

Abstract

Valleytronic applications utilize the valley degree of freedom, the location of a state's wavefunction within the Brillouin zone, to store and process information. In this context, the valley Hall effect is important for reading and writing the valley state. So far, research on this effect has focused on its linear response to an applied current and has not considered nonlinear responses. Here we report the observation of a nonlinear valley Hall effect in a graphene moiré superlattice, indicated by the generation of second-harmonic non-local voltages under a.c. currents. **The nonlinear effect we observe has a magnitude surpassing the linear version** and is highly tunable with a gate voltage. The nonlinear signal shows quadratic scaling with driving current and quartic scaling with local resistance, setting it apart from its linear counterpart. We further reveal a nonlinear inverse valley Hall effect by observing the third- and fourth-harmonic non-local voltages. This effect provides a mechanism for valley manipulation and may enable a **valley rectifier** device that converts a.c. charge current into d.c. valley current.

Device Characterizations

- ◆ High-quality graphene–hBN moiré superlattices enable a tunable gap and Berry curvature.

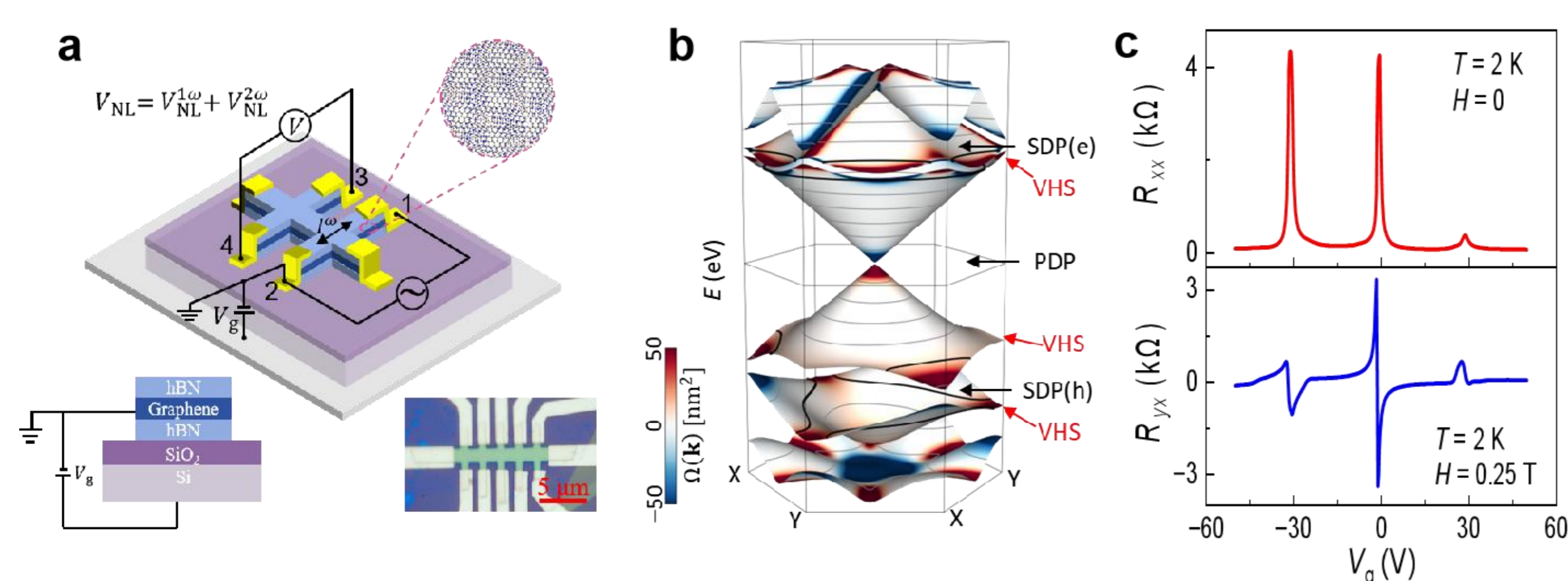


Fig. 1 **a**, Schematic of the device structure of hBN-encapsulated monolayer graphene and the non-local electric harmonic measurements. **b**, Band structure of graphene–hBN moiré superlattices. **c**, The longitudinal resistance R_{xx} (top) and Hall resistance R_{yx} (bottom) as functions of gate voltage V_g . The SDPs emerge at $V_g = -31$ V and 29 V.

VHE and nonlinear VHE

AC electric field E_y^ω applied at middle Hall arms induces a (nonlinear) valley current $J_x^{v,\omega}$ ($J_x^{v,2\omega}$) in the transverse direction via (nonlinear) VHE. This valley current is converted into a nonlocal (nonlinear) charge current via inverse VHE at the left and right arms, producing $V_{NL}^{1\omega}$ ($V_{NL}^{2\omega}$).

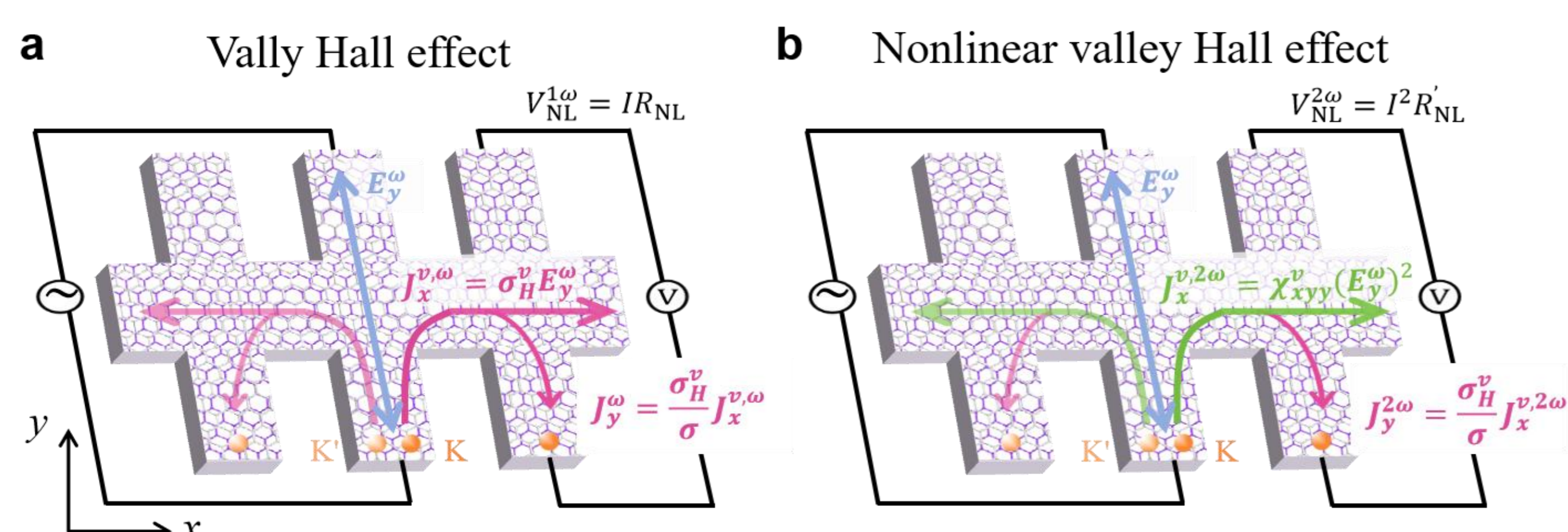


Fig. 2 Schematic of the VHE (a) and nonlinear VHE (b) and the non-local measurement via the inverse VHE.

Observation of nonlinear VHE

- ◆ $V_{NL}^{2\omega}$ exhibits a pair of opposite peaks and changes sign across each Dirac point region.
- ◆ $V_{NL}^{2\omega}$ can indeed be larger than $V_{NL}^{1\omega}$ at $I > 2 \mu A$ near the Dirac points.

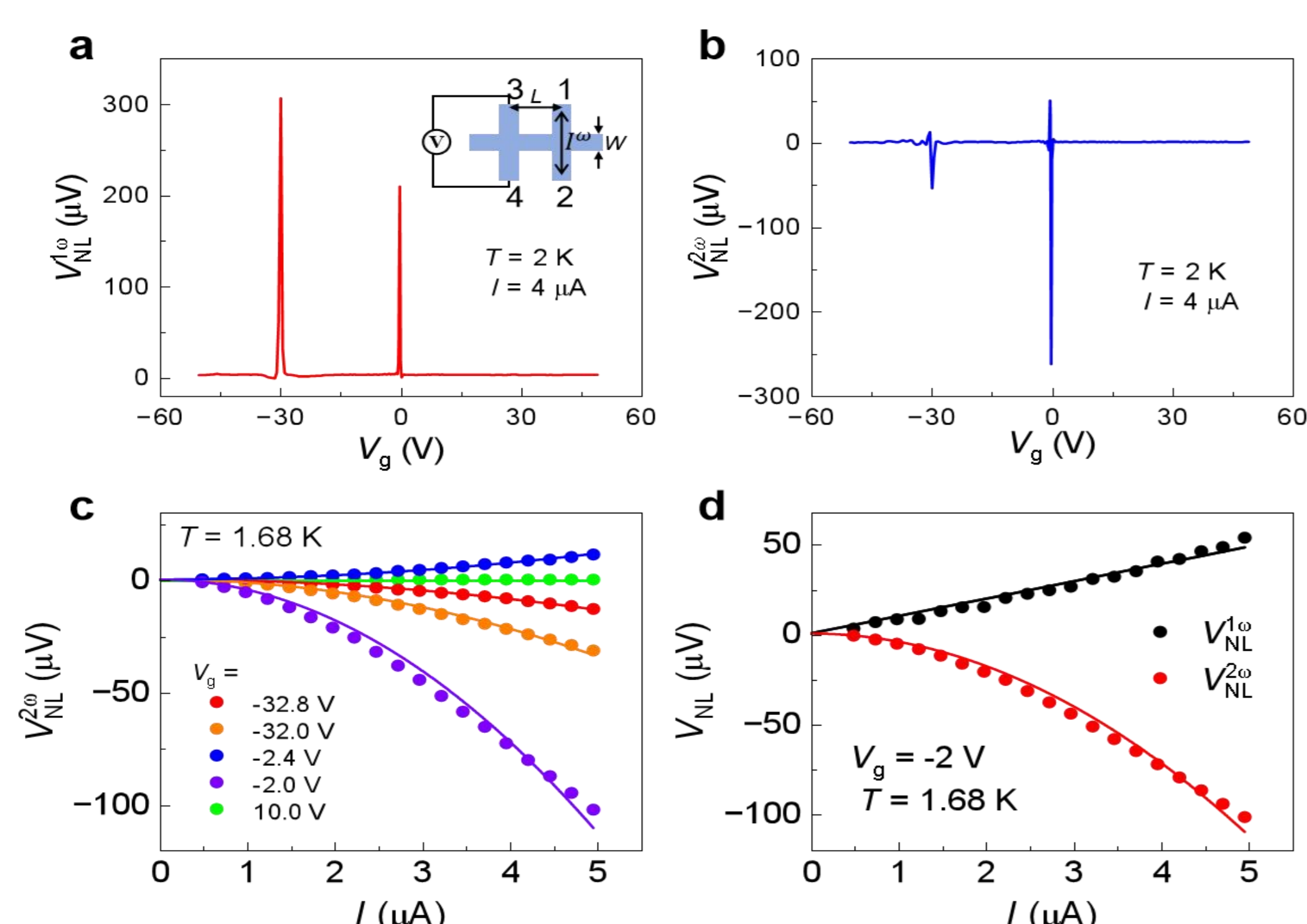


Fig. 3 Gate voltage and current dependence.

Scaling relation

- ◆ $V_{NL}^{2\omega}$ decays exponentially with $T \rightarrow$ thermal activation behaviour
- ◆ $R_{NL}^{2\omega} \propto R_{xx}^4 \rightarrow \chi^v \propto \rho_{xx}^0$ (intrinsic or zeroth-order extrinsic mechanism)
- ◆ $V_{NL}^{2\omega}$ decays with distance $L \rightarrow$ valley diffusion length $l_v \sim 1 \mu m$

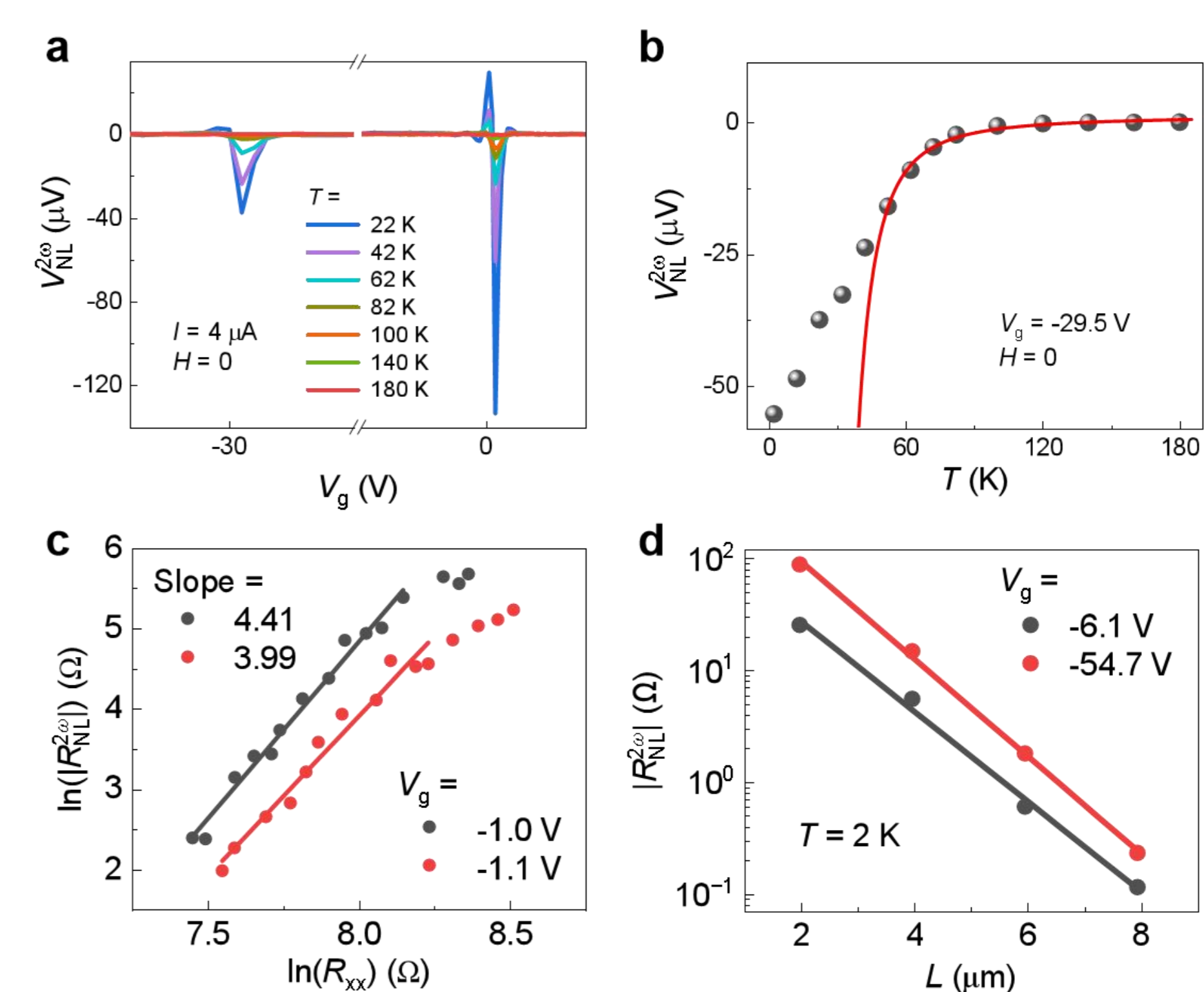


Fig. 4 Temperature dependence and the scaling relation.

Evidence of nonlinear inverse VHE

- ◆ Nonlinear inverse VHE + nonlinear VHE $\rightarrow V_{NL}^{3\omega}$ and $V_{NL}^{4\omega}$
- ◆ Scaling: $R_{NL}^{3\omega} \propto R_{xx}^5$

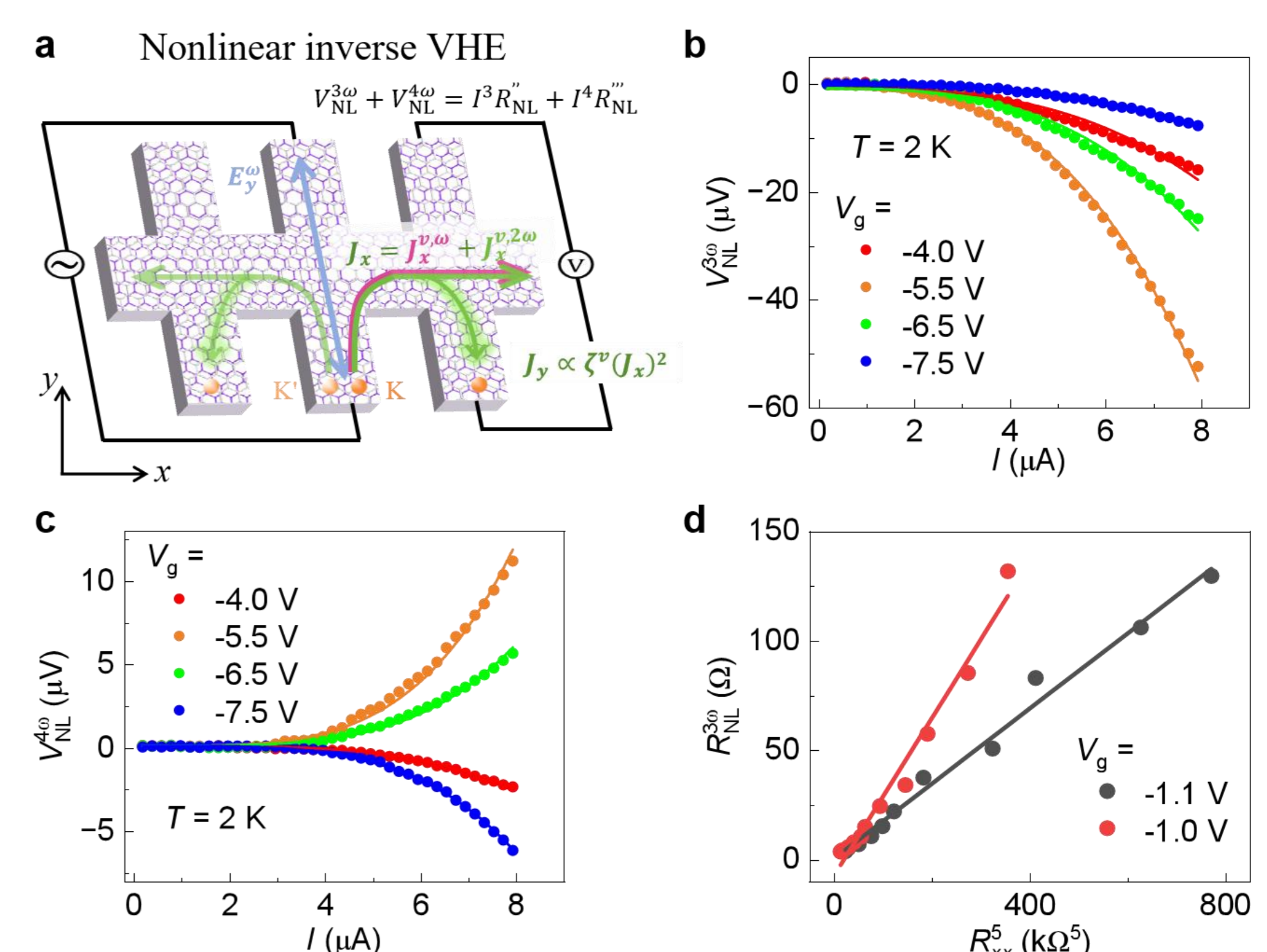


Fig. 5 The nonlinear inverse VHE and the third- and fourth-harmonic non-local voltages.

Conclusion

1. We observe a giant, highly gate tunable nonlinear VHE that surpasses its linear counterpart near Dirac points.
2. The second-harmonic signal $V_{NL}^{2\omega}$ scales quadratically with current and quartically with resistivity, reverses sign across Dirac points, indicating a nonlinear zeroth-order extrinsic origin.
3. For a potential application, the nonlinear VHE enables a valley rectifier that converts a.c. electric currents into d.c. valley current.

Future Work: Detect the Valley-contrasting orbital moment.

References

1. Gorbachev, R. V. et al. *Science* **346**,448-451(2014).
2. Xiao, D. et al. *Phys. Rev. Lett.* **99**, 236809 (2007).
3. Das, K. et al., *Phys. Rev. Lett.* **132**, 096302 (2024).