Interaction-Induced Shubnikov–de Haas Oscillations in Optical Conductivity of Monolayer $MoSe_2$

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We report polarization-resolved resonant reflection spectroscopy of a charge-tunable atomically thin valley semiconductor hosting tightly bound excitons coupled to a dilute system of fully spin- and valley-polarized holes in the presence of a strong magnetic field. We find that exciton-hole interactions manifest themselves in hole-density dependent, Shubnikov–de Haas–like oscillations in the energy and line broadening of the excitonic resonances. These oscillations are evidenced to be precisely correlated with the occupation of Landau levels, thus demonstrating that strong interactions between the excitons and Landau-quantized itinerant carriers enable optical investigation of quantum-Hall physics in transition metal dichalcogenides.

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Optical excitations of a semiconductor hosting a two-dimensional electron (2DES) or hole (2DHS) system subjected to a strong magnetic field provide a direct tool for investigating quantum-Hall states arising from Landau-level (LL) quantization of the carrier orbital motion [1–3]. Over the last three decades, this approach has been widely applied to explore a plethora of fascinating many-body phenomena in III-V or II-VI quantum-well heterostructures, such as modulation of the screening responsible for magneto-oscillations of the excitonic luminescence at integer [4,5] and fractional filling factors ν [6–8], or the formation of collective spin excitations (Skyrmions) around $\nu = 1$ [9,10].

Qualitatively new avenues for the magneto-optical studies of 2DES or 2DHS have emerged with the advent of atomically thin transition metal dichalcogenides (TMD), which are direct band gap semiconductors with two nonequivalent valleys of conduction and valence bands appearing at the $K^{\pm}$ points of the hexagonal Brillouin zone [11–14]. Owing to heavy effective carrier masses and reduced screening, the cyclotron energy in a TMD monolayer is much smaller than the binding energies of the exciton (0.5 eV) or trion (30 meV) [15–18] even at high magnetic fields ~10 T. In this regard, TMD monolayers remain in stark contrast to conventional semiconductors, such as GaAs. Moreover, spin-valley locking by the spin-orbit coupling [19] as well as the presence of the valley-contrasting π Berry phase [12] gives rise to a unique ladder of spin- and valley-polarized LLs in TMD monolayers [20–22], the degeneracy of which is further lifted by the strong Zeeman effect [23,24]. Although the fingerprints of such LLs have been recently reported in several transport experiments [25–30], their optical signatures have been obtained only for a WSe_2 monolayer in the limit where exciton binding is suppressed due to screening; in this regime, the optical excitation spectrum is qualitatively similar to that of GaAs 2DES at moderate fields and displays a multitude of equidistant, band-to-band inter-LL transitions [31].

In this Letter, we report the optical response of a Landau-quantized 2DHS in a $MoSe_2$ monolayer placed in a strong magnetic field of 8–16 T in a fundamentally different limit, where the excitons remain tightly bound and interaction effects are manifest. This is experimentally realized at sufficiently low hole densities $p \lesssim 3 \times 10^{12}$ cm^{-2} (corresponding to a Wigner-Seitz radius of $r_s \gtrsim 5$), at which the screening is too weak to compromise the ultralarge exciton binding in the $MoSe_2$ monolayer. At such densities, the itinerant holes are fully spin- and valley-polarized in the K^+ valley (for $B > 0$) and the optical excitation spectrum is dominated by two cross-circularly polarized resonances corresponding to the bare exciton in the K^+ valley and exciton-polaron in the K^- valley: collective many-body excitation emerging from the attractive interaction between the exciton and a fermionic bath [32–35]. We demonstrate that the transition energies and/or linewidths of both resonances exhibit Shubnikov–de Haas–like (SdH-like) oscillations with the hole density, which are correlated with LL filling, thus providing a first direct evidence for the influence of quantum-Hall states on the excitonic excitations in a TMD monolayer.

Our experiments have been carried out on three different van der Waals heterostructures, each consisting of a gate-controlled $MoSe_2$ monolayer encapsulated between two hexagonal boron-nitride (h-BN) layers. In the main text, we present the data acquired for one of these devices [Fig. 1(a)], where density-dependent oscillations are manifest in the hole-doped regime; while all three devices show such oscillations in the presence of a 2DHS, the third device, described in the Supplemental Material [36], displays analogous oscillations for a 2DES. The valley-selective optical response of all samples has been analyzed by means of low-temperature ($T \approx 4$ K), circular polarization-resolved, white light reflection magneto-spectroscopy (for details, see Supplemental Material [36]).

We first concentrate on the spectral reflectance contrast measured as a function of the gate voltage V_g at $B = 0$, which is shown in Fig. 1(b). For $-7\text{V} \lesssim V_g \lesssim 4$ V, when the sample is devoid of free carriers, the spectrum features only the bare exciton resonance. Once V_g is increased (decreased) beyond this range, free electrons (holes) start to be injected to the monolayer. As demonstrated previously [32–35], the attractive interaction between these carriers and the excitons dress the latter into exciton-polarons, which in turn qualitatively alters the nature of the optical transitions.

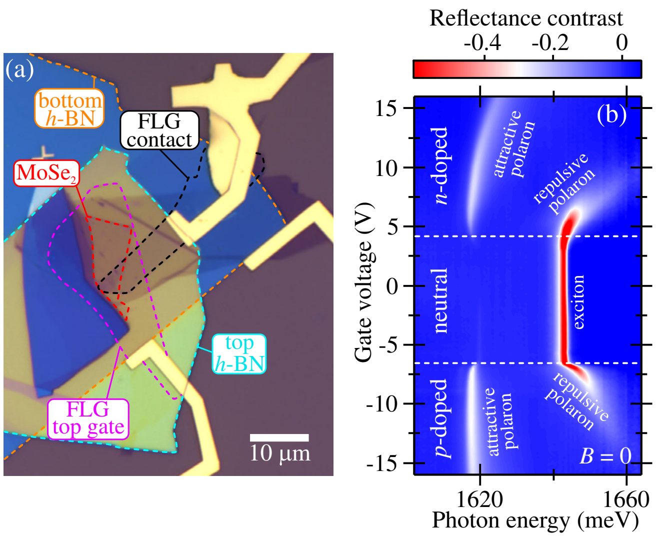

FIG. 1. (a) Optical microscope image of the gate-controlled $MoSe_2$ monolayer studied in this Letter. The monolayer was encapsulated between two layers of h-BN, electrically contacted with a few-layer graphene (FLG) flake, and capped with the second FLG flake serving as a top gate (the boundaries of the flakes are marked with dashed lines). The carrier density was tuned by applying a gate voltage between Au/Ti electrodes connected to the FLG flakes. (b) Color-scale map presenting reflectance contrast spectra measured as a function of the gate voltage at $B = 0$. The horizontal dashed lines mark the transitions between neutral and n- or p-doped regimes.

A prominent signature of this crossover is an emergence of a second, lower-energy resonance originating from an attractive exciton-polaron. Concurrently, the exciton resonance transforms into a repulsive exciton-polaron resonance, which exhibits a strong blue-shift and line broadening. Due to the transfer of oscillator strength to the attractive polaron, the repulsive polaron resonance becomes indiscernible for carrier densities $\gtrsim 1 \times 10^{12}$ cm^{-2}.

Figures 2(a) and 2(b) present the gate-voltage dependence of the reflectance contrast spectra detected in two circular polarizations upon application of the magnetic field of $B = 16$ T perpendicularly to the monolayer plane. Such a field lifts the degeneracy of the electronic states in $K^{\pm}$ valleys due to the Zeeman splitting [see Fig. 2(c)], whose magnitude has been previously found to be significantly enhanced by the exchange interactions in the presence of free carriers [48]. Consequently, at appropriately low densities the electrons (holes) occupy only the states in K^- (K^+) valley. Because the polaron dressing of the excitons arises predominantly due to the intervalley exciton-carrier interaction [33,35,48], at such densities the exciton-polaron resonances appear exclusively in σ^+ (σ^-) polarization, whereas the spectrum in the opposite

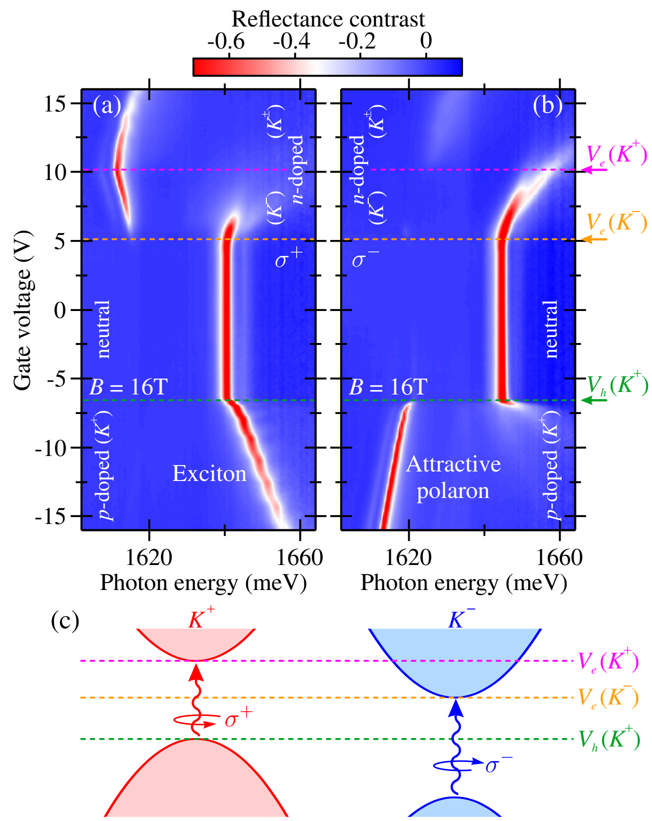

FIG. 2. (a), (b) Reflectance contrast spectra measured at $B = 16$ T as a function of the gate voltage in σ^+ (a) or σ^- (b) circular polarization. (c) Schematic illustrating the lowest-energy electronic subbands at the K^+ and K^- valleys for a $MoSe_2$ monolayer subjected to an external magnetic field. The horizontal dashed lines show the positions of the Fermi level at the crossovers between different doping regimes, which are marked in (a) and (b).

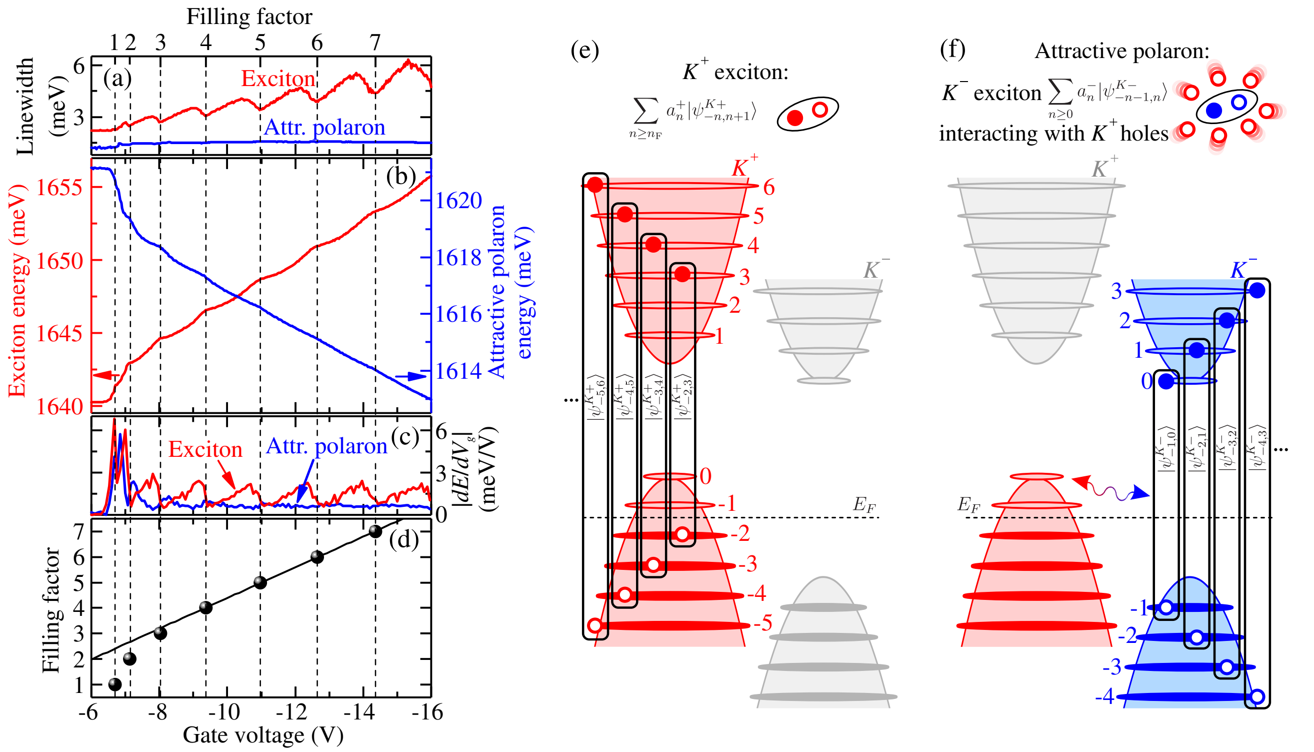

FIG. 3. (a),(b) Gate-voltage dependencies of the linewidths (a) and energies (b) of the exciton (red) and attractive polaron (blue) resonances on the hole-doped side at $B = 16$ T. The data was extracted by fitting the spectral profiles of both resonances in the polarization-resolved reflectance contrast spectra from Figs. 2(a) and 2(b). (c) Absolute values of the derivatives of the energies of both resonances with respect to the gate voltage. (d) Gate voltages V_g corresponding to integer filling factors ν determined based on the positions of the local minima of the exciton linewidth (independently marked by vertical dashed lines). Solid line represents the linear fit to the data for $V_g \lesssim -8$ V, when the Schottky effects at the contact to $MoSe_2$ can be neglected, and hence when the hole density depends linearly on V_g (for details, see the Supplemental Material [36]). (e),(f) Cartoons illustrating the exciton (e) and attractive polaron (f) configurations in the presence of Landau-quantized system of spin-polarized holes occupying only the states in K^+ valley under the influence of external magnetic field $B > 0$.

polarization shows only the bare exciton resonance. As seen in Figs. 2(a) and 2(b), this valley-polarized regime covers the whole experimentally accessible gate-voltage range $-16\text{V} \lesssim V_g \lesssim -7$ V on the hole-doped side, while for the electrons it holds for $5\text{V} \lesssim V_g \lesssim 10$ V. Most importantly, there is a striking difference between these two cases: at electron doping the optical transitions evolve smoothly with V_g, whereas at hole doping they exhibit a pronounced oscillatory behavior. As we show below, these oscillations are due to sequential filling of the hole LLs.

Although we have also observed similar oscillations on the electron-doped side for a better quality sample featuring narrower optical transitions (see the Supplemental Material [36]), they have been found to be much less pronounced than the oscillations at the hole doping for all studied devices. We speculate that this asymmetry may be, at least partially, a consequence of electron-to-hole effective mass ratio that exceeds unity, in contrast to density functional theory (DFT) calculations predicting both masses to be similar [49–51]. This conjecture is supported by the fact that while the hole mass was shown by ARPES measurements [52–55] to remain in excellent agreement with DFT, recent transport measurements of MoS_2 and $MoSe_2$ monolayers indicated that the electron mass may be larger by a factor of ∼2 [28,29].

In the following we focus on the central finding of our work—the signatures of LL filling in the optical spectra of the exciton and attractive polaron transitions in the hole-doped regime (the repulsive polaron transition is not examined, since it disappears at very low hole densities). As seen in Figs. 3(a)–3(c), both resonances exhibit aforementioned oscillatory behavior, which is particularly striking for the exciton, whose linewidth displays sharp, periodic minima [Fig. 3(a)] that appear to be correlated with cusplike changes of the slope of the energy increase with V_g [Figs. 3(b) and 3(c)]. Remarkably, this effect may be independently understood as SdH oscillations in $MoSe_2$ optical conductivity $\sigma(E_X)$ at the excitonic energy [56–59], since $\text{Re}[\sigma(E_X)]$ is determined by the excitonic linewidth [60]. However, the origin of those oscillations is different to that of transport SdH oscillations in static conductivity. Specifically, we find that the oscillations in our system are due to the influence of the LL occupation on the strength of the interactions between the exciton and Landau-quantized

2DHS. This is particularly clear for the case of the exciton linewidth, the narrowing of which arises directly from the modulation of the efficiency of the intravalley exciton-hole coupling. This coupling becomes enhanced each time the lowest-energy LL is only partially filled (around half-integer ν), as in such a case the holes can be effectively scattered between the empty states belonging to this LL. Concurrently, when the Fermi level lies in the gap between the LLs (around integer ν), the possibility of such a hole scattering is substantially reduced due to lack of available final states. This leads to a suppression of the exciton-hole interaction, and thereby gives rise to a pronounced minima in the exciton linewidth.

The above reasoning is corroborated by the results of our theoretical simulations of the exciton absorption spectrum, which, for simplicity, were intended to reproduce the LL-related oscillatory features on a qualitative level only. First, using a variational approach, we calculate the energy of the exciton embedded in the reservoir of valley-polarized holes, taking into account both the phase-space filling and hole-hole exchange interaction. Then, assuming contactlike repulsive intravalley exciton-hole interaction, we calculate (to second order in the interaction) the correlation energy and finite lifetime acquired by the exciton due to dressing with electron-hole pairs from the Fermi sea (for details, see the Supplemental Material [36]). As shown in Figs. 4(a)–4(c), this simple model correctly captures periodic narrowing of the exciton transition around integer ν. Moreover, it also predicts the slope of exciton energy dependence to be altered for the same hole densities, thus explaining the presence of cusplike features in Figs. 3(b) and 3(c). We emphasize that although analogous oscillatory features appear in the model including only the phase-space filling and ignoring interactions [see the black curves in Figs. 4(b) and 4(c)], in such a case the cusps have an opposite direction to that in the experiment: the slope of the exciton energy gets steeper around integer ν instead of becoming flatter. This finding unequivocally demonstrates that the correct description of the experimental data requires inclusion of the interactions, which in turn confirms their primary role in LL-filling-dependent modification of excitonic spectra.

Remarkably, the signatures of LL-filling emerge also for the K^- exciton dressed into an attractive polaron, but are found to be much less pronounced. In fact, they are barely visible in the gate-voltage evolution of the polaron linewidth [Fig. 3(a)] and appear only in the V_g dependence of the energy, taking form of a familiar cusplike features around integer ν that, however, tend to vanish for larger hole densities [Figs. 3(b) and 3(c)]. Although the reason behind this tendency remains not entirely clear, the presence of even faint polaron energy oscillations is by itself supportive of our explanation of the LL signatures to emerge in the optical spectra due to the interactions. This stems from the fact that, unlike the exciton, the polaron transition is not affected by the phase-space filling in the investigated valley-polarized regime [compare the schematics in Figs. 3(e) and 3(f)].

As demonstrated above, the minima of the exciton linewidth occur at gate voltages corresponding to subsequent integer ν, which should imply these voltages $V_g(\nu, B)$ to be equally spaced, as long as the hole density p remains proportional to V_g. Although this prediction holds for

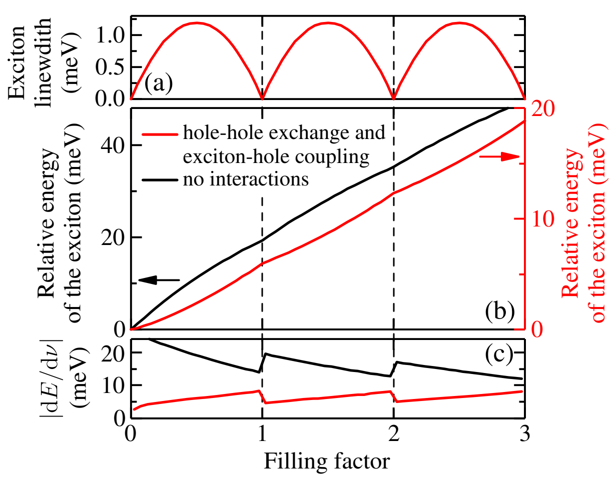

FIG. 4. (a)–(c) Exciton linewidth (a), relative energy (b), and derivative of the energy with respect to ν (c) calculated at $B = 16$ T as a function of the filling factor using the developed theoretical model. The red (black) curves represent the results of the model including (excluding) the interaction effects.

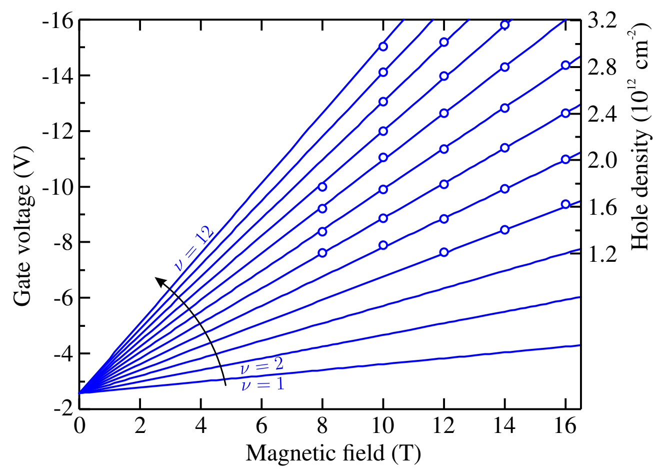

FIG. 5. Gate voltages $V_g(\nu, B)$ corresponding to integer filling factors ν determined based on the positions of the exciton linewidth minima for different magnetic fields B. The data are presented only in a linear-response regime of the gate (i.e., for $V_g \lesssim -8$ V), in which the hole density depends linearly on V_g, and its value (marked on the right axis) can be obtained within the frame of the parallel-plate capacitor model (for details, see the Supplemental Material [36]). Solid lines represent the fit to the experimental data with a set of linear dependencies $V_g(\nu, B) = V_0 - \delta\nu B$ with two fitting parameters: V_0 representing the common origin of these curves, and δ controlling their slopes.

$\nu > 3$ [see Fig. 3(d)], at lower densities the inter-LL voltage gaps turn out to be significantly narrower. We attribute these deviations (that are found to be specific to the investigated device, see the Supplemental Material [36]) to the presence of a Schottky barrier at the contact to the $MoSe_2$, which gives rise to initially nonlinear increase of p with V_g (for details, see the Supplemental Material [36]). This effect, however, becomes negligible at larger-density regime (i.e., for $V_g \lesssim -8$ V), where the voltages $V_g(\nu, B)$ extracted at different fields B indeed scale proportionally to both ν and B, as evidenced by their perfect agreement with a fitted set of linear dependencies forming a characteristic LL fan chart (Fig. 5). The fit allows us to determine the voltage change $\delta \approx 0.1$ V/T needed to fill a LL in a unit magnetic field. On this basis, as well as based on the geometrical capacitance C_{geom} of our device (obtained within a parallel-plate capacitor approximation, see the Supplemental Material [36]), we finally extract the number of states in each LL $C_{\mathrm{geom}}\delta B/e$ that yields $(1.0 \pm 0.2)eB/h$ (where e is the electron charge, while h is the Planck constant). This finding indicates complete lifting of the LL degeneracy, exactly as expected for LLs that are valley and spin polarized.

In conclusion, our experiments provide the first signatures of LL-quantization in the optical spectra of a TMD monolayer hosting a dilute system of spin-valley polarized holes. These signatures are evidenced to emerge due to the influence of LL occupation on the strength of interactions between itinerant holes and tightly bound excitons, which gives rise to prominent filling-factor-dependent SdH-like oscillations in the energy and linewidth of the excitonic transitions. The interaction-enabled optical access to the quantum-Hall physics demonstrated in our work constitutes the first step towards optical investigation of a rich field of strongly correlated phenomena at integer and fractional filling factors in atomically thin semiconductors.

The data that support the findings of this Letter are available in the ETH Research Collection [61].

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