Temperature-induced Valley Polarization in WS_2 Heterostructures

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Motivation

We investigate the temperature dependence of valley polarization in WS_2 heterostructures. The influence of heterostructures of different materials on the degree of valley polarization is depicted. The intervalley scattering rates under resonant and non-resonant excitation energy as the crucial parameters to see the temperature dependence by considering Fröhlich coupling are calculated. The results show the scattering rate is almost independent of temperature due to a large phonon energy. Subsequently, the major contribution of the observed valley depolarization should come from the change in the radiative lifetime.

Structure

A schematic structure of the heterostuctures is depicted in Fig. 1. Atomically thin materials 1 and 2 are bounded between two semi-infinite substrate materials 3 and 4, characterized by their dielectric functions.

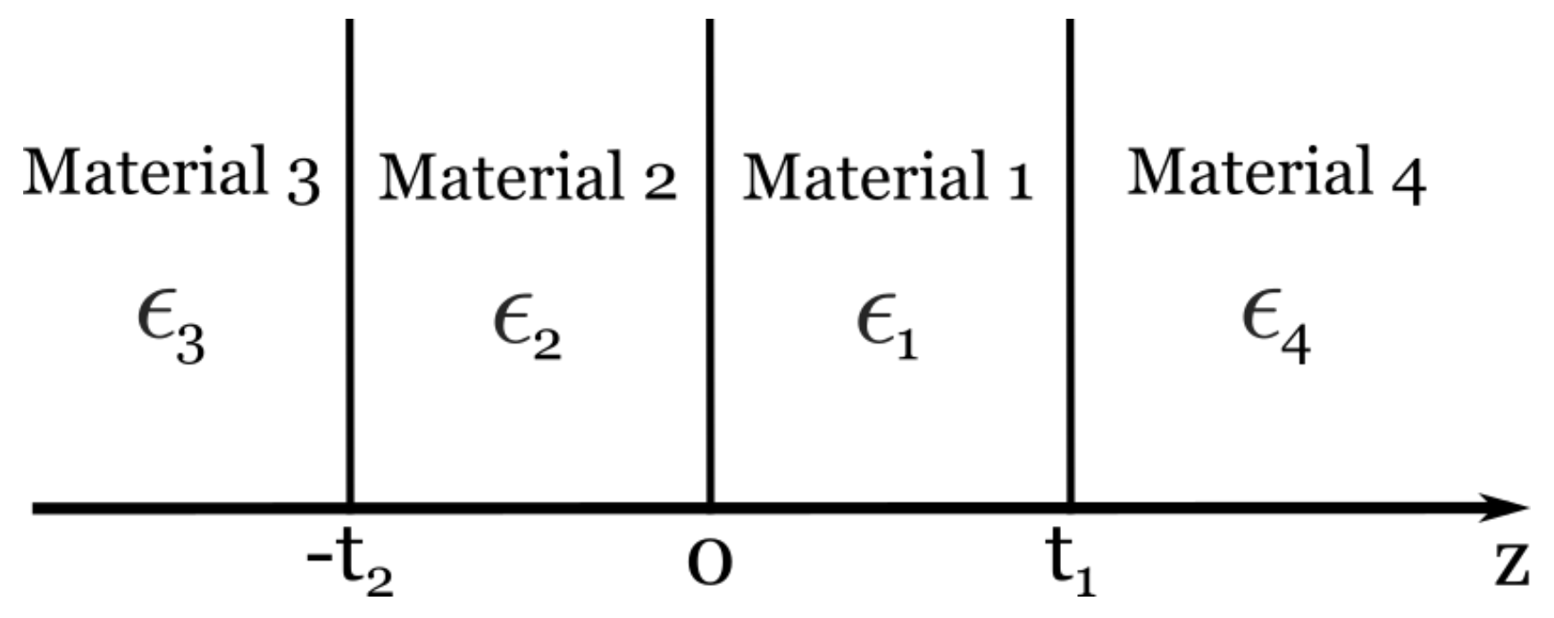

Figure 1: Schematic structure of heterostrutures.

Theory

To find the Fröhlich coupling, we solve Maxwell equations in the form of[1]

$$\phi(\boldsymbol{\rho}, z) = \sum_q \phi(q,z) e^{i\boldsymbol{q}\cdot\boldsymbol{\rho}},$$

$$\phi(q,z) = \phi_0 \begin{cases} (\alpha_1+1)e^{-qz} & z \le t_1 \\ \alpha_1 e^{q(z-2t_1)} + e^{-qz} & 0 < z \le t_1 \\ (1+\alpha_1 e^{-2qt_1})\left[(1-\alpha_2 e^{-qt_2})e^{qz} + \alpha_2 e^{-q(z+t_2)}\right] & -t_2 < z \le 0 \\ (\alpha_2(1-e^{-2qt_2}) + e^{-qt_2})(1+\alpha_1 e^{-2qt_1})e^{q(z+t_2)} & z \le -t_2 \end{cases},$$

$$\alpha_1 = \frac{\epsilon_1 - \epsilon_4}{\epsilon_1 + \epsilon_4}, \qquad \alpha_2 = \frac{(\epsilon_2 - \epsilon_3)\exp(-qt_2)}{\epsilon_2 + \epsilon_3 + (\epsilon_2 - \epsilon_3)\exp(-2qt_2)},$$

The frequencies can be obtained by the secular equation and ϕ_0 is related to the energy of excitation quanta $\hbar\omega_q$.

The Hamiltonain can be written as

$$H = \sum_q -e\phi(q,z) e^{i\boldsymbol{q}\cdot\boldsymbol{\rho}} (a_q + a_{-q}^\dagger),$$

The scattering rate is calculated using Fermi Golden's rule:

$$\frac{1}{\tau_k} = \frac{2\pi}{\hbar} \sum_{k'} |\Gamma(q)|^2 \begin{bmatrix} N(T)\delta(\varepsilon_{k'} - \varepsilon_k - \hbar\omega) + \\ \big(1 + N(T)\big)\delta(\varepsilon_{k'} - \varepsilon_k + \hbar\omega) \end{bmatrix}$$

Table 1 - Energies and dielectric constants of used materials.

	$\hbar\omega_{TO}[meV]$	$\hbar\omega_{LO}[meV]$	$\varepsilon(0)$	$\varepsilon(\infty)$
WS_2	42.9	43	13.7	13.6
hBN	97.6	103	5.1	4.58
SiO_2	55.7	60	3.9	3.36

Results

The results of the measurements and theoretical calculations are shown below[2].

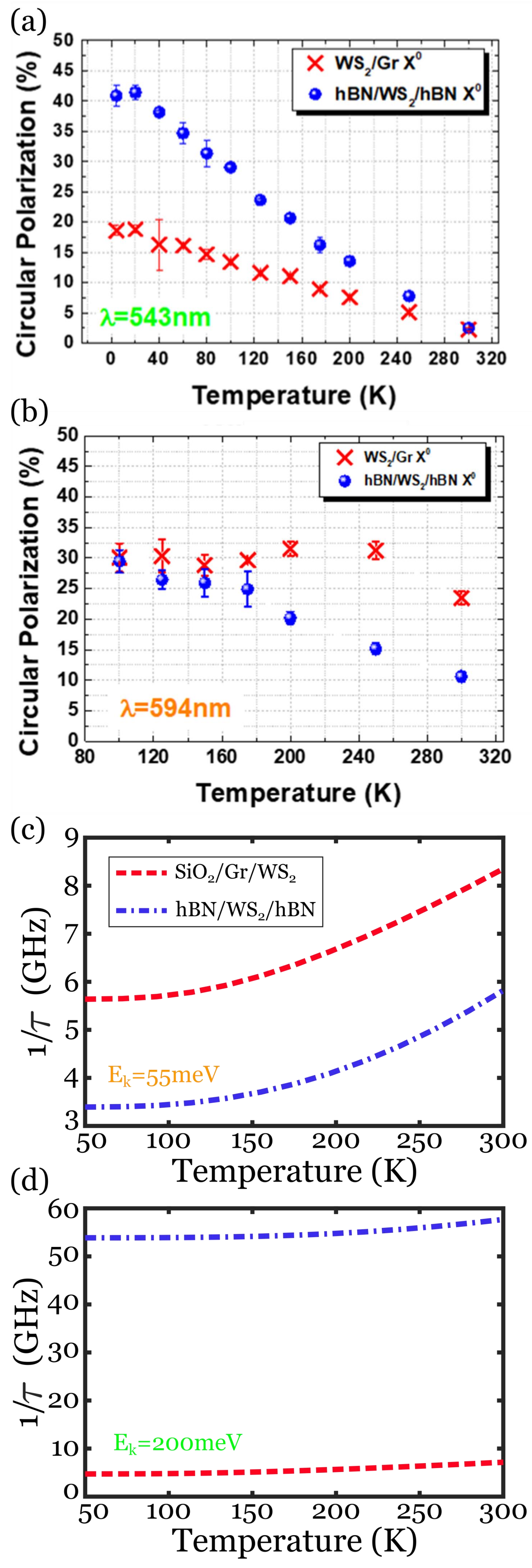

Figure 2: Temperature dependent valley polarization degree for a neutral exciton (X^0) in hBN / WS_2 / hBN and WS_2 / Gr heterostructures as a function of temperature under (a) 543nm and (b) 594nm excitations. The inter-valley scattering rates in WS_2 heterostructures. (c) ε_k=55meV *(near-resonant), and (d)* ε_k=200meV *(off-resonant).*

To measure the polarization, the system is excited with right-handed circularly polarized light (σ+), and then the resultant photoluminescence is analysed for co-polarized (σ+) and cross-polarized/left-handed circularly polarized light (σ-). The degree of valley polarization is related to the circularly polarized emission (P_{circ})1,2 where $P_{circ}=(I_+-I_-)/(I_++I_-)$, and I_+ (I_-) is the intensity of the σ+ (σ-) helicity PL component. In Figs. 2a and 2b, the X^0 temperature-dependent polarization degree of hBN/WS_2/hBN and WS2/Gr are shown for non- and near-resonant excitation conditions using the 543nm (2.283eV) and 594nm (2.087eV) laser sources, respectively. Figs. 2c and 2d show scattering rate from the bottom of the conduction band at the K point to all available states near the K' point. The bandgap energy shift due to electron-phonon coupling is shown in Fig. 3b, where the temperature dependence of the bandgap shift is much weaker than observed in the experiment (Fig. 3a).

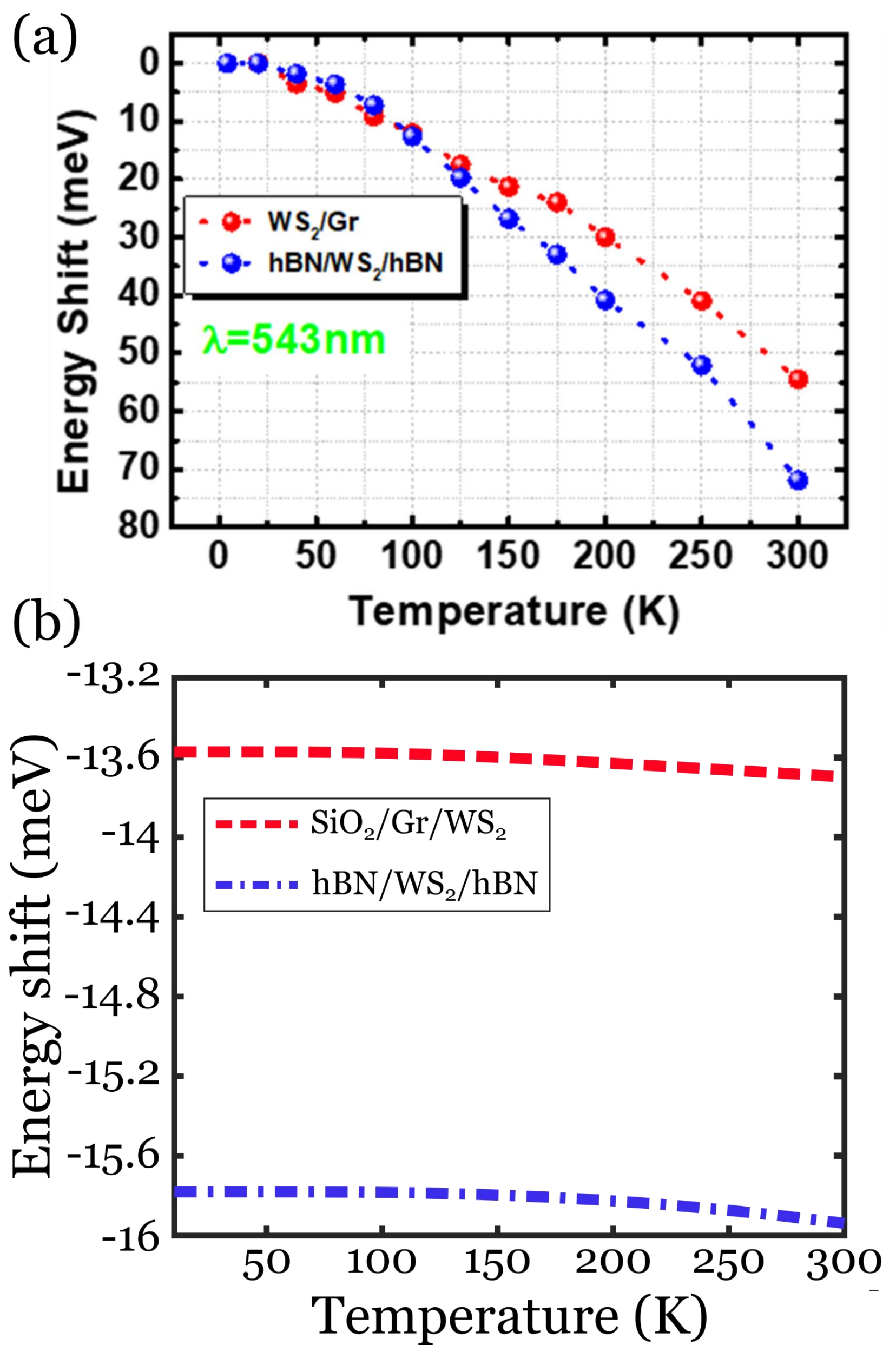

Figure 3: Temperature dependent energy shift of X^0 in hBN / WS_2 / hBN and WS_2 / Gr. (a) Experimental and (b) theory. In (a) the energy shift is normalized and set to 0 at 4K in both cases.

Conclusion

We have demonstrated temperature dependent valley polarization measurements in CVD-grown a) monolayer WS_2 encapsulated in hBN and b) monolayer WS_2 on top of graphene excited by two different photon energies. The electron-phonon model partly explains for the observed temperature dependency.

References

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2. Paradisanos, I., McCreary, K.M., Adinehloo, D., Mouchliadis, L., Robinson, J.T., Chuang, H.J., Hanbicki, A.T., Perebeinos, V., Jonker, B.T., Stratakis, E. and Kioseoglou, G., Appl. Phys. Lett. 116, 203102 (2020)

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