

Interlayer coupling enables EPC-linked polarity inversion and enhanced heat dissipation in MoS₂/CrOCl heterostructures

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ABSTRACT: Interlayer coupling is a central knob for engineering electronic reconstruction and energy dissipation in van der Waals heterostructures. However, a unified understanding that connects interlayer coupling to both electrical and thermal responses is still lacking. Electron–phonon coupling (EPC) could bridge this gap by linking interlayer coupling to both momentum and energy relaxation. Here we establish an electron–phonon-coupling-linked interpretation in MoS₂/CrOCl heterostructures, where interlayer coupling-driven interfacial charge transfer reshapes the MoS₂ electronic structure and, at the same time, renormalizes electron–phonon interactions that govern momentum relaxation and carrier–lattice energy relaxation. By using Raman and second-harmonic-generation (SHG) fingerprints, we

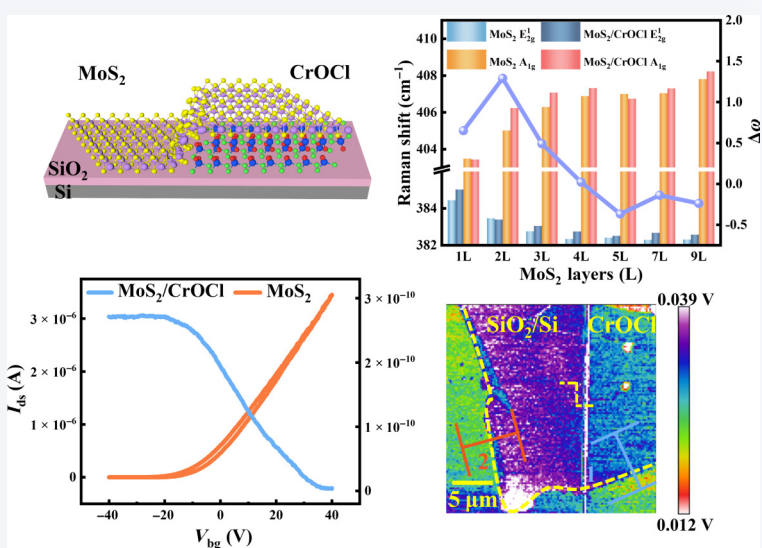
identify 2L-MoS₂/CrOCl as the strongest coupling configuration. This is evidenced by the largest change in mode separation of 1.29 cm^{−1} and a pronounced SHG suppression from 100% to 11.9% relative to pristine MoS₂. In this regime, interfacial charge redistribution downshifts the MoS₂ Fermi level and converts its native n-type character to p-type conduction. Along with electrical transport experiments, we also study the thermal transport and characterize the steady-state temperature rise through scanning thermal microscopy. The MoS₂/CrOCl heterostructure exhibits more efficient heat evacuation, reducing the temperature rise by 38.5% under identical thermal loading conditions compared with the situation for pristine MoS₂ on SiO₂/Si. According to the observed EPC-linked electron and phonon transport properties, interlayer coupling in MoS₂/CrOCl heterostructure has induced both carrier-polarity inversion and enhanced interfacial heat dissipation, providing a basis for codesigning electro-thermal multifunctional devices.

KEYWORDS: interlayer coupling, MoS₂/CrOCl, electron–phonon coupling, carrier-polarity inversion, heat dissipation

1 Introduction

Via moiré engineering, external fields, and hydrostatic pressure, a wide range of interfacial properties in van der Waals (vdW)

heterostructures can be tuned, including charge transport, symmetry responses, and heat dissipation [1–6]. Among them, tuning electrical and thermal characteristics is crucial for enhancing device performance and reliability. These approaches work essentially by modulating the effective interlayer coupling, which strongly reshapes the interfacial electronic structure and lattice dynamics [7]. For example, interlayer coupling could lead to symmetry breaking, interfacial charge transfer, and phonon renormalization in MoS₂-based vdW heterostructures [1, 8, 9].



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Interlayer coupling has also been shown to effectively modulate thermal transport as well as electronic structure. For instance, in graphene/MoS₂ heterostructures, the interfacial thermal conductance G increases with the interfacial coupling strength [10]. However, existing studies have rarely established a direct connection from the interlayer coupling to electron and phonon transport. How coupling-induced interfacial charge redistribution further jointly affects electrical transport and heat dissipation performance still lacks a unified understanding.

With twofold influences on both carrier transport dynamics and thermal transport dynamics, electron-phonon coupling (EPC) provides an ideal path towards deeper comprehension on interlayer coupling. EPC affects momentum relaxation that limits carrier mobility and electrical transport, while it mediates carrier-to-lattice energy relaxation that governs hot-carrier cooling and heat release [11, 12]. Importantly, interlayer coupling modifies interfacial hybridization and band alignment, thereby driving interfacial charge redistribution and a corresponding Fermi-level shift. Such charge redistribution not only changes the available electron-phonon scattering phase space, but can also alter the carrier screening environment of the phonon-induced perturbation potential, thereby renormalizing the screened electron-phonon matrix elements [13, 14]. As a result, EPC can be modulated through both scattering phase space and matrix-element renormalization, allowing interlayer coupling to simultaneously influence electrical and thermal transport [13–15]. Consistent with this picture, Raman measurements on twisted bilayer MoS₂ have revealed pronounced interlayer coupling-induced phonon renormalization associated with moiré-lattice reconstruction [16]. Moreover, sufficiently strong interlayer interaction may enable interlayer EPC channels, offering additional pathways for interfacial energy transfer [17].

Inspired by these EPC-related studies, we systematically investigated interlayer coupling in MoS₂/CrOCl heterostructures. According to our previous report, CrOCl could form strong vdW coupling with MoS₂, enabling pronounced interfacial charge transfer, while its high and anisotropic thermal conductivity provides an efficient heat-spreading pathway [18]. The coexistence of strong interfacial charge transfer and efficient heat spreading characteristics renders MoS₂/CrOCl an ideal platform for investigating how interlayer coupling simultaneously affects electrical transport and heat dissipation through EPC-linked interpretation. To this end, we fabricated thickness-dependent MoS₂/CrOCl heterostructures and evaluated the interlayer coupling effect via Raman and second-harmonic generation (SHG). The strongest coupling configuration is observed to be 2L-MoS₂/CrOCl, as evidenced by the largest change in mode separation of 1.29 cm⁻¹ and a pronounced SHG suppression from 100% to 11.9% relative to pristine MoS₂. Driven by strong interlayer coupling, the depletion of electrons in MoS₂ and the consequent downshift of its Fermi level enable a gate-tunable polarity inversion. Such tunability unlocks potential polarity-programmable applications. Scanning thermal microscopy (SThM) further shows more efficient heat evacuation in MoS₂/CrOCl, with the temperature rise reduced by 38.5% compared with the situation for MoS₂ on SiO₂/Si. Overall, our results establish interlayer coupling as a practical knob for codesigning electrical polarity and thermal management in vdW heterostructures.

2 Results and discussion

In our experiments, MoS₂ flakes with well-defined thicknesses (1–9 layers, 1L–9L) were prepared by mechanical exfoliation from bulk

MoS₂ crystals, using polydimethylsiloxane (PDMS) as the transfer medium. As shown in Fig. 1(d), layer numbers (1L–9L) were determined by combining atomic force microscopy (AFM) line profiles with previous literature calibration. On SiO₂/Si, monolayer MoS₂ typically shows an apparent AFM height of 0.8–1.0 nm, which can be larger than the pristine thickness of MoS₂ due to interfacial adsorbate effects. For thicker regions, the thickness increases stepwise by 0.6–0.7 nm per added layer, consistent with the crystallographic interlayer spacing of 3R-MoS₂ (~0.62 nm), establishing the thickness-layer correspondence used here [19]. To further validate the layer assignment, the characteristic Raman spectra of our samples show excellent agreement with standard layer-dependent Raman spectra (Fig. S5 in the Electronic Supplementary Material (ESM)) [20]. We investigated the interlayer coupling between MoS₂ and CrOCl by dry-transferring a MoS₂ flake onto a pre-exfoliated approximately 13-layer CrOCl flake on SiO₂/Si (Fig. S1 in the ESM). The CrOCl flake used in this work has a thickness of 12 nm, as confirmed by AFM (Fig. S2 in the ESM). This thickness lies within the thin-flake regime, where CrOCl has been reported to support strong interfacial coupling while preserving pronounced in-plane thermal anisotropy [9, 21].

To enhance the interfacial contact and coupling, the resulting heterostructure was subsequently annealed at 120 °C for 2 h under an argon atmosphere. Figures 1(a) and 1(b) show the schematic illustration and optical microscopy (OM) image of the MoS₂/CrOCl heterostructure, respectively. As shown in Fig. 1(b), the orange, red, yellow, blue, and purple dashed lines correspond to 1L, 2L, 3L, 5L, and 9L MoS₂ stacked on CrOCl, respectively. Notably, the entire region of the MoS₂/CrOCl heterostructure displayed a clear and uniform morphology under OM, with no cracks or wrinkles observed. These features directly indicate good interfacial contact quality between MoS₂ and CrOCl. This conclusion was further corroborated by AFM (Fig. 1(c)) and Raman mapping (Fig. S4 in the ESM).

SHG is a symmetry-sensitive optical probe in vdW heterostructures, because interlayer coupling can modify lattice symmetry and electronic structure, thereby strongly modulating the effective second-order nonlinear susceptibility $\chi^{(2)}$ [22–24]. Here we employed SHG as an experimentally accessible and symmetry-sensitive indicator of interlayer coupling in the MoS₂/CrOCl heterostructure. After calibrating the MoS₂ thickness by AFM, spatially resolved SHG spectra (Fig. 1(e)) acquired across disparate regions show a clear suppression of SHG intensity upon heterostructure formation for all tested layers. Quantitatively, the SHG intensities are reduced to 19.3%, 11.9%, 15.2%, 18.4%, and 15.8% of their respective pristine values for 1L, 2L, 3L, 5L, and 9L MoS₂, as shown by the purple solid line in Fig. 2(i). Notably, the suppression exhibits a pronounced nonmonotonic layer dependence and reaches its maximum for 2L-MoS₂/CrOCl, where the SHG intensity decreases to 11.9% of the pristine value. This nonmonotonic SHG modulation likely reflects a competition between the intrinsically thickness-enhanced SHG of 3R-MoS₂ and the interface-induced symmetry perturbation introduced by CrOCl. The especially strong suppression in 2L-MoS₂/CrOCl may result from the high sensitivity of bilayer 3R-MoS₂ to interfacial perturbations and the efficient transmission of such perturbations across the entire bilayer.

To elucidate the thickness-dependent SHG suppression in MoS₂/CrOCl heterostructures, we probed interlayer coupling from the lattice-vibration perspective. Figure 2(a) shows a representative Raman spectrum acquired from the MoS₂/CrOCl heterostructure

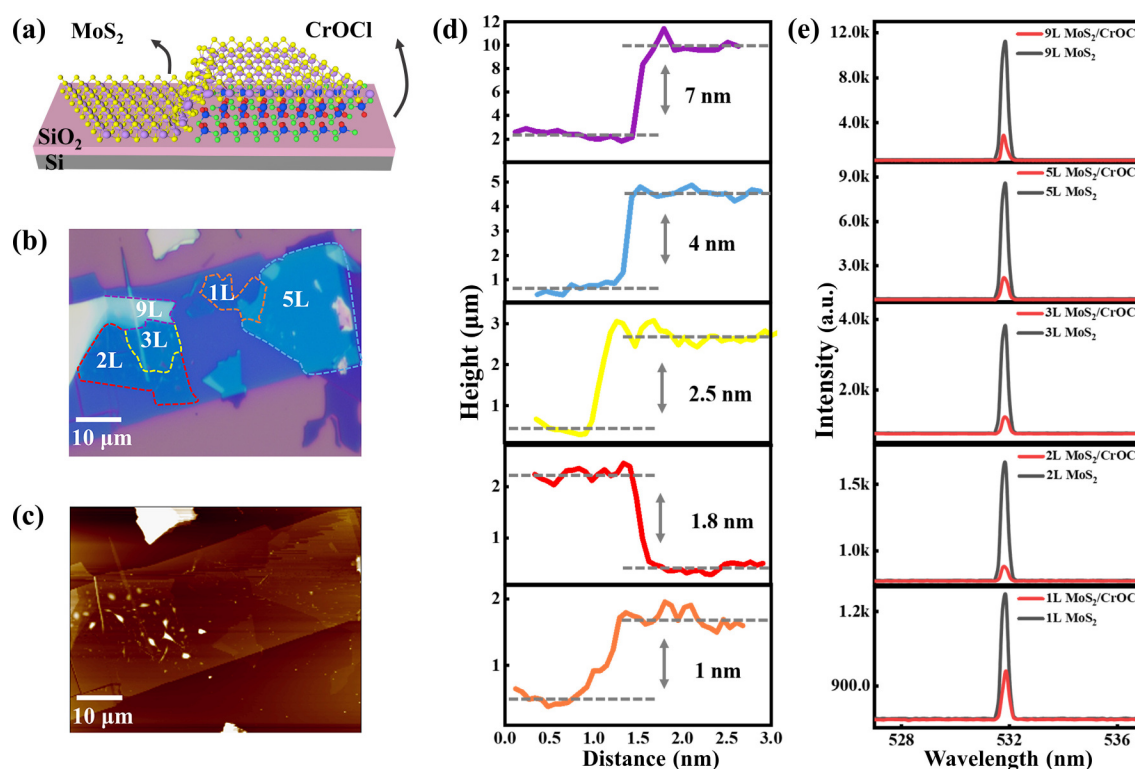


Figure 1 Fabrication and structural characterization of MoS₂/CrOCl heterostructures. (a) Schematic illustration of the MoS₂/CrOCl heterostructure fabrication process on SiO₂/Si substrates. (b) OM image of the MoS₂/CrOCl heterostructure, where the orange, red, yellow, blue, and purple dashed lines correspond to 1L, 2L, 3L, 5L, and 9L MoS₂ stacked on CrOCl, respectively. Scale bar: 10 μm. (c) AFM topographic image of the MoS₂/CrOCl heterostructure. Scale bar: 10 μm. (d) AFM line profile for different numbers of MoS₂ layers. Scale bar: 10 μm. (e) Room-temperature SHG spectra of pristine MoS₂ (black curve) with 1L–9L and the MoS₂/CrOCl heterostructure (red curve) formed therefrom.

region. The prominent peaks at ~ 383.5 and ~ 405.0 cm⁻¹ are assigned to the in-plane E_{2g}^1 and out-of-plane A_{1g} modes of MoS₂, while the peaks at ~ 207.4 , ~ 415.1 , and ~ 455.4 cm⁻¹ attributed to the A_g^1 , A_g^2 , and A_g^3 modes of CrOCl, respectively [1]. After annealing, both phonon modes of MoS₂ exhibited blueshifts, with A_{1g} and E_{2g}^1 shifting by 1.39 and 0.27 cm⁻¹, respectively, increasing the mode separation $\Delta\omega$ from 21.70 to 22.82 cm⁻¹ (Fig. 2(b)). Since thermal annealing is known to effectively remove interfacial residues and enhance intimate interlayer contact at vdW interfaces, the increased $\Delta\omega$ provides evidence that interlayer coupling is strengthened (Fig. S3 in the ESM) [25, 26]. Motivated by this observation, we further performed Raman measurements on pristine MoS₂ flakes (1L–9L) and the corresponding MoS₂/CrOCl heterostructures (Fig. S5 in the ESM). As summarized in Fig. 2(c), the A_{1g} mode exhibited a consistent blue shift (0.07–1.31 cm⁻¹) across all MoS₂ thicknesses (1L–9L), while the E_{2g}^1 mode showed relatively small and thickness-dependent shifts. Notably, the change in mode separation systematically increases upon heterostructure formation and displays a distinct layer-number dependence, with the largest enhancement magnitude of 1.29 cm⁻¹ observed for the 2L-MoS₂/CrOCl. This trend correlates with the SHG response (Fig. 2(i)): The 2L-MoS₂/CrOCl heterostructure also exhibited the strongest SHG suppression, with the SHG intensity reduced to 11.9% of its respective pristine value. Collectively, the consistent layer-dependent Raman and SHG fingerprints indicate that the 2L-MoS₂/CrOCl exhibits the strongest interlayer coupling regime among all the studied configurations. This thickness dependence suggests that interfacial charge transfer is the primary factor governing the coupling strength. Nevertheless, interfacial strain and

other structural effects at the heterointerface may also modulate the final coupling response, particularly for non-2L heterostructures.

Given the intrinsically low symmetry of CrOCl, we performed polarization-resolved Raman spectroscopy on the 2L-MoS₂/CrOCl heterostructure to further probe interlayer coupling-induced symmetry reduction [27]. The sample was mounted on a rotational stage, with the horizontal direction defined as $\theta = 0^\circ$. For pristine MoS₂ on SiO₂/Si, the E_{2g}^1 and A_{1g} intensities remain unchanged with incident polarization under our scattering configuration, consistent with the high in-plane symmetry of MoS₂. In contrast, both modes in the 2L-MoS₂/CrOCl heterostructure exhibited a clear twofold symmetry in their angle-dependent Raman intensities, as shown in Figs. 2(e) and 2(f). As θ increased from 0° to 90° , the intensities of both modes progressively increased, yielding anisotropy ratios of ~ 1.45 for E_{2g}^1 and ~ 2.05 for A_{1g} at 90° (intensity ratio defined as $I_{\text{hetero}}/I_{\text{pristine}}$). The contour map in Fig. 2(d) further visualized this symmetry-breaking behavior, with the A_{1g} mode showing more pronounced intensity modulation. The coupling-induced symmetry reduction is further supported by polarization-dependent SHG measurements. The SHG intensity can be described as $I(\theta) = I_0 \cos^2(3\theta)$, where I_0 is the maximum SHG intensity, and θ is the angle between the incident polarization and the armchair (AC) axis of pristine MoS₂. As shown by the orange polar plot in Fig. 2(h), pristine MoS₂ exhibited a clear six-fold symmetry, which is typical characteristic of the D_{3h} point group. In contrast, the SHG response of the MoS₂/CrOCl heterostructure reveals a distinctly anisotropic intensity pattern (blue data in Fig. 2(h)), with SHG intensity enhanced along specific AC directions and suppressed along others, as further visualized in the

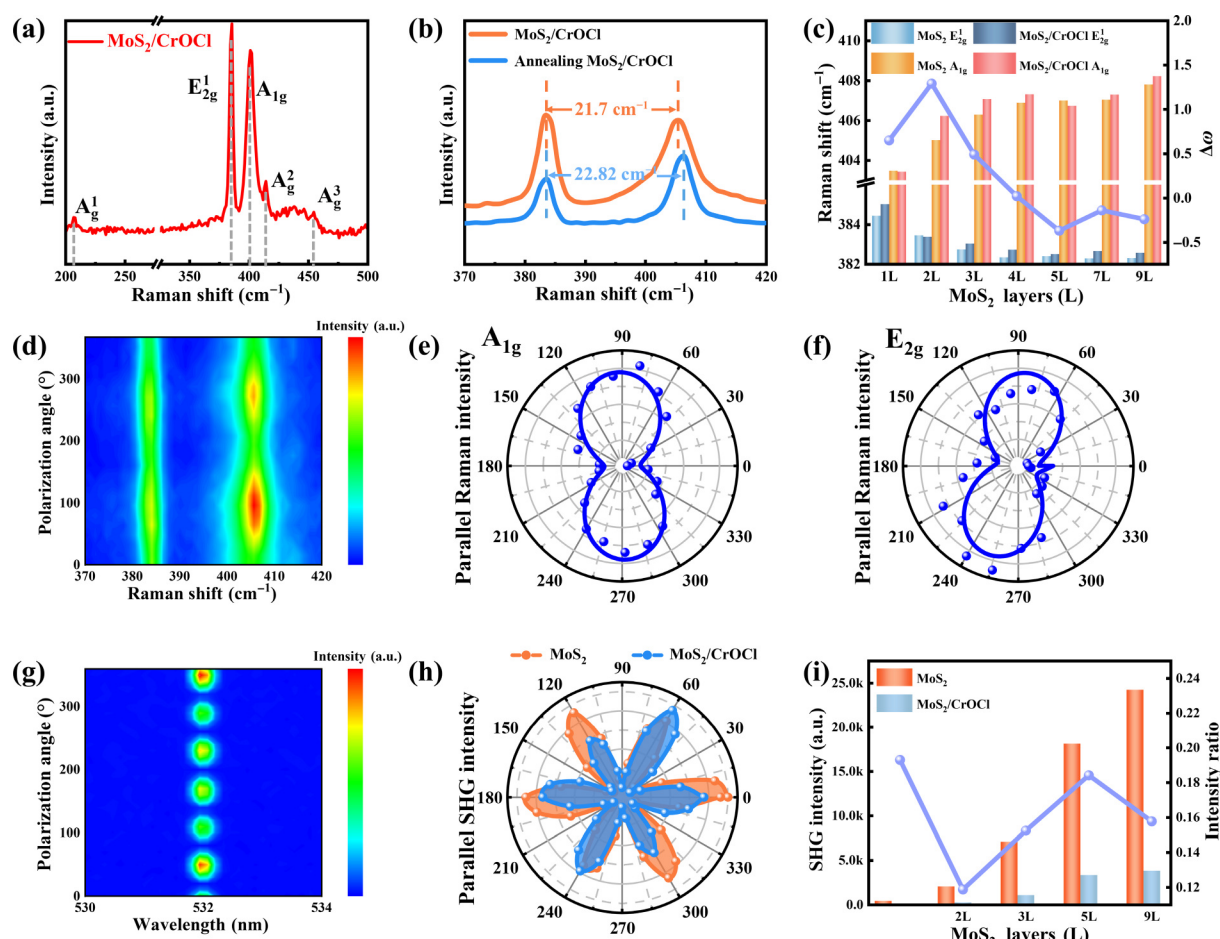


Figure 2 Probing thickness-dependent interlayer coupling in MoS₂/CrOCl heterostructures. (a) Raman spectrum of MoS₂/CrOCl heterostructures. (b) Raman spectra of the heterostructure before (orange) and after (blue) annealing. The dashed lines indicate the positions of E_{2g}¹ and A_{1g} modes, with the mode separation defined as $\Delta\omega = \omega(A_{1g}) - \omega(E_{2g}^1)$ (cm⁻¹). (c) Raman peak positions of E_{2g}¹ and A_{1g} for pristine MoS₂ (1L-9L) and the constructed MoS₂/CrOCl heterostructures. The purple solid line indicates the change in mode separation of MoS₂ before and after stacking. ((d) and (g)) Contour maps of angle-dependent Raman and SHG intensity in the 2L-MoS₂/CrOCl heterostructure. ((e) and (f)) Polarization plots and fitted curves for E_{2g}¹ and A_{1g} intensities, with blue dots denoting the experimental data. (h) Angle-resolved SHG intensity polarization map of the 2L-MoS₂/CrOCl heterostructure. (i) Summary bar plot of SHG intensity for 1L-9L pristine MoS₂ and the corresponding MoS₂/CrOCl heterostructure; the purple solid line represents the thickness-dependent SHG intensity ratio.

intensity contour map (Fig. 2(g)). Consistent symmetry breaking observed in both polarization-resolved Raman and SHG measurements provides robust evidence for pronounced interlayer coupling in the 2L-MoS₂/CrOCl heterostructure.

To elucidate the physical origin of the optical anisotropy induced by strong interlayer coupling, we further investigated interfacial interactions in the 2L-MoS₂/CrOCl heterostructure from the perspective of interfacial charge transfer. The peak positions of the A_{1g} and E_{2g}¹ modes are sensitive to electron doping and strain, respectively, enabling us to quantitatively disentangle their contributions to the observed optical anisotropy [28–30]. As shown in Fig. 3(a), we compared the Raman spectra acquired from pristine MoS₂ on SiO₂/Si (q₁) and MoS₂/CrOCl heterostructure (q₂). At the point q₂, the A_{1g} mode exhibited a significant blue shift of 1.22 cm⁻¹ with a reduced full width at half maximum (FWHM), while the E_{2g}¹ mode showed a negligible red shift. This indicates a synergistic effect of interfacial charge transfer and strain at the heterointerface, where charge transfer dominates. This conclusion is consistent with the angle-dependent Raman contour map in Fig. 2(d), which showed higher anisotropy in the A_{1g} mode than in the E_{2g}¹ mode.

To further corroborate the dominant role of interfacial charge

transfer inferred from Raman analysis, we performed photoluminescence (PL) measurements, as the PL intensity and peak energy are highly sensitive to the exciton population and the renormalized optical transition energy [2, 31, 32]. As shown in Fig. S6 in the ESM, the heterostructure region (q₂) exhibited pronounced PL quenching accompanied by a clear blue shift of the A exciton peak ($\Delta E = 12$ meV), consistent with interfacial charge transfer behavior and suppressed radiative recombination due to efficient interfacial charge separation. We conducted Gaussian fitting on the A exciton emission (Figs. 3(b) and 3(c)) to elucidate the charge transfer mechanism and quantify corresponding transfer efficiency at the MoS₂/CrOCl heterointerface. The A exciton line shape can be divided into two components: the neutral exciton (A⁰) originating from the pristine Coulomb interaction and the trion (A⁻) dominated by carrier-doping effects [33]. For pristine 2L MoS₂ on SiO₂/Si (q₁, Fig. 3(b)), the emission was dominated by the A⁻ peak located at ~1.85 eV, indicative of n-type doping characteristics [34]. In contrast, in the MoS₂/CrOCl heterostructure region (q₂, Fig. 3(c)), the emission was dominated by the A⁰ peak at ~1.89 eV, with its fitted area percentage increasing to nearly four times that of the pristine MoS₂ (Table S1 in the ESM). This marked

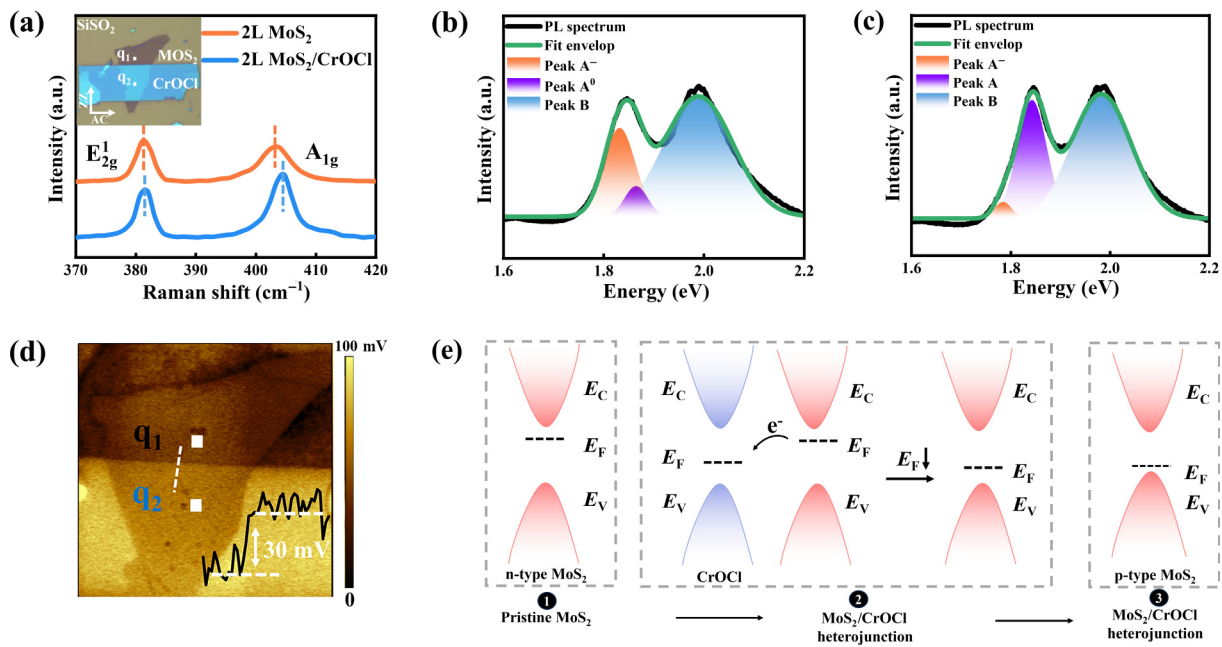


Figure 3 Charge Transfer in the 2L-MoS₂/CrOCl hHeterostructures driven by strong interlayer coupling. (a) Raman spectrum of the 2L-MoS₂/CrOCl heterostructure. Inset shows the OM image. Orange line: pristine MoS₂ on SiO₂/Si (point q₁). Blue line: MoS₂ on CrOCl (point q₂). (b) and (c) PL spectra extracted from points q₁ and q₂ in (a), showing the experimental PL intensity (black line) and fitted curve (green line), where the A exciton peak is decomposed into A⁻ and A⁰ via a Gaussian function. (d) KPFM surface potential image of the 2L-MoS₂/CrOCl heterostructure, with the surface potential difference across the white dashed line shown in the bottom-right corner. (e) Density functional theory calculations of the band structures of 2L MoS₂ and CrOCl. Left panel: Band structure of pristine n-type 2L MoS₂. Middle panel: charge transfer at the interface during the heterostructure formation, inducing band realignment (Fermi level shifts toward the valence band of MoS₂). Right panel: applying a finite negative electric field ($V_{ds} = 0.1$ V, corresponding to a negative source-drain configuration) further lowers the Fermi level, resulting in p-type conduction characteristics for the heterostructure.

transition from trion-dominant to neutral-exciton-dominant PL provides spectroscopic evidence for a strong interlayer coupling-induced interfacial charge transfer from MoS₂ to CrOCl. To further validate this charge transfer process, we conducted Kelvin probe force microscopy (KPFM) measurements. As shown in Fig. 3(d), the surface potential in the heterostructure region (q₂) was ~ 30 mV higher than that in the pristine region (q₁), providing independent electrical evidence for the effective electron depletion in MoS₂ upon interfacing with CrOCl [2, 34]. These PL and KPFM signatures align with our previous studies on MoS₂/CrOCl heterostructures [1, 28], which revealed a higher KPFM surface potential in the MoS₂/CrOCl overlapping region than in pristine MoS₂. We deconvoluted the A exciton emission into neutral-exciton and trion components, then applied a trion mass-action model to quantify the extent of interfacial electron depletion in MoS₂. Here, the fitted area ratio A_{A^-}/A_{A^0} is used as a relative indicator of the free-electron population in MoS₂.

Based on the above optical and electrical evidence, we propose a schematic model illustrated in Fig. 3(e). In the absence of an external bias, 2L MoS₂ and CrOCl form a Type-II band-alignment heterostructure that favors interfacial charge separation, which commonly manifests as PL quenching in atomically thin heterostructures [31, 35]. Strong interlayer coupling can further renormalize the MoS₂ electronic structure and facilitate interfacial charge transfer from MoS₂ to CrOCl, consistent with the observed reduction of trion emission and the KPFM surface potential contrast [1]. The enhanced electron-hole separation in MoS₂ promotes the formation of indirect excitons, which predominantly recombine nonradiatively [36, 37]. Importantly, interfacial charge transfer causes a significant downward shift of the Fermi level in

2L MoS₂. Under a negative gate bias, the Fermi level is shifted further toward the valence-band maximum, endowing the system with pronounced p-type conduction. Thus, strong interlayer coupling-induced charge transfer provides an effective route to convert MoS₂ from its native n-type behavior toward p-type conduction [9, 38]. Beyond carrier-polarity control, interfacial charge redistribution induced by interlayer coupling offers a route to tune EPC, which concurrently governs electrical and thermal transport in vdW heterostructures [39]. Moreover, strong interlayer coupling may activate interlayer EPC channels, as has been directly observed in vdW stacks such as WSe₂/hexagonal boron nitride (hBN), where phonons in hBN are rendered Raman-active through interlayer resonant coupling to electronic transitions in the WSe₂. These interlayer channels provide additional cross-layer pathways for interfacial energy transfer [40].

As discussed above, the strongly coupled Type-II MoS₂/CrOCl vdW heterostructure drives charge transfer from MoS₂ to CrOCl. This process depletes electrons in MoS₂, downshifts its Fermi level, and enables gate-tunable polarity inversion. Moreover, the accompanying interfacial charge redistribution further modifies dielectric screening and can renormalize EPC, thus providing an additional handle to regulate heat dissipation. To verify the discussion, we fabricated MoS₂/CrOCl heterostructure field-effect transistors (FETs) and examined their gate-tunable electrical characteristics. In Fig. 4(a), the channel between electrodes 1 and 2 corresponds to pristine 2L MoS₂ FET, whereas the channel between electrodes 3 and 4 belongs to 2L-MoS₂/CrOCl heterostructure FET, with each constituent layer outlined by the green and red dashed lines, respectively. All the electrodes (5 nm Cr/70 nm Au) were fabricated via standard e-beam lithography and e-beam evaporation

to ensure reliable interfacial contacts. As shown in Fig. 4(b), both FETs exhibited near-Ohmic output characteristics in the low-bias regime, indicating that contact nonlinearity is not the dominant factor in the polarity characterization. More critically, the transfer characteristics provided direct evidence of carrier-polarity inversion in MoS₂. As shown in Fig. 4(c), the pristine 2L MoS₂ FET turned on under positive gate bias and exhibited a current on/off modulation exceeding four orders of magnitude (from $\sim 10^{-10}$ to more than 2×10^{-6} A), consistent with the commonly observed n-type behavior of pristine MoS₂ arising from defect-related doping and Fermi-level pinning at metal–MoS₂ interfaces [41]. In contrast, the 2L-MoS₂/CrOCl FET exhibited unambiguous p-type conduction at $V_{ds} = 0.1$ V (Fig. 4(d)). Together with the optical evidence in Fig. 2, this result supports an electron-depletion mechanism: Strong interlayer coupling drives substantial interfacial charge transfer from MoS₂ to CrOCl, downshifting the Fermi level and stabilizing p-type transport under gate bias. This successful polarity inversion establishes a versatile platform for complementary electronics, enabling both n- and p-type operation from the same material through tailored interlayer coupling.

Beyond polarity inversion, such interlayer coupling-driven interfacial charge redistribution at the 2L-MoS₂/CrOCl interface is expected to reshape energy dissipation pathways through EPC. Guided by our EPC-linked interpretation, we quantitatively

characterized the heat evacuation of the MoS₂/CrOCl heterostructure using SThM. A laser-heated resistive probe (configured in a Wheatstone bridge) exchanges heat with the sample upon contact, leading to a change in probe temperature that is transduced into an output voltage signal U . Following the established SThM framework, the relationship between the U and the thermal conductivity (κ) of the sample can be expressed as [42]

$$\frac{dU}{d(\kappa^{-1})} = \begin{cases} \frac{\beta Q}{\pi r}, \kappa < C \\ \frac{\beta Q}{2\pi\delta} \left(\ln \frac{\alpha}{vr} - \frac{3}{2} \right), \kappa \geq C \end{cases} \quad (1)$$

where β is a constant determined by the parameters of the Wheatstone bridge, Q is the heat flow from the thermal probe to the sample, r is the thermal contact radius, δ is the length of the cylindrical heat source, α is the thermal diffusivity, v is the scanning rate, and C is a probe-dependent conductivity scale factor. Through tight control of the scanning conditions and probe-sample contact, $\frac{\alpha}{vr}$ remains approximately constant during measurement, yielding a nearly linear dependence of the SThM output on κ^{-1} under our experimental operating regime. Consequently, after calibrating the probe voltage sensitivity coefficient (S), we convert the thermal signal to the corresponding

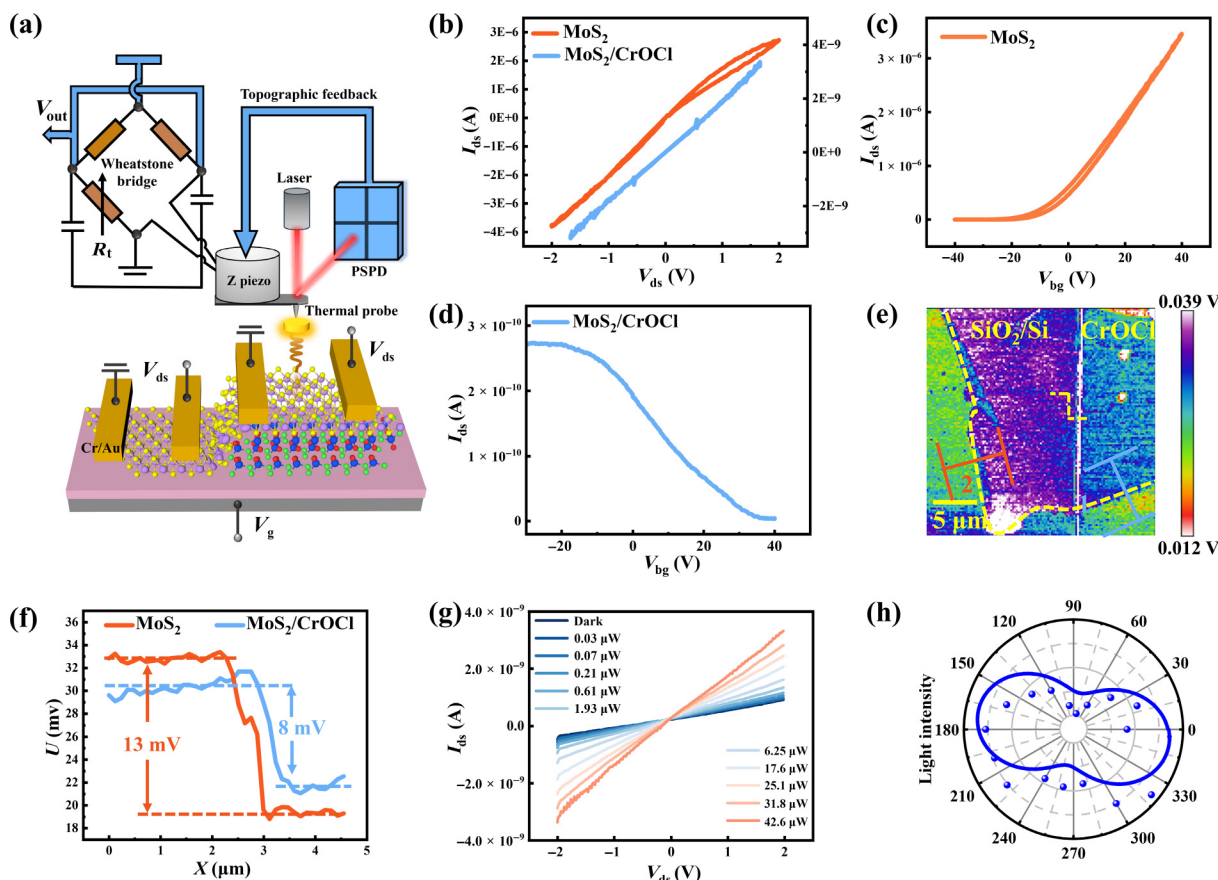


Figure 4 Polarity engineering and electro-thermal modulation in 2L MoS₂ via strong interlayer coupling with CrOCl. (a) Schematic diagram of the 2L-MoS₂/CrOCl heterostructure FET and the SThM configuration. The local temperature is probed using a thermoresistive sensor, whose output signal is read by a Wheatstone bridge circuit. (b) Output characteristics curves (I_{ds} – V_{ds}) of the pristine 2L MoS₂ FET (orange) and 2L-MoS₂/CrOCl heterostructure FET (blue) under low-bias regime, showing near-Ohmic behavior. (c) and (d) Transfer curves (I_{ds} – V_{bg}) of pristine 2L MoS₂ FET and 2L-MoS₂/CrOCl heterostructure FET, respectively, showing a clear polarity inversion from n-type to p-type conduction. (e) SThM image of the pristine MoS₂ region (left) and the MoS₂/CrOCl heterostructure region (right). Scale bar: 5 μ m. (f) Corresponding thermal voltage line-scan profile extracted from (e). (g) Power-dependent photocurrent–voltage (I – V) characteristics of the 2L-MoS₂/CrOCl heterostructure under 532 nm illumination. (h) Angle-dependent photoresponse of the 2L-MoS₂/CrOCl heterostructure (polar plot).

temperature (T) rise via $T = \frac{U}{g \cdot S}$, where g is the lock-in amplifier gain factor [43]. The temperature in the 2L-MoS₂/CrOCl heterostructure was reduced by 38.5% compared with the situation for pristine MoS₂ on SiO₂/Si, confirming a significantly more efficient heat dissipation capability. Furthermore, based on the calibrated SThM contrast across the heterostructure (Figs. 4(e) and 4(f), and Fig. S8 in the ESM), the MoS₂/CrOCl region exhibits a thermal conductivity of $\sim 1.63\times$ higher than that of the pristine MoS₂ region.

These results indicate an interlayer coupling-enhanced heat dissipation pathway in the 2L-MoS₂/CrOCl heterostructure. Interlayer coupling drives interfacial charge transfer and establishes carrier screening, which could strongly renormalize EPC and accelerate electron to phonon energy relaxation [39]. Simultaneously, strong interlayer coupling introduces additional phonon-assisted interlayer energy transfer pathways, thereby enhancing heat flow from MoS₂ into CrOCl. Owing to the intrinsically high and anisotropic thermal conductivity of CrOCl, the injected heat spreads rapidly along the CrOCl layers, leading to a reduction of 38.5% in the local operating temperature of the heterostructure compared to the pristine MoS₂ [21]. Therefore, we established an EPC-linked approach to jointly tune electronic polarity and heat dissipation through the interlayer coupling, which help address challenges associated with both reconfigurable device functionality and thermal reliability in 2D electronics.

Combined with the reduced local temperature rise revealed by our SThM results, this thermal-management capability might help suppress local photothermal accumulation under laser illumination and thereby support more stable photocurrent output in the MoS₂/CrOCl photodetector [44]. Systematic characterizations revealed that the optoelectronic response of the MoS₂/CrOCl heterostructure could be effectively modulated by both the optical power and polarization state of the incident light. Under 532 nm illumination with incident power ranging from 0 to 42.6 μ W, the output photocurrent increases monotonically with optical power (Fig. 4(g)), reaching a maximum responsivity of ~ 3.31 mA/W. This enhancement is primarily attributed to the improved separation and transport efficiency of the photogenerated carriers promoted by the strong interlayer coupling. Angle-resolved measurements reveal a robust two-fold symmetric photoresponse polarization pattern (Fig. 4(h)) with an anisotropy ratio of 1.08, which is attributable to symmetry breaking at the heterointerface. The obtained responsivity and anisotropy ratio are relatively modest compared with some recent related photodetectors [45]. Such performance is still physically reasonable for the present 2L-MoS₂/CrOCl heterostructure, where the polarization-dependent photoresponse mainly arises from interfacial symmetry breaking. The relatively low responsivity further indicates that the device is not yet optimized for maximum photogain. These results therefore support a proof-of-concept demonstration of interface-induced anisotropic photodetection.

3 Conclusions

We systematically investigated thickness-dependent interlayer coupling in MoS₂/CrOCl heterostructures and proposed an EPC-linked interpretation to elucidate how tunable interlayer coupling governs their structure–property relationships. We identify 2L-MoS₂/CrOCl as the strongest coupling configuration, evidenced by

its largest change in mode separation of 1.29 cm⁻¹, and strong SHG suppression from 100% to 11.9% relative to pristine MoS₂. Through the strong interlayer coupling, interfacial charge redistribution downshifts the Fermi level of 2L MoS₂, converting its native n-type character into a p-type conduction, consistent with electrical transport measurements. Beyond polarity inversion, these SThM results further reveal enhanced heat dissipation, with the MoS₂/CrOCl heterostructure temperature rise suppressed by 38.5% compared with the pristine MoS₂ on SiO₂/Si. We attribute this electrical-thermal regulation to an interlayer coupling-enhanced EPC pathway, which reshapes carrier scattering and energy relaxation at the vdW interface. Photoresponse measurements additionally show a power-dependent nonlinear response with a pronounced twofold in-plane symmetry, consistent with anisotropic interfacial coupling. Overall, this work links interlayer coupling to electrical-thermal responses through EPC, providing practical guidelines for codesigning electro-thermal characteristics in vdW heterostructure devices.

4 Experimental section

4.1 Fabrication of MoS₂/CrOCl heterostructures

CrOCl flakes were mechanically exfoliated from bulk CrOCl crystals (Shanghai On-way Technology Co., Ltd.) using blue tape (Sunnano (Taizhou) New Energy Co., Ltd.) and transferred onto the surface of a 285 nm SiO₂/Si substrate doped with phosphorus. MoS₂ flakes (Shanghai On-way Technology Co., Ltd.) of various thicknesses were prepared using the same method. A piece of PDMS was attached to a glass slide. After placing a polyvinyl butyral (PVB) film on the PDMS surface, the glass slide was mounted on a micro-translation stage under an optical microscope with a 3D positioning adjustment platform. The PVB film was then gently brought into contact with the MoS₂ surface. After the PVB film covered the flake, the sample was heated to 60 °C for 2 min. The glass slide was subsequently lifted rapidly, leaving the MoS₂ on the PVB film. The PVB film carrying the MoS₂ was then transferred onto the exfoliated CrOCl flake, completing the heterostructure assembly via thermal release of the PVB film. Electrodes (5/70 nm Cr/Au) for the MoS₂/CrOCl heterostructure devices were fabricated using standard electron-beam lithography (Raith Eline Plus, Germany) and electron-beam evaporation procedures (TECHNOL TEMD500, China). For the representative device, channel length (L) was 7 μ m, channel width (W) was 2 μ m, and contact area (A_c) was 14 μ m².

4.2 Morphological and spectroscopic characterization

The sample morphology was examined using OM (SOPTOP). The thickness and surface potential were characterized using AFM and KPFM (NT-MDT NTEGRA P9, Russia). Raman and PL spectra were acquired using a confocal Raman microscopy system (Witec Alpha 300, Germany) with a 532 nm excitation laser. The laser power was set to 0.8 mW. To obtain high signal-to-noise ratio spectral data, the integration time for a single spectrum was set to 1 s, accumulated over 10 scans. For high-resolution Raman and PL mapping, the integration time per pixel was set to 0.2 s, with 3 points scanned per μ m. SHG response tests were conducted using a confocal Raman microscope system (Witec Alpha 300, Germany) with an incident laser wavelength of 1064 nm. The integration time was set to 0.5 s. The heat evacuation was characterized using SThM

(NT-MDT NTEGRA P9, Russia). All the electrical and photoelectric properties were measured using a probe-station equipped with a Keithly 2636B source meter unit. The 532 nm laser was used as the light source for photoelectric measurement, and its power was adjusted by rotating the light attenuator and verified by a power meter. Note that all the device fabrication and measurements were performed under ambient air condition.

Electronic Supplementary Material: Supplementary material (the orientation determination and stacking strategy of MoS₂/CrOCl heterostructures based on polarization-dependent optical anisotropy (Fig. S1), AFM characterization of the representative CrOCl thin flake (Fig. S2), optical microscopy and Raman spectroscopic characterization of annealed and exfoliated MoS₂/CrOCl heterostructures (Figs. S3 and S5), Raman mapping confirming interfacial uniformity (Fig. S4), photoluminescence spectra, AFM morphology, and scanning thermal microscopy images of the heterostructures (Figs. S6–S8), and Gaussian multi-peak deconvolution parameters of the PL spectra used to evaluate excitonic components and interlayer charge-transfer effects (Table S1)) is available in the online version of this article at <https://doi.org/10.26599/NR.2026.94908743>.

Data availability

All data needed to support the conclusions in the paper are presented in the manuscript and the Electronic Supplementary Material. Additional data related to this paper may be requested from the corresponding author upon request.

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Declaration of competing interest

All the contributing authors report no conflict of interests in this work.

Author contribution statement

C. Y. D. conceived the idea and designed experiments. G. P., W. L., G. W., H. M. Z., and S. F. L. supervised the overall project. W. Q. W., C. Y. Z., and J. Y. fabricated the devices. J. Y. and J. H. X. performed electrical measurements and thermal characterization. T. Z. and J. Y. performed photodetection. J. Y. carried out Raman analysis and discussed on material characterization. C. Y. D., J. Y., and T. Z. conducted data analysis. C. Y. D., J. Y., and G. P. wrote and revised the manuscript. All the authors participated in the discussion of the results.

Use of AI statement

None.

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