Controlling exciton many-body states by the electric-field effect in monolayer MoS₂J. Klein^{1,2,*}, A. Hötger,¹ M. Florian,³ A. Steinhoff,³ A. Delhomme,⁴ T. Taniguchi,⁵ K. Watanabe^{1,5}, F. Jahnke,³ A. W. Holleitner,¹ M. Potemski⁴, C. Faugeras⁴, J. J. Finley,^{1,†} and A. V. Stier^{1,‡}¹Walter Schottky Institut and Physik Department, Technische Universität München, Am Coulombwall 4, 85748 Garching, Germany²Department of Materials Science and Engineering, Massachusetts Institute of Technology, Cambridge, Massachusetts 02139, USA³Institut für Theoretische Physik, Universität Bremen, P.O. Box 330 440, 28334 Bremen, Germany⁴Université Grenoble Alpes, INSA Toulouse, Univ. Toulouse Paul Sabatier, EMFL, CNRS, LNCMI, 38000 Grenoble, France⁵Research Center for Functional Materials, National Institute for Materials Science, 1-1 Namiki, Tsukuba 305-0044, Japan

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We report magneto-optical spectroscopy of gated monolayer MoS₂ in high magnetic fields up to 28 T and obtain new insights on the many-body interaction of neutral and charged excitons with the resident charges of distinct spin and valley texture. For neutral excitons at low electron doping, we observe a nonlinear valley Zeeman shift due to dipolar spin-interactions that depends sensitively on the local carrier concentration. As the Fermi energy increases to dominate over the other relevant energy scales in the system, the magneto-optical response depends on the occupation of the fully spin-polarized Landau levels (LL) in both K/K' valleys. This manifests itself in a many-body state. Our experiments demonstrate that the exciton in monolayer semiconductors is only a single particle boson close to charge neutrality. We find that away from charge neutrality it smoothly transitions into polaronic states with a distinct spin-valley flavor that is defined by the LL quantized spin and valley texture.

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A bosonic exciton immersed in a Fermi gas is an example of the *impurity model* that can be understood in terms of a polaronic picture—dressing of the exciton by collective excitations of the many-body environment [1–8]. The role of electron-electron interactions on the properties of two-dimensional (2D) electron and hole gases has been extensively studied in GaAs [9–11], AlAs [12,13], and recently also in graphene [14,15]. Typically the spin- and valley susceptibility of the system is found to be enhanced by interactions. Signatures of collective phenomena are particularly pronounced in 2D materials [16–19] such as semiconducting transition metal dichalcogenide (TMDC) MoS₂ owing to strong Coulomb interaction [20,21]. Elegant control knobs, such as static electric-field effects, have been used for tuning many-body interactions for the study of Fermi polarons [22–27]. Specifically in the very low-density regime, valley dichroism in the presence of a magnetic field generates a unique local spin and valley texture of excess charge carriers. Here, only few spin-polarized Landau levels (LLs) with low filling factors are involved in shaping the many-body environment. Therefore, the interaction of excitons immersed in the fermionic

bath depends on those LLs. Quantum transport studies are intrinsically restricted to the high-density regime where LLs with high filling factors are typically probed [28–35], whereas optical measurements facilitate access to the entire density regime [25,27]. The large variation of reported g factors for excitons in MoS₂ [36–40] and other 2D materials [25,27,36,39,41–48] with arbitrary carrier concentration further highlights the necessity for studying valley Zeeman shifts with tunable and controlled carrier densities in 2D semiconductors.

In this Rapid Communication, we fully address the variation of g factors in the literature which is a direct consequence of the many-body interaction with the fermionic bath. We control the carrier concentration n in a dual-gate field-effect device [Fig. 1(a)] and study the magneto-optical response in high magnetic fields up to $B = 28$ T. Simultaneous control of n and B allows us to prepare a unique global spin texture originating from the quantization of excess carriers in fully spin-polarized LLs. We study the magnetic field dependence of neutral and charged excitons (Fermi polarons) which encode the evolution of the total magnetic energy including carrier spin, valley magnetic moment (Berry phase), and cyclotron phenomena arising from quantization of electrons and/or holes into discrete LLs [25,27,41–45,48]. From our measurements, we directly observe that shape and magnitude of the valley Zeeman shift ΔE_{VZ} of excitons very sensitively depend on the spin and valley texture. Our results suggest that the interaction of the exciton with the Fermi-bath at low densities is driven by dipolar spin-interaction which markedly differs from previous observations that have not taken into account the unique LL quantization in 2D TMDCs [22–27].

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FIG. 1. (a) Schematic of the dual-gate monolayer MoS₂ vdW device in Faraday geometry. A monolayer MoS₂ is sandwiched between hBN and contacted with thin graphite (FLG) electrodes. (b) Carrier concentration-dependent low-temperature ($T = 5$ K) μ -PL at $B = -28$ T. Dashed lines show the region of charge neutrality ($n \sim 0 \text{ cm}^{-2}$). Arrows for $B = -28$ T highlight the $+0K$ and $+1K$ LLs in the X^- . (c) Spin and valley exciton band structure at the K and K' points of monolayer MoS₂ for restored time-reversal symmetry ($B = 0$ T). (d) A finite magnetic field ($B \neq 0$ T) breaks time-reversal symmetry inducing an electron spin imbalance ($N_\uparrow \neq N_\downarrow$). For positive magnetic field, the dipolar-spin-interaction is of *intravalley*-type, while for negative magnetic field of *intervalley*-type. (e) Calculated LL fan diagram. (f) Calculated degree of spin-polarization η_s showing the density and magnetic field-dependent spin and valley texture in the conduction bands.

We excite the sample with unpolarized light at $\lambda = 514$ nm and an excitation power of $30 \mu\text{W}$ and detect σ^- -polarized PL at $T = 5$ K. Due to the robust optical selection rules in monolayer TMDCs, we only probe the emission from excitonic recombination in the K' valley for positive and negative polarities of the B field [49]. Figure 1(b) shows a carrier concentration-dependent false color PL map recorded at $B = -28$ T. We symmetrically tune the voltage applied to the top and bottom gates, which effectively counteracts Fermi-level pinning due to the reduction of band tail states [50,51]. This allows us to observe the X^+ transition in the p-charged regime I for the first time in MoS₂. The data reveal narrow emission lines of the X^0 due to the hexagonal boron nitride (hBN) encapsulation [38,52,53] and LL oscillations in the *intravalley* X^- at higher magnetic field. Importantly, another PL feature, labeled X'^- , appears red shifted from X^- at $n_2 \sim 4 \times 10^{12} \text{ cm}^{-2}$. This peak quickly gains oscillator strength with increasing n to dominate the spectrum at the highest

value of n studied. X'^- is commonly observed for n-doping in MoS₂ [28,54] and WSe₂ [55]. It has recently been attributed to a Mahan-like exciton [54], or an exciton-plasmon-like excitation [56], but its precise origin is not yet fully understood. We repeated the measurement for various static magnetic fields ranging from -28 T to 28 T.

Figure 1(c) depicts the spin and valley band sequence of monolayer MoS₂ in an excitonic picture. Electron-hole exchange leads to an optically dark alignment of the spin-orbit split conduction bands c_1 and c_2 ($\Delta_{db} \sim 14 \text{ meV}$), which has been theoretically proposed [57] and experimentally verified [28,58]. Throughout this Rapid Communication, we describe all observed effects on the basis of this picture. For $B = 0$ and E_F situated slightly above the lower conduction band minima (c_1), an equal number of spin-up ($\uparrow$) and spin-down ($\downarrow$) electrons occupy the bands at K and K' ($N_\uparrow = N_\downarrow$) resulting in zero net spin polarization [Fig. 1(c)]. However, an applied magnetic field breaks time-reversal symmetry and shifts the conduction band minima, and the resident electrons condense into LLs resulting in an occupation imbalance between the valleys. A direct consequence is the emergence of a spin-polarized Fermi sea ($N_\uparrow \neq N_\downarrow$) in either K or K' for positive and negative magnetic fields [Fig. 1(d)]. Therefore, the resulting spin-valley texture of the electrons depends on the carrier concentration (E_F) and the direction and magnitude of the applied magnetic field as visualized for monolayer MoS₂ in Fig. 1(f). For other monolayer TMDCs, this depends on the details of the respective band structure. In general, the global degree of spin-polarization $\eta_s = (N_\uparrow - N_\downarrow)/(N_\uparrow + N_\downarrow)$ enters different regimes ranging from $|\eta_s| = \pm 1$ at low carrier concentration and high magnetic fields (regime II), through intermediate densities with $|\eta_s| < 1$ (regime III) to the absence of η_s at highest n where E_F resides well above the minimum of the upper conduction band c_2 (regime IV). For our consideration, we model only the relevant LLs in the conduction bands with a finite inhomogeneous broadening (4 meV) and determine the number of spin $\uparrow$ and spin $\downarrow$ electrons from integrating over the corresponding density of states [see Fig. 1(e) and further details in the Supplemental Material [59]].

An applied magnetic field lifts the K/K' valley degeneracy by shifting time-reversed pairs of states in opposite directions in accord with the Zeeman energy $-\mu_B \cdot B$ [60]. This effect will shift the exciton energy when the magnetic moment of conduction and valence bands are not equal, $\Delta E_{VZ} = -(\mu^c - \mu^v) \cdot B = \frac{1}{2} g \mu_B B$. Figures 2(a) and 2(b) show the magnetic field-dependent X^0 PL close to charge neutrality ($n \sim 0 \text{ cm}^{-2}$) and for low electron densities ($n \sim 1.45 \times 10^{12} \text{ cm}^{-2}$). While ΔE_{VZ} is completely linear at charge neutrality, strikingly it becomes nonlinear with electron doping. The observation is summarized in Fig. 2(c), where the B -field-dependent peak positions for a sequence of charge concentrations are shown.

At charge neutrality, we measure a g factor of X^0 , $g_{X^0} = -1.27 \pm 0.09$. This value is consistent with recent reports in high-quality and hBN-encapsulated MoS₂ [38,40] but contrasts earlier work on nonencapsulated samples [36,37]. Our measured g factor appears to be unusually small compared with other TMDCs where a linear valley Zeeman shift for X^0 near $g \sim -4$ is reported [21,36,39,41–44,61]. In

FIG. 2. (a) and (b) Normalized μ -PL spectra of X^0 for applied magnetic fields at $n = 0 \text{ cm}^{-2}$ ($n = 1.45 \times 10^{12} \text{ cm}^{-2}$) from -28 T to 28 T. The solid lines are a guide to the eye. (c) Carrier density-dependent valley Zeeman shift ΔE_{VZ} of the X^0 in the electron and hole charged regime.

general, the total exciton magnetic moment has contributions from an orbital and a spin component $\mu = \mu^{\text{orb}} + \mu^s$, where $\mu^{\text{orb}} = -\frac{e}{2m_e} l_z$ and $\mu^s = \frac{eg_e}{2m_e} s_z$. Due to the hybridization of the $d_{x^2-y^2}$ and d_{xy} orbitals in the valence band, the Bloch electrons at K/K' provide an angular momentum of $l_z = \pm 2\hbar$ such that an estimate for the valley Zeeman splitting in a simple, noninteracting model is $\mu = -4 \mu_B$. [42] Such g factors are reproduced well by recent first-principle studies including excitonic effects [62,63]. However, as pointed out in Ref. [61] and demonstrated in the supplemental material [59], the decomposition of μ^{orb} in a contribution from intercellular hopping, captured by a lattice model, and atomic intracellular corrections is a delicate problem. Localized orbitals that do not allow for atomlike dipole transitions (e.g., d_{z^2} - and $d_{x^2-y^2}/d_{xy}$ -orbitals in MoS_2) result in a vanishing intracellular correction rendering $\mu^{\text{orb}} = 0$. Breaking of the electron-hole symmetry is expected due to transitions from each band to energetically higher- and lower-lying bands resulting in a finite intercellular contribution. However, even the most sophisticated theoretical approaches to date ignore the many-body nature of the electron-hole pair which we demonstrate here. We further note that the exciton g factor results from couplings of energetically remote bands [64], which may be perturbed by free carriers as well as impurities [51].

The nonlinear valley Zeeman shift in the n-charging regime directly correlates with η_s and is a direct consequence of a dipolar spin-interaction of the exciton with the local spin-polarized fermionic bath. An enhancement in the context of conduction band filling has been observed in WSe_2 at high densities [24]. However, our reported results clearly show

that the interaction is driven by few electrons occupying fully spin-polarized LLs. Similar physics can be found for electrons interacting with local magnetic moments in dilute magnetic semiconductors [65]. In our case, the effective magnetic moment originates from a magnetic field-induced net spin-polarization of excess carriers as depicted schematically in Fig 1(d). Qualitatively, this can be pictured as a paramagnetic spin-ordering effect of conduction band electrons where the MoS_2 effectively undergoes a *ferrimagnetic-to-ferromagnetic* phase transition [34]. When a sufficiently large magnetic field is applied, the valley degeneracy is lifted, which leads to a repopulation of electrons into the lowest conduction band LL. As such a net spin imbalance, $S_z(B) = -1/2 \cdot B_J(x)$, between the valleys occurs at higher B . Here, the Brillouin function [65] is $B_J(x) = \frac{2s_z+1}{2s_z} \coth\left(\frac{2s_z+1}{2s_z}x\right) - \frac{1}{2s_z} \coth\left(\frac{1}{2s_z}x\right)$, where $s_z = 1/2$ is the electron spin in c_1 and $x = g\mu_B s_z B/k_B T$. The net magnetization is then $M \sim \Delta N \cdot S_z(B)$ with the total number of polarized electron spins $\Delta N = |N_\downarrow - N_\uparrow|$. Therefore, the total valley Zeeman g factor for the exciton in regime II can be expressed as the addition of spin and valley net magnetization effects as

$$g^*(n, B) = g_{X^0} + g_m(n) \cdot S_z(B), \quad (1)$$

such that the nonlinear valley Zeeman shift is $\Delta E_{VZ}^{X^0}(n, B) = 1/2 g^*(n, B) \mu_B B$. As shown by the fits to the data in Fig. 2(c), this simple model captures the nonlinear valley Zeeman shift surprisingly well. Note that we do not observe a nonlinearity in the p-charging regime I. This may be expected as interaction effects are more prominent in the conduction band [28].

Finally, we summarize the valley susceptibility of all features in our accessible density range in Fig. 3(a).

Regime I.—The X^0 g factor monotonically increases from ~ -1.27 at charge neutrality to -2.1 at the highest hole density, while the X^+ g factor exhibits a value of $-(3.52 \pm 0.35)$ suggesting that the extra hole adds $\sim 1.5 \mu_B$ to the valley susceptibility of the exciton. Most strikingly, we find that g^* monotonically increases from the value observed close to charge neutrality to the trion g factor, showing the direct dependence of the exciton g factor on the carrier density and strongly pointing toward the exciton having the character of a spin-polarized magnetic polaron at finite electron density.

Regime II.—Introducing electrons to the system and using Eq. (1) to obtain $g^*(n, B)$, the magnetic moment of X^0 [see Fig. 2(c)] increases monotonically with carrier concentration, peaking at $g^* \sim -5.5$ for a density of $n_1 \sim 2 \times 10^{12} \text{ cm}^{-2}$. This confirms our expectation that for $n < n_1$, the increased magnetic susceptibility of X^0 arises from the density of spin-polarized electrons in the lower K' valley [see Fig. 1(f)] and the magnitude of $g^*(n, B)$ is highly sensitive to the local electron concentration. This conclusion may explain the large variation in the literature of reported g factors for excitons in MoS_2 [36–40]. Crucially, $g^*(n, B)$ smoothly approaches the g factor observed for X^- at higher electron concentrations, indicating that both states have a similar spin and valley structure. The X^0 interacts with the spin-polarized Fermi sea, while the strength of the interaction is tuned by the Fermi energy, very similar to the Kondo problem of an isolated impurity spin interacting with a spin-polarized Fermi sea [66]. For high densities, the spin structure of X^0 increasingly resembles that

FIG. 3. (a) Carrier density-dependent g factors of X^0 (black and gray data), the negatively (red and blue data) and positively (green data) charged excitons X^- and X^+ , and the X'^- feature (yellow data). (b) Carrier density dependence of the X'^- binding energy E_B . For $n_2 < n < n_3$ E_B is linear with E_F while it becomes sub-linear for $n > n_3$. For the highest carrier densities (regime IV), the quasi-particle excitation is strongly interacting and dissolving into a many-body state X'_{MB} . (c) Electron concentration dependence of the degree of global spin-polarization η_s for $B = -20 \text{ T}$ and 20 T .

of X^- . Indeed, at the carrier density of n_1 , X^0 sees on average ~ 0.1 electrons within its wave function.

Regime III.—Increasing the electron concentration from moderate (n_1) to high (n_2) densities, we find an asymmetry of the X^- valley Zeeman shift in positive [$X^-(s^+)$] and negative [$X^-(s^-)$] magnetic fields (see Supplemental Fig. S5 in the supplemental material [59]). We attribute this to LL occupation differences in the K/K' valley in the lower conduction bands c_1 [see Fig. 1(e)]. The g factor for negative B increases to ~ -10 . Similar to X^0 , X^- also interacts with electrons in the Fermi sea. However, unlike X^0 , the local spin-valley texture admits electrons residing in both valleys since E_F is located in the lower conduction bands (regime III). This picture is also supported by the spin texture, shown in Fig. 3(c).

Regime IV.—For $n > n_2$, the PL is quickly dominated by the feature X'^- which emerges precisely at the carrier density when E_F shifts into the upper conduction bands c_2 ($n_2 \sim 4 \times 10^{12} \text{ cm}^{-2}$ in our device) [28]. We find that ΔE_{VZ} of X'^- is symmetric and linear in B for all accessible densities [see inset Fig. 3(a)]. The g factor of X'^- equals $g_{X^-(s^-)}$ at n_2 , which suggests a qualitatively similar magnetic moment. Indeed, a bound complex with the excess electron occupying the $\nu = +0K$ LL in the c_2 is such a configuration [see inset Fig. 3(b)]. This is further substantiated by the very strong valley polarization of this feature (see Supplemental Figs. S6 and S7 in the supplemental material [59]). Both, η_s and the g factor of X'^- simultaneously diminish above n_3 with $\eta_s \sim 0$ and a minimum g factor of ~ -5 . Strikingly, at exactly this concentration, a change in the de-

pendence of the X'^- binding energy $E_B = E(X^-) - E(X'^-)$ occurs from a linear to a sublinear dependence on E_F [see Fig. 3(b)]. For $n > n_3$, all valleys and electron spin species are available since E_F is situated well within c_2 . The electron concentration approaches the Mott density [67] where strong many-body effects start to dominate the interaction of X'^- and electrons of all spins forming a strongly dressed many-body state [see inset Fig. 3(b)]. Here the precise local spin structure becomes less relevant, and mean-field-“like” theories become applicable. A detailed description of this state would call for dedicated many-body calculations of the spin susceptibility.

Our results show that all excitons in 2D materials are many-body correlated states that have a magneto-optical response that is sensitive to the local carrier density and related spin and valley textures. We explain the large variation of g factors observed in the literature as arising from lack of control of local doping. The findings of our study represent an important step toward studying and engineering many-body-related phases and novel interaction phenomena in atomically thin materials.

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