Photo-assisted modulation of thermal transport and thermopower in a single-layer transition metal dichalcogenide

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Transition metal dichalcogenides (TMDCs) have the representative formula MX_2, where *M* is a transition metal element from group IV–VI and *X* belongs to the set of elements S, Se, and Te, collectively identified as chalcogens [1]. They are layered materials of covalently bonded atoms held together by weak van der Waals forces. Thin TMDC films are considered promising thermal materials with the possibility of a large figure of merit, $ZT = S^2 \sigma T \kappa^{-1}$. Here, ***S*** is the Seebeck coefficient, the electrical (thermal) conductivity is denoted by $\boldsymbol{\sigma}$ ($\boldsymbol{\kappa}$), and ***T*** is the temperature. Beginning with a *k.p* representation [2] of the Hamiltonian that describes the carriers in the vicinity of the two valleys, ***K*** and ***K`*,** as massive Dirac fermions, we theoretically demonstrate the modulation of ***S*** aided by a driven periodic perturbation. We use a high-intensity circularly-polarized illumination to drive the TMDC into a Floquet *off-resonant* phase [3] that enlarges the fundamental band gap, say, at ***K***, while ***K`*** suffers an equal reduction. This dual transformation shown in Fig. 1 is simply an outcome of the time-reversal principle connecting the two valleys and manifests as unequal conductivities for respective carriers. This inequality is also mirrored in their thermopower (***S***) behaviour. The conductivity (intra-band) calculations are performed using the Kubo formalism. To determine the thermopower for carriers (that lie close to *K* and *K`*), in the low-temperature limit, we use Mott's formula furnishing a valley-resolved thermopower. Specifically, the carriers from the valley that has an optically-lowered band gap (Fig. 2) reveal a higher thermopower vis-à-vis the ensemble located in its time-reversed counterpart [4]. Further, a simple application of the Wiedemann-Franz law (WFL) relates σ to κ from which we obtain low-temperature *ZT*, using the pre-computed thermopower.

The thermal conductivity is integral to Peltier-type solid state cooling methods and thermopower generation; while the former requires a higher κ for unimpeded heat flow from deep-seated hot-spots in a miniaturized chip, the latter relies on localized heat production for efficient energy-conversion. To this end, in accord with WFL, to achieve a desirable κ, it is prudent to investigate conditions that permit an adjustable σ. We show that in conjunction with optical modulation, disorder, which can under appropriate concentration establish a variable hopping regime and quench σ, allows the sought control over κ within the purview of our stated applications.

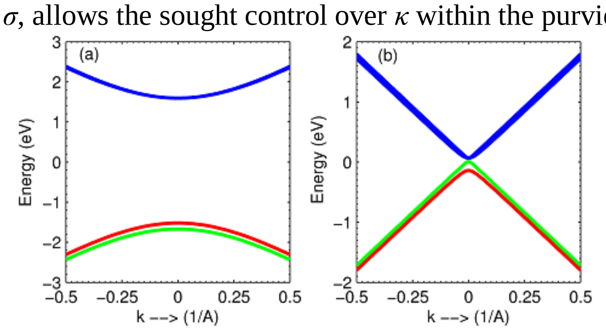

Figure 1: The dispersion of monolayer MoS_2 under *off-resonant* condition. The *K* (*K`*) valley band gap on the left is enlarged (shrunk) when transformed into the Floquet phase. The frequency of the light beam corresponds to 10.0 eV.

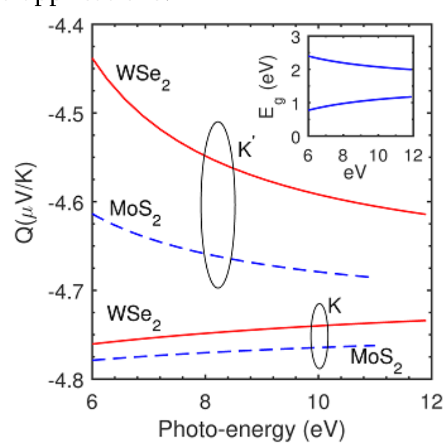

Figure 2: The low-temperature (at T = 10 K) valley-resolved longitudinal thermopower (*Q*) for MoS_2 and WSe_2. The lower band gap at *K`* from a right-circularly polarized photo-illumination gives a higher *Q* compared to the *K* valley. In general, the lower band gap of $WSe_2$ offers a higher *Q*. The inset shows the progression of band gap at *K* (upper curve) and *K`* for a range of illumination energies that fulfill the *off-resonant* conditions in a Floquet phase.

1. Wang Q H, Kalantar-Zadeh K, Kis A, Coleman J N and Strano M S, *Nat. Nanotechnology.* **7** 699 (2012).
2. Xiao D, Liu G, Feng W, Xu X and Yao W, *Phys. Rev. Lett.* **108** 196802, (2012).
3. Kitagawa T, Oka T, Brataas A, Fu L and Demler E, *Phys. Rev.* B **84** 235108 (2011).
4. Sengupta P, Tan Y, Klimeck G and Shi J, *Jour of Phys. Cond Matt*, **29**, 405701 (2017).

Supplementary material

In the one-page submission, we described an optical technique to modulate the thermopower and the thermal conductivity. The thermal conductivity was also found amenable to disorder (see Fig. 3 for a comparison between pristine and disordered case) through corresponding changes to the its electrical counterpart. These studies could also be potentially extended to material systems, for instance, ultra-thin films of 3D topological insulators and graphene-like silicene and stanene that host massive Dirac fermions of the kind found in single-layer TMDCs around the *K* and *K*` valley edges. Silicene and stanene [1] can carry heavy fermions via a gate-applied electric field while the helical states of a 3D TI (for example, Bi_2Te_3,) under surface hybridization in an ultra-thin film change from a zero-mass linear set of bands to massive Dirac hyperbolas. In principle, we can observe a quantitative change of thermopower in these materials and the presented results would differ insofar as band/Hamilton parameters are concerned. It is pertinent to note though that since most room temperature 3D TIs are characterized by a single Dirac cone, a valley-resolved quantity is not possible unlike in multi-valleyed silicene and stanene. From a theoretical standpoint, such light-matter interactions can bring about significant topological phase changes [2] primarily by creating a band gap (the Floquet topological insulator phase) to transform a zero-gapped material (as in the surface states of a 3D TI) into a trivial insulator. Such microscopic rearrangements can be observed from the emitted photo-electrons (in an ARPES experiment) which mirror the change in fundamental Chern numbers as the dynamically-driven material undergoes topological phase transitions [3].

Additionally, we must mention here that the described modulations are not just limited to thermopower and thermoelectric effects but can be used for a variety of applications with appropriate optical sources. For instance, it is possible to demonstrate an intriguing rise in anisotropy with far-reaching effects in another widely researched two-dimensional (2D) material, the multi-layered black phosphorous (BP). When doped with potassium, four-layered BP [4] with a Dirac crossing at Γ (at an energy of 0.18 eV) has a hybrid dispersion of parabolic/quadratic (along the zigzag) and linear (along armchair) branches imparting a highly anisotropic character which is further augmented (Fig. 4) by driving it into a Floquet phase (as before, the periodic field is a light source) through introduction of an extra linear term. This time-dependent contribution gives rise to a subtle inter-play [5] between the linear and quadratic terms manifesting as a change in the density of states (DOS) and measured using a simple quantum capacitance technique (Fig. 5) in prototype field-effect transistor setup (Fig. 6) with dual gates, one of which is optical and the other a conventional metal-type. Quantum capacitance measurements exactly map the anisotropy of the DOS and electronic dispersion. The DOS, in turn, greatly determines the heat response pointing to a similar microscopic origin to the modified thermopower in single-layer TMDCs.

In essence, the genesis of such exotic realizations lie in the photo-induced fundamental change to the topology of electron states either through band gap modification or altered arrangement of energy levels. In our case, the single-layer TMDCs, the electrons located close to the valley edges carry a unique topological marker, the finite Berry curvature (the momentum-space analog of magnetic field) which governs the interaction with the periodic electromagnetic field of light as revealed in their amended response.

1. Liu C, Wanxiang F, Yugui Y, *Phys. Rev. Lett*. **107** 076802, (2011).
2. Ezawa M, *Phys. Rev. Lett.* **110** 026603, (2013).
3. Gavensky L, Gonzalo U, Balseiro C, *Scientific Reports* **6**, 36577 (2016).
4. Kim J, Baik S, Ryu S, Sohn Y, Park S, Park B, Denling J *et al. Science* **349**, 6249, (2015).
5. Sengupta P, Rakheja S, preprint*: arXiv*:1708.02350 (2017).

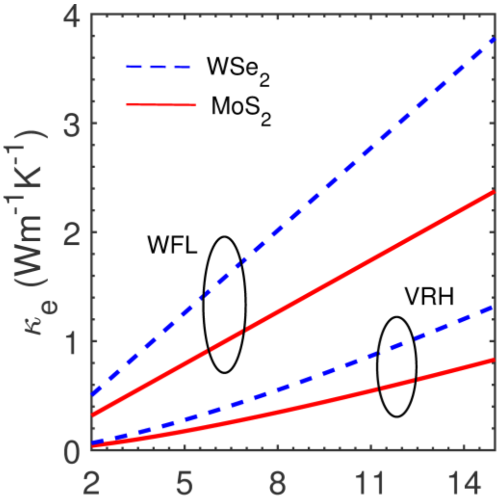

Fig. **3**: The low-temperature thermal conductivity for two cases is shown. The curves marked as `WFL' are obtained by a straightforward application of the Wiedemann-Franz law; the other group denoted by `VRH' pertains to the state when variable range hopping is active and modifies the result of WFL. The two semiconducting TMDCs are MoS_2 and WSe_2 (dashed line).

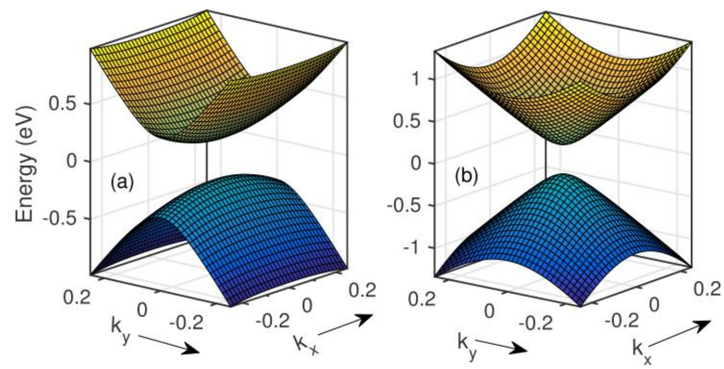

Fig. **4**. The numerically calculated electronic dispersion of a four-layered BP in shown in the left panel (a) where the bands appear distinctly parabolic. However, under an intense light beam the bands are rearranged and acquire a linear character (b) as revealed in the funnel-like shape. For visual clarity, we have artificially set the power of the beam to a large value of 5.0 eV while the frequency corresponds to an optical source of energy 7.0 eV. The *k*-components are in 1/A. This behaviour is in marked contrast to that of a single-layer TMDC which shows a band gap alteration but not a fundamental re-arrangement of states around the valley edge.

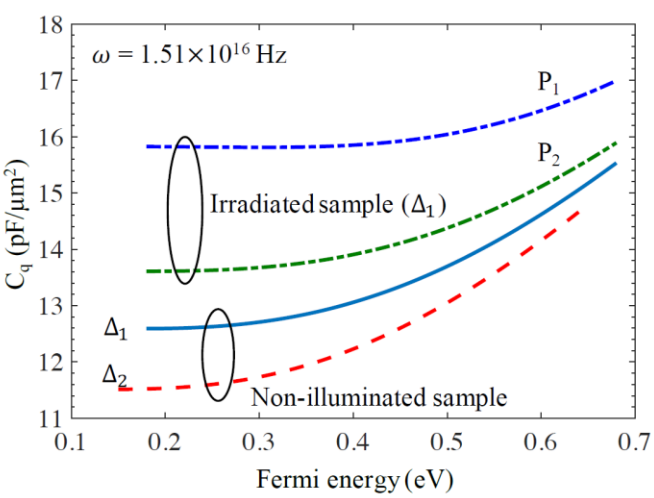

Fig. **5**. The numerically computed quantum capacitance (C_q) in vicinity of the Dirac crossing (0.18 eV) is shown for non-illuminated and irradiated four-layered gapped BP (*K*-doped) sample. Two sets of photo-illumination with different powers, P_1 = 1.0 eV and P_2 = 2.0 eV but identical frequency (ε = 10 eV) modulate the quantum capacitance. The non-illuminated quantum capacitance is lower for a smaller band gap (Δ) Here, Δ_1 = 0. 18 eV and Δ_2 = 0. 15 eV. This is another instance of light-induced change to an experimentally observable quantity.

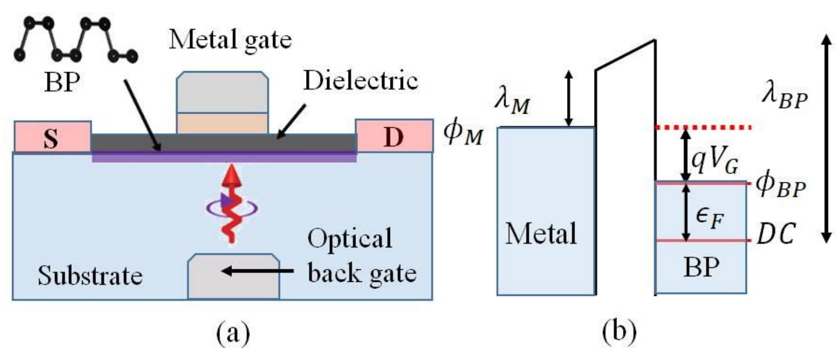

Fig. **6**. The left panel (a) shows the schematic of a metal-gated (V_g) four-layered *K*-doped BP (with a puckered unit cell sketched in black) device with source (S) and drain (D) contacts. The optical back gate supplies the external time-dependent perturbation to drive BP into Floquet phase The energy edges are drawn in (b) where ε_f is the Fermi energy measured from the Dirac crossing. The electrochemical potential of the metal (BP) layer is Φ_M (Φ_{BP}) while the work functions are identified by λ.