A rocky pathway through a valley

The Valley Hall Effect in MoS₂ Transistors

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Recommended with a commentary by Anton Akhmerov, Delft University of Technology.

An important distinction between the conventional quantum wells and the graphene-like two dimensional crystals with honeycomb lattice is that instead of a single Fermi surface the latter have two isolated valleys. They are positioned in the opposite corners of the Brillouin zone and are related to each other by time reversal symmetry. The first observation of relativistic dispersion of single layer graphene [1] was also a solid proof of the existence of valleys, apparent from the $4(n + 1/2)e^2/h$ quantization of the Hall conductance, and the corresponding four-fold (2 spin × 2 valley) degeneracy of the Landau levels. Manipulation to the valley degree of freedom (dubbed "valleytronics" [2]) has however proven to be hard in graphene. If implemented naively, it requires unprecedented control of the lattice structure on the scale of a single atom, something far beyond the current experimental capabilities.

A recent successful approach for selectively coupling to a single valley was demonstrated in a single layer molybdenum disulfide. Unlike graphene, MoS₂ strongly breaks inversion symmetry: one of the sublattices of its honeycomb lattice is a single molybdenum atom, while the other one consists of two sulphur atoms on top of each other. The result of the inversion symmetry breaking is the opening of the 1.9 eV band gap in both valleys. A neat consequence of the fact that valleys are centered at the K and K' corners of the Brillouin zone and of its orbital structure is the appearance of a new selection rule: a circularly polarized light may only create electron-hole pairs in one valley, but not in the other. The selection rule appears due to the interplay between the orbital structure of the wave functions at the bottom of the conduction band and the top of the valence band together with the structure of the Bloch wave functions becoming eigenstates of the $2\pi/3$ rotation in the corners of the Brillouin zone [3]. More specifically, the states at the bottom of the conduction band have a total azimuthal quantum number $l = \pm 1$ and can only be coupled to the l = 0 states at the top of the valence band by light with matching polarization. The selective coupling of circularly polarized light to a single valley was successfully demonstrated in Ref. [4], by showing that circularly polarized light causes photoluminescence of MoS₂ with the same polarization. A bilayer MoS₂ has

inversion symmetry, so that the selection rule is no longer effective, and accordingly exhibits no polarized signal in photoluminescence.

The work of McEuen group that appeared on arXiv in March 2014, arXiv:1403.5039, makes the next step towards valleytronics. Like their predecessors they use circularly polarized laser light to selectively excite carriers only in a single valley. However, beyond merely selectively exciting carriers in one valley, the researchers showed that the two valleys have different properties. While the total Hall conductance of MoS_2 must be equal zero in the absence of magnetic field, this zero is composed out of equal magnitude and opposite sign contributions originating from two valleys. This means that if carriers only in a single valley are excited, they will experience a small but non-vanishing effective magnetic field that will generate a corresponding Hall voltage.

The Hall voltage was indeed observed by McEuen group, and it satisfied all the necessary sanity checks. Finite Hall conductivity only appeared when the photon energy was sufficient to excite the quasiparticles across the band gap. The magnitude of the voltage was proportional to the degree of circular polarization of the incoming light. Finally, the Hall voltage also vanished in a bilayer, unambiguously proving that it is only developed due to the imbalance of the valley populations.

Personally, I find one aspect of this preprint the most intriguing. When considering MoS_2 and other dichalcogenides one must keep in mind that these materials are not simply graphene with different Hamiltonian parameters. Graphene now starts competing with quantum wells in purity and quality, with its mobility approaching a million cm^2/Vs and even the early graphene experiments had mobilities around ten thousand. On the other hand, MoS_2 used in the Cornell experiment only had mobility below a hundred, and this is not anyhow atypical. This means that the ideal picture of band structure, Berry curvature and the optical selection rules is only a crude starting point for understanding what does actually happen. If a lot of disorder originates from atomic scale defects, it should cause intervalley scattering and reduce the effectiveness of the selection rules as well as the Hall conductance. Understanding what is actually happening requires an additional investigation both theoretical and experimental. The observed quick drop of Hall conductance at laser frequency above the resonace suggests strong intervalley relaxation, while the Hall voltage on resonance matches well the theoretical expectations corresponding to perfect valley polarization of the excited carriers. The combination of the conventional conductance measurements with the anomalous Hall response should allow to disentangle different scattering mechanisms in this material and to answer the following open questions. What are the relevant sources of scattering present in MoS_2 , and what are their relative strengths? Looking further into future, could better understanding of the scattering in MoS_2 explain what are the next steps towards the long standing goal of valleytronics?

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