 13.5°. This twist angle is confirmed by the K points of graphene and h-BN from energy contours at the Fermi energy (Fig. 3c) and −2.9 eV (Fig. 3d). By performing ARPES measurements with in situ Rb deposition, the evolution of the band dispersion upon electron doping is revealed. Figure 3e–g shows the dispersions measured at different carrier density, with a characteristic diamond-shaped

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**Fig. 2** Formation of plasmaron bands upon doping in a 0° aligned graphene/h-BN. a–h Evolution of the electronic structure through the K point (indicated by black dotted line in i) upon electron doping. $E_0$, $E_{SDC}$, and $E_{pm}$ indicate the Dirac point energy, second-generation Dirac point energy, and plasmaron crossing energy respectively. i Fermi surface map at doping level corresponding to data shown in g, j Dirac point energy $E_0$ and plasmaron crossing energy $E_{pm}$ as a function of electron doping. Black broken curves are guides to the eyes.

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**Fig. 3** Plasmaron dispersion observed in a 13.5° twisted graphene/h-BN heterostructure. a, b Optical images of exfoliated monolayer graphene and graphene/h-BN heterostructure. The twist angle determined from the straight edge is 73.5° (equivalent to 13.5°). Scale bar indicates 100 μm. c, d Energy contours at Fermi energy and −2.9 eV to reveal the K points of graphene and h-BN, which confirm the twist angle of 13.5°. e–g Dispersion of 13.5° graphene/h-BN at different carrier density to show the evolution of plasmaron bands upon doping. h Dispersion of 0° graphene/h-BN at doping level of $3.7 \times 10^{13}$ cm$^{-2}$ (also shown in Fig. 2g) to compare with results on 13.5° graphene/h-BN sample. i–l 2D curvature results of e–h, which clearly show the diamond-shaped dispersion (indicated by red arrows) formed by the Dirac cone and plasmaron bands.
dispersion formed by the crossing of Dirac cone (blue curve) and plasmaron bands (red). The plasmaron bands are more clearly observed in the 2D curvature plots shown in Fig. 3i–k. A comparison of dispersions measured at high electron doping for 13.5° twisted (Fig. 3f) and 0° aligned (Fig. 3h) graphene/h-BN heterostructures shows that the diamond-shaped dispersion has the same size despite the different twist angle. We further note that the Rb doping does not induce any additional reconstruction, as revealed by the Fermi surface map in the doped sample as shown in Supplementary Fig. 4. Therefore, the diamond-shaped dispersion near the Dirac point is a result of the plasmaron at high electron doping, independent of the stacking angle between monolayer graphene and h-BN and the distribution of Rb atoms.

In the case of magic-angle twisted bilayer graphene where the flat band emerges \( \alpha \gg 1 \), the plasmon dispersion will depend on the twisting angle between the two graphene layers, leading to flat plasmons with a much larger fine structure constant compared to the Dirac cone due to disorder-induced damping and near the Fermi level, the dispersion is dominated by the moiré replica bands (gray curves). In addition, as shown in Fig. 3f, h, the diamond-shaped dispersion formed by the Dirac cone and plasmaron is observed in both aligned and twisted graphene/h-BN heterostructures with the same size, indicating the same strength of electron-plasmon interaction.

**The plasmaron dispersion at the highest electron doping**

To extract the plasmaron dispersion, we show in Fig. 4 the band structure analysis at the highest doping of \( 5.8 \times 10^{13} \text{ cm}^{-2} \) through the K point along the K-K direction. Dispersions extracted from MDCs are appended as blue, red and gray curves in Fig. 4a, b. c MDCs with energy range from \( e_i \) to \( e_f \) shown in b, and the colored marks indicate peaks and are over-plotted in b. d A schematic of electronic structure at the highest doping level. Blue, red and gray colors represent the original graphene Dirac cone, plasmaron bands and the moiré superlattice bands, respectively. e–i Cuts parallel to the K-K direction throughout the momentum range of the diamond shape (indicated by gray lines in d).

**The extracted effective fine structure constant**

The plasmaron dispersion in graphene/h-BN heterostructure provides critical information about the electron-electron interaction and the effective fine structure constant. Although observations of plasmons and plasmarons require large electron concentration, the dimensionless effective fine structure constant \( e^2/\epsilon \hbar v_F \) is directly related to the effective dielectric constant \( \epsilon \) and is independent of the carrier density. In particular, as a result of the unique linear dispersion, the normalized energy separation \( \delta E \) (by the separation between the Dirac point energy and the

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**Fig. 4** Electronic structure at doping level of \( 5.8 \times 10^{13} \text{ cm}^{-2} \) to show the plasmaron dispersion. a, b Electronic structure at the highest doping (\( 5.8 \times 10^{13} \text{ cm}^{-2} \)) through the K point along the K-K direction. Dispersions extracted from MDCs are appended as blue, red and gray curves. c MDCs with energy range from \( e_i \) to \( e_f \) shown in b, and the colored marks indicate peaks and are over-plotted in b. d A schematic of electronic structure at the highest doping level. Blue, red and gray colors represent the original graphene Dirac cone, plasmaron bands and the moiré superlattice bands, respectively. e–i Cuts parallel to the K-K direction throughout the momentum range of the diamond shape (indicated by gray lines in d).
are uniquely determined by the Fermi energy and momentum separations $\delta k$ (by Fermi momentum $k_F$) between the graphene Dirac cone and the plasmaron dispersion is determined by $\alpha_{ee}$\textsuperscript{15,17}. Therefore, the dimensionless effective fine structure constant $\alpha_{ee}$ can be directly extracted by analyzing the diamond-shaped dispersion.

To quantify the energy and momentum separation between the plasmaron bands and the graphene Dirac cone, we show in Fig. 5 a detailed analysis of the plasmaron dispersion upon doping for 0° aligned graphene/h-BN. Figure Sa–e shows the renormalized dispersions with energy $E'$ scaled to $|E_{0f}|$, with $E_{0f}$ defined as the shift of the Dirac point from $E_F$. In the renormalized plot, the diamond-shaped dispersion is positioned at the same energy with similar energy range $E_F$ and momentum separations $\Delta k$ without dielectric screening from the environment, which is an upper limit for $\alpha_{ee}$\textsuperscript{18}. When graphene is placed on a substrate with dielectric constant $\epsilon_s$, the effective fine structure constant is related to the dielectric environment by $\alpha_{ee}\approx e^2/\epsilon_s c^2$, which is directly determined by the dielectric environment. We note that similar spectral features have also been discussed theoretically as satellite bands induced by weakly interacting electron-plasmon interaction\textsuperscript{19}.

Figure Si shows a comparison of experimental $\alpha_{ee}$\textsuperscript{4} values for graphene samples reported so far, including graphene samples grown on Au-, fluoride-, and hydrogen-treated SiC substrate as well as the carbon face of SiC\textsuperscript{17,18}. Our graphene/h-BN heterostructure shows the largest reported $\alpha_{ee}$ value among all graphene samples. At the lowest order approximation, the effective fine structure constant is related to the dielectric constant by $\alpha_{ee}\approx e^2/\epsilon_s c^2$, which is directly determined by the dielectric environment. Therefore, our extracted value of $\alpha_{ee}\approx 0.9\pm 0.1$ gives the lower limit of the effective fine structure constant in graphene/h-BN heterostructure. Without considering the dielectric screening from the valence electrons and the RB atoms, which is quite reasonable considering that $\delta E$ and $\delta k$ are both independent of carrier concentration or amount of RB deposited, the effective dielectric constant is taken as the average value between dielectric constants of materials on both sides of graphene. In the extreme case for free-standing graphene without dielectric screening from the environment, $\epsilon = 1$ and the effective fine structure constant is $\alpha_{ee}\approx 2.2$ (orange symbol in Fig. Si), which is an upper limit for $\alpha_{ee}$. When graphene is placed on a substrate with dielectric constant $\epsilon_s$, the effective fine structure constant depends on the substrate dielectric constant $\epsilon_s$ by $\alpha_{ee}\approx 4.4/\epsilon_s$\textsuperscript{18}, where the effective dielectric constant $\epsilon$ is taken as the average between the vacuum $\epsilon_{vac} = 1$ and $\epsilon_s$. The fitting function of $\alpha_{ee}\approx 4.4/\epsilon_s$ is also plotted in Fig. Si. From this relation, the extracted $\alpha_{ee}\approx 0.9$ for graphene/h-BN gives an
effective dielectric constant of $\epsilon \approx 2.5$, which corresponds to
substrate dielectric contribution $\epsilon_{s} \approx 4$, similar to the reported
substrate dielectric constant of $\epsilon_{s, BN} \approx 3$\textsuperscript{26,34}. Considering the dielectric constant of h-BN, the large fine structure constant of graphene/h-
BN is not surprising, however, being able to experimentally observing it is still an important experimental progress.

DISCUSSION
In summary, we report the experimental evidence of plasmaron and extract the fine structure constant of graphene/h-BN. We note that experimental values for the effective fine structure constant have been reported through optical transparency\textsuperscript{5} and inelastic x-ray scattering measurement\textsuperscript{5,30} on both graphene and graphite, and the fitting of the Dirac point velocity\textsuperscript{9}, which involves both effects of the carrier screening and dielectric screening. Here by observing not only the Dirac cone but also the dispersion of the previously inaccessible plasmarons in graphene/h-BN at high electron density, we extract the dressed effective fine structure constant $\alpha'_{\text{ep}} \approx 0.9$. Such large effective fine structure constant reveals the important role of the small dielectric constant of h-BN in reducing the dielectric screening. In addition, the dispersions remain quite sharp (Fig. 2c-h) under the presence of such a large number of carriers and strong plasmon-charge interaction, suggesting the insignificant contribution of both scattering channels in the electron scattering. Since many device applications require tunable electronic density, our finding on the small scattering of a highly electron-doped graphene/h-BN provides useful information for applications in gate-tunable nano-electronic and nano-plasmonic applications\textsuperscript{28,40}.

In the past decade, h-BN has been widely used as a substrate and a capping layer, for example, in magic-angle twisted bilayer graphene or ABC stacking trilayer graphene on h-BN, both exhibiting Mott insulator\textsuperscript{7,41} and superconductivity upon dop-
ing\textsuperscript{36,42}. Revealing the effect of dielectric property of h-BN on the effective fine structure constant of graphene can also be helpful for understanding the electron-electron interaction in graphene/h-
BN. Finally, it has been suggested that the effective fine structure constant is relevant to other Dirac systems\textsuperscript{50} including topological insulator surface states\textsuperscript{35,44}, and Dirac or Weyl materials\textsuperscript{45,46}. Therefore, our results on graphene/h-BN hetero-
structure in principle can be extended to other Dirac materials for evaluating the electron-electron interaction and the effective fine structure constant.

METHODS
Sample preparation
Sample preparation of the 0° aligned graphene/h-BN sample. Single crystal h-BN flakes were first exfoliated onto a 300 nm SiO$_2$/Si substrate by mechanical cleaving method. Graphene samples were directly grown on h-BN substrates by the epitaxial method, as specified in previous work\textsuperscript{14}. As-grown samples were characterized by tapping mode atomic force microscopy at room temperature in ambient atmosphere. We used freshly cleaved mica as shadow masks for metal electrode deposition. The contact metal (2 nm Cr on 90 nm Au) was deposited on the non-mica-covered area with a small part of target graphene/h-BN samples. The samples were then annealed at 200 °C, after removing the mica flakes.

Sample preparation of the 13.5° twisted graphene/h-BN sample. Firstly, h-BN flake was exfoliated onto a PDMS stamp. Then the graphene flake on a SiO$_2$/Si substrate was picked up sequentially with h-BN on PDMS, and the twist angle between graphene and h-BN was determined by the edge of the flakes. Subsequently, the graphene/h-BN structure was flipped over and picked up with a second PDMS stamp and then transferred onto the gold-plated substrate. Finally, a piece of graphite was placed to connect graphene and gold to make sure the conductivity.

ARPES measurements
ARPES measurements were performed at beamline 4.0.3 and 12.0.1 of the Advanced Light Source at Lawrence Berkeley National Laboratory (LBNL). The optimal spot size was set to 30 µm. The data were recorded with photon energies of 50 and 60 eV. The overall energy and angle resolution are better than 26 meV and 0.1°, respectively. Before measurements, the samples were annealed at 200–300°C until sharp dispersions were observed. All measurements were performed below 20 K and under a vacuum better than $5 \times 10^{-11}$ torr. The Rb deposition was achieved by heating an SAES commercial dispenser in situ.

DATA AVAILABILITY
The data that support the findings of this study are available from the corresponding author upon reasonable request.

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**AUTHOR CONTRIBUTIONS**

S.Z. designed the research project. Hongyun Z., E.W., C.B., K.D., H.Z., A.F., J.D. and S.Z. performed the ARPES measurements and analyzed the ARPES data. S.W., X.L., Q.L., G.C. and G.Z. prepared the graphene samples. K.W. and T.T. prepared h-BN crystals. Hongyun Z. and S.Z. wrote the manuscript, and all authors commented on the manuscript.

**COMPETING INTERESTS**

The authors declare no competing interests.

**ADDITIONAL INFORMATION**

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