

博士論文（要約）

Topological valley current in bilayer graphene
with broken inversion symmetry

（反転対称性の破れた二層グラフェンにおける
トポロジカルバレー流の研究）

島崎 佑也

An electron has well-defined quantum numbers; charge and spin. Spintronics, which is the technology based on manipulation of the spin degree of freedom, has been intensively studied, leading to development of various kinds of spin effects such as spin current and spin Hall effect. On the other hand “valleytronics” which utilizes the valley degree of freedom has more recently been proposed and is now beginning to be experimentally addressed. In certain solid crystals, energetically degenerate but non-equivalent local band structures called valleys exist. Graphene and transitional metal dichalcogenides (TMDCs) are two typical 2D material systems having the valleys. Both materials are processed into devices for studying the electrical properties using a mechanical exfoliation technique. Graphene and TMDCs have two-component valley degree of freedom (K or K') and are considered to be promising platforms for valleytronics owing to valley dependent Berry phase and accompanying valley contrasting phenomena. The most important phenomena in valleytronics are valley Hall effect and its inverse effect that are analogous to spin Hall effect and its inverse effect in spintronics. These effects enable electrical generation and detection of pure valley current, which is a counter flow of the K-valley and K'-valley electrons. Similar to pure spin current, this pure valley current accompanies no net charge current and is a promising information carrier for non-dissipative electronics. The important requirement to realize the Berry phase driven valley contrasting phenomena is that the system has broken inversion symmetry. The monolayer semiconducting TMDC materials such as MoS₂ and WSe₂ have broken inversion symmetry in themselves. Indeed valley Hall effect has recently been demonstrated in monolayer MoS₂ by opto-electrical measurement. Compared to TMDC materials, graphene has less crystal defects which cause inter-valley scattering, and is therefore more appropriate to study the valley current transport. However, pristine graphene has inversion symmetry and generates neither valley Hall effect nor its inverse effect before the inversion symmetry is broken in some way. One of the ways is to use hexagonal BN (h-BN) as a contact layer. h-BN has staggered honeycomb lattice consisting of Boron and Nitrogen atoms and has often been used as high quality insulating substrate for graphene. When the crystal orientation of monolayer graphene is carefully aligned to that of h-BN, the potential from the h-BN substrate breaks inversion symmetry of monolayer graphene to open up the band gap. Using such an aligned monolayer graphene/h-BN stack, both valley Hall effect and valley current transport have been observed. The other is to apply an external electric field to break the inversion symmetry. For the case of AB-stacked bilayer graphene, a perpendicular electric field induces a potential difference between the two layers. This potential difference effectively works as a staggered potential to break the inversion symmetry. In this scheme, the sign of the valley Hall conductivity (or Berry curvature, which works as an effective magnetic field) and the size of band gap are varied as a function of perpendicular electric field. Note that this is not possible in the above mentioned way because the inversion symmetry is structurally broken and both the sign of the valley Hall conductivity and the size of the band gap are fixed after the device fabrication. Furthermore, there is an advantage that crystal orientation

alignment is not necessary in this scheme. Therefore this scheme is more relevant for mass production. However, the valley current transport in the band gap still remains to be revealed experimentally. It has been pointed out that gapped bilayer graphene can be viewed as a “marginal topological insulator”, where the existence of the edge states is significantly affected by the edge properties, and usual bulk-edge correspondence does not hold. Nevertheless, the bulk valley Hall conductivity remains non-zero in the band gap. This raises a profound question: If a marginal topological insulator without edge states can host a bulk topological current or not in its gap. It is therefore still challenging to reveal valley Hall effect or topological valley current in the insulating regime. The higher controllability of bilayer graphene should reveal the nature of bulk topological current in the gap, which has not been discussed in the study of usual topological insulators with edge states. In this thesis, we studied topological valley current transport in bilayer graphene with broken inversion symmetry. We utilized a dual-gate structure to independently control the perpendicular electric field or displacement field and the carrier density. The bilayer graphene is encapsulated between two h-BN insulating layers to reduce the inhomogeneity of the device as much as possible. Both graphene and h-BN are prepared by the mechanical exfoliation technique and are obtained as flakes whose size is typically several tens of micrometers. We stack these flakes together manually under an optical microscope to make the encapsulated structure. Here we have developed specific transfer techniques to realize such an encapsulated device structure with high quality. To detect the valley current transport, we measured local resistivity and nonlocal resistance using a Hall bar device having several sets of current or voltage terminals attached to a central channel. The valley current mediated nonlocal transport is understood in the following way: Via valley Hall effect, the injected current between two terminals across the central channel generates a valley current along the channel but away from the current flowing part. After propagating several micrometers, this valley current is detected by measuring voltage between two terminals across the central channel via inverse valley Hall effect. Nonlocal resistance, which is defined as the detected voltage divided by the injected current, is a measure of valley current transport. We evaluated the displacement field and the carrier density dependence of the nonlocal resistance from 1.5K to 200K. By breaking inversion symmetry with applying the displacement field, a huge nonlocal resistance emerges. This resistance is 3 orders of magnitude larger than that expected for contributions from the trivial current diffusion. To discuss the origin of the observed nonlocal transport, we developed simple analytical models by assuming bulk valley current mediated transport or edge transport. We find that the difference in the nonlocal transport mechanism leads to the difference in the scaling relation between the local resistivity ρ and the nonlocal resistance R_{NL} . In the case of bulk valley current mediated nonlocal transport, we expect a cubic scaling relation of $R_{\text{NL}} \propto \rho^3$ at the charge neutrality point for the case of a small valley Hall angle, which is defined as the ratio of valley Hall conductivity to the conductivity and corresponds to the conversion efficiency from the charge current to the valley current.

On the other hand, nonlocal transport also happens due to edge transport. In this case, we can readily derive a linear scaling relation between the 4 terminal local resistance and nonlocal resistance at the charge neutrality point by neglecting shunting effect by bulk residual conductivity. We found that the observed nonlocal transport originates from the bulk valley current transport by evaluating the $R_{\text{NL}} - \rho$ relation in the following way. The $R_{\text{NL}} - \rho$ scaling relation was obtained from both measurements of the displacement field dependence and temperature dependence of R_{NL} and ρ at the charge neutrality point. In both measurements we observe the cubic scaling relation of $R_{\text{NL}} \propto \rho^3$ in the smaller resistivity regime, which is considered to be the band transport regime. This cubic scaling relation is also detected for both polarities of the displacement field. These findings strongly support the observed is valley current mediated nonlocal transport. For both local resistivity and nonlocal resistance at the charge neutrality point we realize that the temperature dependence shows a clear activation behavior and find that the obtained relation between these two activation energies is consistent with that predicted for valley current transport. From the cubic scaling relation, there is a relation between the activation energy of local resistivity (E_{L}) and that of nonlocal resistance (E_{NL}), that is $E_{\text{NL}} = 3E_{\text{L}}$. Actually the displacement field dependence of E_{NL} and E_{L} shows the relation of $dE_{\text{NL}}/dD = 3dE_{\text{L}}/dD$. This supports the cubic scaling relation, of the thermally activated valley transport although there appears an offset of E_{NL} . Depending on the temperature, both the local resistivity and nonlocal resistance show a crossover feature between the high and low temperature regime. The crossover of the local resistivity is attributed to that of the transport mechanism between the band conduction at high temperatures and hopping conduction at low temperatures. For the larger displacement field, the crossover temperature of local resistivity and nonlocal resistance coincide, indicating the correlation between the crossover of the conduction mechanism (band conduction or hopping conduction) and the crossover of the nonlocal transport. On the other hand for the hopping conduction, the $R_{\text{NL}} - \rho$ relation deviates from the cubic scaling. The reason for this has yet to be revealed, but we realize so analogy with the anomalous Hall effect. A crossover behavior of anomalous Hall conductivity between the band conduction and hopping conduction has been reported in various materials. Magnetic field dependence of the nonlocal transport is also evaluated at 70K, where quantum Hall effect is not observed. There are two worth noting features. Even for zero displacement field, a large nonlocal resistance emerges around the charge neutrality point by applying a perpendicular magnetic field. This behavior still remains to be understood, while a similar behavior is observed for monolayer graphene without broken inversion symmetry where spin/valley splitting induced spin/valley Hall effect is pointed out as a possible origin. When a high displacement field is applied, nonlocal resistance keeps increasing as the perpendicular magnetic field is increased, even though the local resistivity changes only slightly. This feature also remains to be revealed. In conclusion, we observed the topological valley current transport in bilayer graphene with broken inversion symmetry for the first time. This is evidenced by the observation of the cubic

scaling between the local resistivity and the nonlocal resistance at the charge neutrality point. The relation between the local and nonlocal resistance activation energies is also consistent with this observation. In addition, we found a crossover behavior of the nonlocal transport and the peculiar magnetic field dependence of the nonlocal transport, while some parts of these findings still remain to be revealed. Our study on the topological valley transport in highly controllable bilayer graphene system makes a breakthrough for further study on graphene-based valleytronics and topological transport in honeycomb lattice systems.