Large effective mass and interaction-enhanced Zeeman splitting of $K$-valley electrons in MoSe$_2$

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We study the magnetotransport of high-mobility electrons in monolayer and bilayer MoSe$_2$, which show Shubnikov-de Haas (SdH) oscillations and quantum Hall states in high magnetic fields. An electron effective mass of $0.8m_e$ is extracted from the SdH oscillations' temperature dependence; $m_e$ is the bare electron mass. At a fixed electron density the longitudinal resistance shows minima at filling factors (FFs) that are either predominantly odd, or predominantly even, with a parity that changes as the density is tuned. The SdH oscillations are insensitive to an in-plane magnetic field, consistent with an out-of-plane spin orientation of electrons at the K-point. We attribute the FFs parity transitions to an interaction enhancement of the Zeeman energy as the density is reduced, resulting in an increased Zeeman-to-cyclotron energy ratio.

Group VI transition metal dichalcogenides (TMDs) 1H-monolayers are direct bandgap two-dimensional (2D) semiconductors with band extrema at the corners (K-point) of the hexagonal Brillouin zone [1]. The combination of strong spin-orbit interaction (SOI) and broken inversion symmetry results in a large bandgap at the K-point, and a spin-split bandstructure with coupled spin and valley degrees of freedom [2–4]. Magnetotransport in clean TMD samples can be used to probe the energy-momentum dependence at the band extrema, the Landau level (LL) structure, and assess the impact of electron-electron interaction via negative compressibility or enhanced Zeeman splitting. Shubnikov-de Haas (SdH) oscillations of K-valley holes in mono- and bilayer WSe$_2$ have revealed predominantly two-fold degenerate LLs [5], and interaction-enhanced Zeeman splitting [6, 7]. Similarly, $\Gamma$-valley holes in few-layer WSe$_2$ show large effective masses and enhanced Zeeman splitting [8]. Magnetotransport of 2D electrons in TMDs has been hindered by challenges in obtaining high-mobility samples and low-temperature Ohmic contacts [9]. Magnetotransport in few-layer MoS$_2$ and WS$_2$ samples reveal three or six-fold degenerate LLs, consistent with Q-valley conduction band (CB) extrema [10–12]. Compressibility studies of monolayer WSe$_2$ reveal comparable $K$-valley electron and hole effective masses, and interaction-enhanced LL Zeeman splitting in the valence band (VB), but not in the CB [7].

Here we report a study of SdH oscillations in high-mobility electrons in dual-gated mono- and bilayer MoSe$_2$, using Pd bottom-contacts. From the temperature dependence of the SdH oscillations amplitude, we extract an electron effective mass of $0.8m_e$; $m_e$ is the bare electron mass. We observe predominantly even or odd filling factors (FFs) depending on the electron density ($n$), an observation explained by an interaction-enhanced Zeeman splitting with reducing density. Tilted magnetic-field measurements indicate that the electron spin is locked perpendicular to the MoSe$_2$ plane.

Our devices are fabricated using MoSe$_2$ flakes exfoliated from synthetic crystals (HQ Graphene). Mono- and bilayer flakes are identified using a combination of Raman and photoluminescence (PL) spectroscopy. Figure 1(a) shows the normalized PL spectra for both mono- and bilayer flakes, at room temperature, using an excitation wavelength of 532 nm. The monolayer (bilayer) PL spectrum features a single prominent peak at 1.57 (1.53) eV, associated with the A exciton [13, 14]. Figure 1(b) shows a cross-section schematic of a dual-gated, hBN-encapsulated MoSe$_2$ device. Outlines of different colors mark the MoSe$_2$ flake (red), Pd contacts (green), top (orange) and bottom (black) graphite gates. (d) $R_{xx}$ (left axis) and $R_{xy}$ (right axis) vs $B$ measured at $T = 0.3$ K and $n = 4.9 \times 10^{12}$ cm$^{-2}$ in bilayer MoSe$_2$ B1.

FIG. 1. (a) Normalized room temperature PL spectra of mono- and bilayer MoSe$_2$. (b) Schematic cross-section and (c) optical micrograph of a dual-gated, hBN-encapsulated MoSe$_2$ device. Outlines of different colors mark the MoSe$_2$ flake (red), Pd contacts (green), top (orange) and bottom (black) graphite gates. (d) $R_{xx}$ (left axis) and $R_{xy}$ (right axis) vs $B$ measured at $T = 0.3$ K and $n = 4.9 \times 10^{12}$ cm$^{-2}$ in bilayer MoSe$_2$ B1.
and magnetic fields up to 35 T.

Figure 1(d) shows the longitudinal ($R_{xx}$) and Hall ($R_{xy}$) resistance as a function of the perpendicular magnetic field ($B$) measured in bilayer MoSe$_2$ sample B1 at $n = 4.9 \times 10^{12}$ cm$^{-2}$, and $T = 0.3$ K. The data show SdH oscillations developing at $B > 6$ T, corresponding to a mobility $\mu = 1650$ cm$^2$/Vs. At high $B$-fields quantum Hall states (QHSs) develop at $\nu = 6, 8, 10; \nu = nh/eB$, where $e$ is the electron charge, and $h$ is Planck’s constant. Similar data measured in monolayer MoSe$_2$ sample A1 are included in the Supplemental Material [17].

Figures 2(a,b) show $R_{xx}$ vs $B$ measured at different bottom-gate biases ($V_{BG}$), in monolayer A1 at $V_{TG} = 8$ V, $T = 0.3$ K and in bilayer B2 at $V_{TG} = 6.5$ V, $T = 1.5$ K, respectively. Figures 2(c) and 2(e) show The Fourier transform (FT) amplitude vs frequency corresponding to $R_{xx}$ vs $B^{-1}$ data of Fig. 2(a) and 2(b), respectively. The FT is performed by first subtracting a polynomial background from the $R_{xx}$ vs $B^{-1}$ data to center it around zero, followed by a Hamming window multiplication, and a fast FT algorithm.

Figure 2(c) data, corresponding to monolayer MoSe$_2$, reveal one principal peak at a frequency ($f$) for $V_{BG} \leq 0$ V. For $V_{BG} > 0$ V, $f$ shows a weaker $V_{BG}$ dependence, and a second, lower frequency peak ($f'$) emerges, indicating a second subband is populated. The subband, $(2e/h)f$ and $(2e/h)f'$, and the total $(2e/h)(f + f')$ densities, along with the $n$ values determined from the $R_{xx}$ slope at low $B$-fields are summarized as a function of $V_{BG}$ in Fig. 2(d). The electron density determined from the SdH oscillation frequency is obtained assuming two-fold degenerate LLs. The total $n$ displays a linear dependence on $V_{BG}$. At $n > 12.5 \times 10^{12}$ cm$^{-2}$ the second subband ($f'$) is populated, as marked in Fig. 2(d). The SOI leads to a splitting of the spin-up and spin-down states at the $K$-point in TMDs. This splitting is $\approx 0.2$ eV and $\approx 25$ meV for monolayer MoSe$_2$ VB [1] and CB [4, 18, 19], respectively. We associate the peaks $f$ and $f'$ in Fig. 2(c-d) with the population of the lower and upper CB spin-split bands of monolayer MoSe$_2$, respectively.

Figure 2(e) data, corresponding to bilayer MoSe$_2$, reveal one principal peak at a frequency $f$, and its second harmonic $(2f)$ indicating a single subband is occupied. The $f$ value increases linearly with $V_{BG}$, consistent with Fig. 2(c) data in monolayer MoSe$_2$ with only the lowest spin-split subband populated. Figure 2(f) shows a comparison between $n = (2e/h)f$ calculated using the $f$ values of Fig. 2(e), and the $n$ values determined from the $R_{xy}$ slope at low $B$-fields as a function of $V_{BG}$.

Figure 3(a) shows $R_{xx}$ vs $B$ data measured at various $T$ values, at constant $n = 4.9 \times 10^{12}$ cm$^{-2}$ in bilayer B1. Using the temperature ($T$) dependence of the SdH oscillations amplitude ($\Delta R_{xx}$) we extract the electron effective mass ($m^*$), as $\Delta R_{xx} \propto \xi / \sinh \xi$, where $\xi = 2\pi k_B T / h \omega_c$, and $\omega_c = eB/m^*$; $k_B$ is the Boltzmann constant, and $h$ is the reduced Planck’s constant [17]. Figure 3(b) shows $m^*/m_e$ vs $B$ data for monolayer A1, and bilayer B1, B2 at $n$ ranging between $4.9 - 12.4 \times 10^{12}$ cm$^{-2}$, where only the lower spin-split CB at the $K$-point is probed.
The average $m^*/m_e = 0.8$ is largely insensitive to $n$ and $B$. Theoretical calculations of $m^*/m_e$ in monolayer MoSe$_2$ range between 0.50 – 0.56 [4, 19, 20]. The measured $m^*$ values, and the corresponding density of states $(m^*/\pi\hbar^2)$ allows us to determine the CB spin-splitting $(2\Delta_{cb})$ in monolayer MoSe$_2$. Considering the threshold density for the population of the upper CB subband $n_T = 12.5 \times 10^{12} \text{ cm}^{-2}$ [Fig. 2(d)], we obtain $2\Delta_{cb} = n_T \cdot \pi\hbar^2/m^* = 37$ meV, a value comparable to, albeit larger than theoretical calculations [4, 18, 19].

The CB minima are expected to be at the $K$-point in monolayer, and at the $Q$-point in bulk MoSe$_2$ [21, 22]. The data of Figs. 1-3 allow us to unambiguously determine the CB minima in mono- and bilayer MoSe$_2$. The two-fold LL degeneracy observed in both mono- and bilayer samples is consistent with CB minima at the $K$-point, as SdH oscillations of carriers at the $Q$-point show three- or six-fold degenerate LLs [10, 11]. The similar $m^*$ values of Fig. 3(b) for mono- and bilayer MoSe$_2$ further support this conclusion. In group VI TMD bilayers, the weak inter-layer coupling of $K$-valley carriers leads to two distinct subbands for each layer [5], with densities that can be independently controlled by $V_{FG}$ and $V_{BG}$. For $V_{FG} > 0$ V and $V_{BG} \leq 0$ V only the top layer is populated, and the bilayer MoSe$_2$ can be effectively treated as a monolayer. The absence of a beating pattern in bilayer SdH oscillations up to $n = 11.0 \times 10^{12} \text{ cm}^{-2}$ [Fig. 2(b)] indicates the electrons populate the lower spin-split subband of the top layer.

Figure 4(a) shows $R_{xx}$ vs $n$ at different values between $2.9 - 11.0 \times 10^{12} \text{ cm}^{-2}$ measured in bilayer B2. For $n$ values larger than $8.6 \times 10^{12} \text{ cm}^{-2}$, $R_{xx}$ minima are present at predominantly odd FFs. At $n = 7.0 \times 10^{12} \text{ cm}^{-2}$, the $R_{xx}$ minima at odd and even FFs are of equal strength up to $n = 36$. As $n$ is lowered to $5.6 \times 10^{12} \text{ cm}^{-2}$ the FF sequence turns predominantly even, and at $n = 4.5 \times 10^{12} \text{ cm}^{-2}$ the odd FFs $R_{xx}$ minima are absent. At the lowest $n = 2.9 \times 10^{12} \text{ cm}^{-2}$ another transition to odd FFs is observed. We note that at fixed $n$ the FF sequence is insensitive to changes in the transverse electric field [17].

To better understand the $n$-dependent FF sequence, we write the LLs CB energies $E_{l,\tau s} = \tau s\Delta_{cb} + (l + 1/2)E_c + s g_s\mu_B B/2 + \tau g_s\mu_B B/2$, where $l = 0, 1, 2, \ldots$ is the LL orbital index, $s = \pm 1$ corresponds to the electron spin $\uparrow$ and $\downarrow$, $\tau = \pm 1$ to the $K$ and $K'$ valleys, $E_c = \hbar\omega_c$ is the cyclotron energy, $\mu_B$ is the Bohr magneton, and $g_s, g_c$ are the valley and spin $g$-factors, respectively. The $\tau s\Delta_{cb}$ term describes the spin-split CB minima where the LLs originate. The $\tau s = \pm 1$ doublets lead to two LL fan diagrams with an energy separation of $2\Delta_{cb}$ at $B = 0$. We assume that electrons reside in the lowest spin-split band ($\tau s = -1$), where the total, spin and valley LL Zeeman energy is $E_{2\tau s = -1} = \tau g_s\mu_B B; g^* = g_s - g_c$ is the effective $g$-factor for LLs of the lowest CB spin-split subband. The LL energies of the $\tau s = -1$ group write: $E_{l,\tau} = (l + 1/2)E_c + \tau g^*\mu_B B/2$. We use here the single-band model convention in which all LLs are two-fold degenerate in absence of Zeeman splitting [4, 23]. Using a model in which the $l = 0$ is non-degenerate [3] is equivalent to a $g^*$ offset by $2m_e/m^*$.

The Zeeman-to-cyclotron energy ratio determines the FF sequence, with even (odd) $E_{2\tau s = -1}/E_c$ values leading to even (odd) FFs. Figure 4(a) data reveal a $B$-field independent FF sequence at a fixed $n$, indicating that $E_{2\tau}/E_c$ does not vary with the $B$-field. The FFs parity transitions can be explained by an $n$-dependent $E_{2\tau}/E_c$, or equivalently by an $n$-dependent, interaction enhanced $g^*$. Consistent with the large effective mass, electron-electron interaction is expected to enhance $g^*$ as $n$ is
reduced, as reported in Si [24, 25], GaAs [26], AlAs [27], and WSe$_2$ [6–8] 2D systems.

Magnetotransport in magnetic fields tilted at an angle ($\theta$) from the 2D plane normal [Fig. 4(b) inset] has been employed to probe the Zeeman splitting in 2D systems. If $E_Z$ is proportional to the total magnetic field ($B_T$) the FF sequence changes with $\theta$ [24]. Figure 4(b) shows $R_{xx}$ vs $B$ at various $\theta$ values and $n = 4.5 \times 10^{12}$ cm$^{-2}$ in bilayer B2. At $\theta = 0^\circ$ the FF sequence is predominantly even, and remains unchanged for all $\theta$ values, indicating that $E_Z$ is insensitive to the parallel magnetic field component. These findings contrast observations in Si [24, 25], GaAs [26], AlAs [27], and few layer WSe$_2$ [8] 2D systems, but are in agreement with observations in trilayer MoS$_2$ [11], and mono- and bilayer WSe$_2$ [6], where the combination of strong SOI and band extrema away from the Brillouin zone center locks the carrier spin perpendicular to the 2D system.

Figure 4(c) shows examples of $R_{xx}$ vs $B$ measured in bilayer B3 at low $n$ values. For $n < 4.0 \times 10^{12}$ cm$^{-2}$ the data show QHSs at consecutive FFs above a density-dependent field ($B_p$), where the occupied LLs have the same spin orientation. Interestingly, the observation of consecutive FFs above $B_p$ is accompanied by a pronounced positive magnetoresistance (MR) background superimposed onto the SDH oscillations for $B < B_p$, similar to the positive MR associated with a parallel magnetic-field-induced spin polarization in Si, GaAs and AlAs 2D systems [25, 27, 28].

A quantitative determination of $g^*$ is possible using FF sequence parity data [Fig. 4(a)], and the spin-polarization field [Fig. 4(c)]. Figure 4(d) illustrates the LL structure, where the $E_c$ and $E_Z$ contributions are shown separately for different $E_Z/E_c$ values and FF sequences. Figures 4(e) and 4(f) summarize the FF sequence parity vs $n$ measured in mono- and bilayer samples respectively. Comparing Fig. 4(d) diagram and the FF sequence ($\nu = 4, 5, 7, 9, 11, \ldots$), associated to $R_{xx}$ vs $B$ data measured at $n = 3.4 \times 10^{12}$ cm$^{-2}$ in bilayer B3 [Fig. 4(c)], allows us to assign $E_Z/E_c = 5$ to the lowest $n$ FF parity group of Fig. 4(f). The observation of consecutive integer FFs above a certain magnetic field [Fig. 4(c)] allows to unambiguously assign $E_Z/E_c$. As $n$ is increased, each FF sequence transition is associated with a decrease in $E_Z$ equal to $E_c$ [Fig. 4(e,f)], consistent with a decreasing $g^*$ as the 2D system becomes less dilute. A FF sequence associated with a transition is assigned to a half integer $E_Z/E_c$ value. Once we assign an $i = E_Z/E_c$ value to each FF sequence group [Figs. 4(e,f)], namely $i = 5, 4, 3$, we determine $g^* = (2m_e/m^*)i$ as a function of $n$ as shown in Fig. 4(g,h) for both mono- and bilayer samples, respectively. At the onset of full spin polarization $E_Z$ is equal to the the Fermi energy, and $B_p = 2\hbar n/(e g^* m^* m_e)$ [28]. At low $n$ values the $B_p$ vs $n$ measurement provides a separate method to determine $g^*$ vs $n$. The $g^*$ values obtained from $B_p$ values and FF sequence transitions are summarized in Fig. 4(h) for bilayer samples.

Quantum Monte Carlo (QMC) spin susceptibility calculations [29] have shown good agreement with experiments in GaAs [26] and AlAs [27] 2D electrons, and in WSe$_2$ 2D holes in the $K$-valley [6]. A comparison between the measured $g^*$ and QMC results requires the band $g$-factor value ($g_b$) in absence of interaction effects. As the $g_b$ value remains to be established for MoSe$_2$ [4, 23, 30], we estimate $g_b = 2.2$ using a fit of the QMC spin susceptibility [29] to the experimental $g^*$ vs $n$ data for both mono- [Fig. 4(g)] and bilayer [Fig. 4(h)] samples assuming implicitly the QMC calculations approximate well the interaction enhancement of $g^*$ in MoSe$_2$ as in other 2D systems [6, 26, 27]. The $n$ value is converted into a dimensionless inter-particle distance $r_s = 1/(\sqrt{\pi}\alpha a_B^*$) where $a_B^* = a_B(m_e/m^*)$ is the effective Bohr radius, and $\alpha$ the effective dielectric constant [31]; $a_B$ is the Bohr radius.

In summary, we report magnetotransport studies in high mobility mono- and bilayer MoSe$_2$. The SDH oscillations reveal a density dependent FF sequence, and a $K$-valley electron effective mass of $0.8m_e$. The FF sequence is insensitive to a parallel magnetic field, indicating the electron’s spin is locked perpendicular to the MoSe$_2$ plane. The interplay between cyclotron and Zeeman energy, along with interaction enhanced, density dependent $g$-factor explains the FF sequence odd-to-even transitions. These findings clarify the LL structure of $K$-valley electrons in MoSe$_2$, and highlight the role of interactions in this large effective mass 2D system.

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Supplemental material:
Large effective mass and interaction-enhanced Zeeman splitting of K-valley electrons in MoSe₂

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MAGNETOTRANSPORT IN MONOLAYER MoSe₂

Figure S1 shows the longitudinal resistance ($R_{xx}$) and Hall resistance ($R_{xy}$) as function of the perpendicular magnetic field ($B$) measured in monolayer MoSe₂ A1 at a temperature $T = 0.3$ K, and at an electron density $n = 6.8 \times 10^{12}$ cm$^{-2}$. The data show SdH oscillations developing at $B > 9$ T, corresponding to a mobility $\mu = 1100$ cm$^2$/Vs. At high $B$-fields quantum Hall states (QHSs) develop at $\nu = 8, 10, 12$; $\nu = nh/eB$, where $e$ is the electron charge, $h$ is Planck’s constant. Both Fig. S1 data measured in monolayer MoSe₂, and Fig. 1(d) data measured in bilayer MoSe₂ show QHSs developing at predominantly even filling factors indicating an apparent two-fold Landau level degeneracy.

FILLING FACTOR SEQUENCE TRANSVERSE ELECTRIC FIELD DEPENDANCE

In our samples a set of gate biases determines $n$, and the transverse electric-field $E = \left|C_{TG}V_{TG} - C_{BG}V_{BG}\right|/2\epsilon_0$; $C_{BG}$ ($C_{TG}$) is the bottom (top)-gate capacitance and $\epsilon_0$ is the vacuum permittivity. Figure S2 shows $R_{xx}$ vs $\nu$ measured at different $E$ ranging between $1.30 - 1.74$ V/nm, at a fixed $n = 9.2 \times 10^{12}$ cm$^{-2}$, and $T = 1.5$ K in bilayer MoSe₂ B2. The $R_{xx}$ minima are insensitive to the $E$-field, which suggest an $E$-field independent band and LL structure in the range of values probed here.

![FIG. S1. $R_{xx}$ (left axis) and $R_{xy}$ (right axis) vs $B$ measured at $T = 0.3$ K and $n = 6.8 \times 10^{12}$ cm$^{-2}$ in monolayer MoSe₂ B1.](image)

![FIG. S2. $R_{xx}$ vs $\nu$ measured in bilayer MoSe₂ B2 at $T = 1.5$ K and $n = 9.2 \times 10^{12}$ cm$^{-2}$, and at different $E$-fields. The traces are offset for clarity.](image)

EFFECTIVE MASS EXTRACTION

Figure 3(a) shows the SdH oscillations $T$ dependence at $n = 4.9 \times 10^{12}$ cm$^{-2}$ for bilayer MoSe₂ samples B1, displaying a clear reduction in the oscillations amplitude ($\Delta R_{xx}$) as the $T$ value is increased. The $\Delta R_{xx}$ temperature dependence is proportional to the Dingle factor, $\xi/\sinh\xi$, where $\xi = 2\pi^2k_BT/\hbar\omega_c$, $\omega_c = eB/m^*$ is the electron effective mass, $k_B$ is the Boltzmann constant, and $\hbar$ is the reduced Planck’s constant. To extract $m^*$ we first obtain the FT amplitude spectra for the $R_{xx}$ vs $B^{-1}$ data of Fig. 3(a) [Fig. S3(a)]. A band pass filter centered around $f$, corresponding to shaded region in Fig. S3(a), is applied to eliminate other frequency components. Figure S3(b) shows $\Delta R_{xx}$ vs $B^{-1}$ data at different $T$, obtained by applying an inverse FT to the filtered spectra. The $\Delta R_{xx}$ vs $T$ data at a fixed $B$-field are fit to the Dingle factor to obtain $m^*$, as shown in Fig. S3(b) inset.
FIG. S3. (a) FT amplitude vs frequency for the $R_{xx}$ vs $B^{-1}$ data of Figure 3(a). (b) $\Delta R_{xx}$ vs $B$ calculated from the inverse FT of panel (a) data, using a bandpass filter centered around $f$ [shaded region in panel (a)]. Inset: $\Delta R_{xx}$ vs $T$ data at fixed $B = 13.5$ T, 10.1 T (symbols), and Dingle factor fit to the experimental data (dashed lines).