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Phase-shift of the wavefunction on the scattering probed by the angle-resolved photoelectron spectroscopy: Resonant replicas of the Dirac cone on graphene/SiC

The Dirac cone of graphene is located at the K-point of the Brillouin zone in the k-space. One can take the polar coordinate sytem around the Dirac point for discribing the wavefuntion of the Dirac cone, and the energy is approximately poportional to the distance from the Dirac point, and nothing to do with the angle θ. Therefore, θ can be regareded as "phase" of the wavefunction of the graphene near the Fermi level. It is known that the photoelectron intensity from single-layered graphene is proportional to $\cos^2\frac{(\theta-\delta)}{2}$ in which δ is an angle where the maximal intensity is achieved, and can be analyzed by angle-resolved photoelectron spectroscopy (ARPES) with the polarized photon beam [1]. Meanwhile, it is known that the duplication of the Dirac cone occurs in the ARPES spectra of the epitaxial graphene, and is of interesting since it gives a crucial information for the modification of the epitaxial graphene. On graphene/SiC, replicas of the $(6\sqrt{3}\times 6\sqrt{3})R30^\circ$ periodicity of the buffer layer is well known[2], and recently, those of the SiC-lattice periodicity are also reported [3]. In these researches, the result is well explained by the photoelectron diffraction at the buffer layer and SiC substrate, then δ in the phototelectron-intensity distribution is preserved during the diffraction. In this paper, we found the strong resonant enhancement in intensity of the replicas at the photon energy of 11eV, and more interstingly, the photoelectron-intensity distribution does not preserved in the photoelectron diffraction. This may indicate that the phase-shift of the wavefunction during the scattering is possible to probe experimentally.

The experiments were carried out at the BL-9A, an ARPES beamline, of the synchrotron radiation facility at Hiroshima University (HiSOR). The sample used was single-layer graphene epitaxially grown on SiC surface, which was cleaned by heating at 700K in UHV and the measurements were made at 10K. Figure 1 shows the photoelectron intensity-map at the Fermi level of graphene/SiC derived by ARPES with the p-polarized photon of 10.8eV. The photon incident plane, which is parallel to the horizontal axis in Fig.1, is set along the Γ-K line of the graphene crystal. Due to the low photon energy, all the Dirac cones observed here are replicas (the K-point is located at k_x=$1.7\mathrm{A}^{-1}$ which is out of the area). The intensity of these replicas are strongly enhanced when the photon energy is around 11eV as shown in the inset of Fig.1. Solid red dots show the simulation for the scattering from the Dirac cone at the K-point at (k_x,k_y) of (1.7,0) and other equivalent two K-point with the combinations of primitive reciprocal graphene-lattice vectors ***G***'s and the primitive reciprocal SiC- lattice vectors ***S***'s (In other words, the K-point at the second Brillouin zone is taken into account). Solid blue dots show those from the Dirac cone at the other three K'-points. Triangles, stars, and small filled circles denote single-, double- and triple scatterings, respectively. Simulation using the scattering vector of the buffer-layer-lattice could not reproduce the experimental results at all, and it is concluded that the photoelectron diffraction occurs at the SiC substrate. Figure 2 shows the schematic model of the scattering and photoelectron-distribution of the Dirac cone replicas. Note that replica A is the result of the scattering from the K-point in the first Brillouin zone but B-D are from those in the second Brillouin zone.

Most peculiar behavior of these replicas is that δ in the intensity distribution is sometimes not preserved on the scattering contradictory to the case of the non-resonant photoexcitation previously reported[3]. In the case of replicas A and B, δ does not change, and the intensity distribution of the replicas are similar to the original K-points from which the photoelectron of the Dirac-cone is scattered. On the other hand, the intensity distribution rotates by 2/3π during the scattering into the replica C, and -2/3π for the replica D. As a result, a 4-fold symmetry is observed in the intensity distributions in the whole replicas while it is of a 6-fold symmetry at the original Dirac cones. We found that the rotation angles depend not only on the scattering vector but also on the polarization and the incident plane of the photon beams (Γ-K or Γ-M) on the graphene crystal. Thus, it is obvious that the process of the replica formation is drastically different in the resonant-photoexcitation to 11eV state from that in the non-resonant photoexcitation. There are two candidates to interpret this behavior. One is that the final state of the 11eV above the Fermi level is of completely different symmetry from the non-resonant case, and the photoelectron intensity-distribution (that cannot be experimentally examined since the kinetic energy of the photoelectron is too small at this photon energy) is completely different, and the phase is preserved during the scattering. However, even in the non-resonant excitation into a state of higher than 20eV, it is claimed that the final state is not the pure free-electron state but the graphene wavefunction [4]. Thus, it is required to assume the final state at 11eV is completely different from the ordinal unoccupied state; which is somewhat strange. The other candidate is that the phase-shift occurs during the photoelectron-diffraction, which is commonly accepted, and this can be observed only at the resonant photoexcitation case. If it is the case, the phase-shift of the wavefunction could be measured by using ARPES. However, this requires the theoretical formulation of the coherent process of photoexcitation and diffraction, and more work is needed to achieve a definite conclusion.

References

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Figures

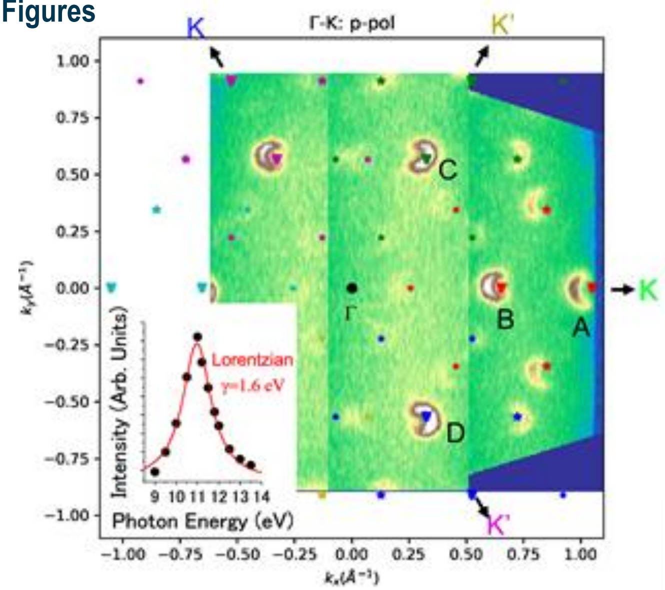

Figure 1: Intensity map of ARPES at the Fermi level taken at hν=10.8eV of p-polarized light whose incident plane is along the Γ-K line. Some of the replicas are indicated as A-D. Blue and red marks shows the simulation of the diffraction assuming the scattering vectors of the SiC-substrate. The photon-energy-dependence of the intensity of the replica is inserted.

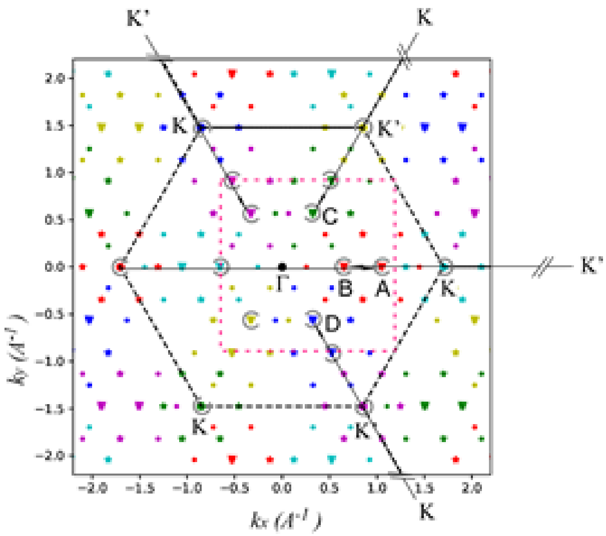

Figure 2: Schematic model of the replicas of Dirac cone in k-space in Fig.1. Red dotted rectangle approximately shows the area observed in Fig.1. Colored marks show the simulation of the scattering by Si-C lattice vector, and marks of A-D corresponds to A-D in Fig.1 Intensity-distributions in the replicas are shown schematically. One can recognize the the original Dirac cone from which the replica is provided with the color of the mark. Arrows indicate the scattering vectors, some of which is comes from the K-point of the second Brillouin zone.