



## Rabi oscillations of a quantum dot exciton coupled to acoustic phonons: coherence and population readout

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While the advanced coherent control of qubits is now routinely carried out in low-frequency (gigahertz) systems like single spins, it is far more challenging to achieve for two-level systems in the optical domain. This is because the latter evolve typically in the terahertz range, calling for tools of ultrafast, coherent, nonlinear optics. Using four-wave mixing microspectroscopy, we here measure the optically driven dynamics of a single exciton quantum state confined in a semiconductor quantum dot. In a combined experimental and theoretical approach, we reveal the intrinsic Rabi oscillation dynamics by monitoring both central exciton quantities, i.e., its occupation and the microscopic coherence, as resolved by the four-wave mixing technique. In the frequency domain, this oscillation generates the Autler–Townes splitting of the light-exciton dressed states, directly seen in the four-wave mixing spectra. We further demonstrate that the coupling to acoustic phonons strongly influences the four-wave mixing dynamics on the picosecond time scale, because it leads to transitions between the dressed states. ©2018 Optical Society of America under the terms of the OSA Open Access Publishing Agreement

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### 1. INTRODUCTION

The coherent control of individual spins and excitons in quantum dots (QDs) has been the scope of semiconductor quantum optics for many years. To unveil the potential lying in QDs and other single-photon emitters for high-speed optical quantum technology, it is urgent to bring the manipulation schemes to perfection. This is nowadays approached using individual spins in electrically defined [1] and epitaxial [2,3] QDs, as well as in color centers in diamond [4,5], which is conditioned by setting up robust spin-photon interfacing. This difficulty can be mitigated using QD excitons: because of their direct coupling to light, the related coherent control protocols operate on the picosecond (ps) time scale [6]. The context of ultrafast control points to the subject of our work, focusing on optically driven coherent dynamics of the exciton state efficiently coupled to longitudinal acoustic (LA) phonons of the GaAs host matrix. Previous research has shown that by observing Ramsey fringes [7,8] the quantum state of single localized excitons can be navigated on the Bloch sphere by varying the delay between two laser pulses with ps duration. It has also been demonstrated that by increasing the applied laser power, the final exciton occupation after a single pulse can be adjusted [9,10] by performing Rabi rotations. Excitations with chirped laser

pulses have been used to realize robust state preparations via the so called rapid adiabatic passage effect [11–17]. A pump-probe experiment [18] and time-resolved resonance fluorescence [19] have been used to study the optically driven Rabi oscillation dynamics of the QD exciton occupation. Rabi oscillations have also been investigated in larger systems, i.e., in QD ensembles [20–22], quantum wells [23,24], and quantum dash ensembles [25]. Note that these Rabi oscillations have to be differentiated from Rabi rotations [9,10,26–31]. The oscillations directly correspond to the dynamical aspect of the genuine Rabi problem [32]. Rabi rotations instead reflect the final exciton state and thus lack direct visibility of the temporal interplay between exciton and phonons.

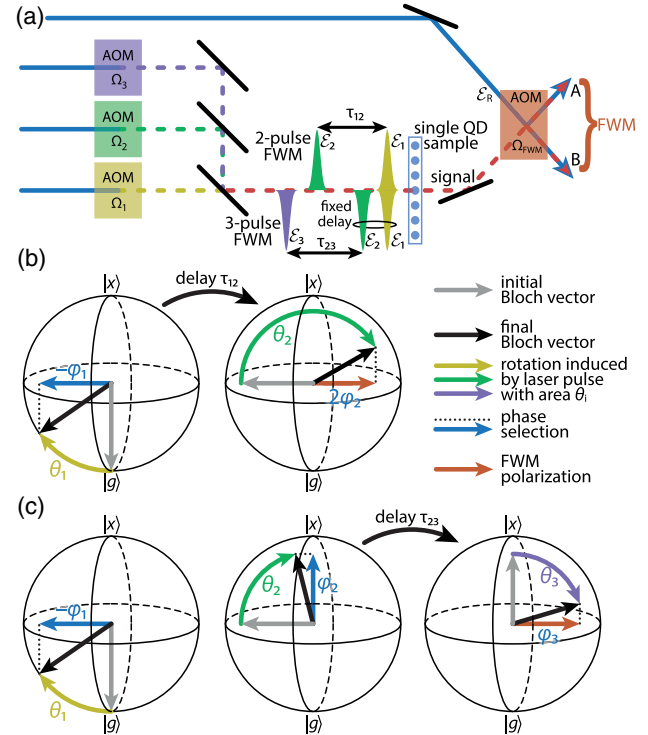
We here propose and realize an original approach to monitor a single exciton's Rabi oscillations in the time domain. Namely, we use the delay dependence of four-wave mixing (FWM) signals to probe the dynamics of both central quantum mechanical quantities of a single QD exciton, i.e., its occupation and its microscopic coherence, which represent the entire quantum state or its Bloch vector. Note that combined with the time-dependence of the emitted signal, this delay dependence can be used to generate two-dimensional spectra. These provide a rich playground to

study spectral features of coupled and uncoupled quantum systems [33–39]. Here, however, our focus is on the temporal evolution of the exciton quantum state. Our dual insight into Rabi oscillation dynamics of a single QD exciton represents a novel aspect and is enabled by the FWM methodology. In a combined experimental and theoretical study, we investigate the dynamics of both quantities in FWM signals. When increasing the intensities of the exciting laser pulses, oscillations on the ps time scale build up during the optical driving. The measured oscillations directly reflect Rabi oscillations of the Bloch vector of an individual QD exciton. In exciton ensembles, the isolation of a single Bloch vector has so far not been realized, because one always deals with a bunch of Bloch vectors undergoing different dynamics. Carrying out this study in the ultimate limit of a single QD exciton allows for an investigation of the fundamental interplay between the optically driven two-level system and its coupling to the host lattice via the exciton-phonon interaction.

## 2. EXPERIMENT AND THEORY

To measure FWM on individual QDs, we employ a three-beam heterodyne spectral interferometry setup optimized for the near-infrared spectral range [38], as schematically depicted in Fig. 1. The FWM is driven using a Ti:Sapphire femtosecond laser. The QD exciton is excited by Gaussian laser pulses, formed by a passive pulse shaper based on a diffraction grating. The pulse train is (i) split into three beams  $\mathcal{E}_i$  ( $i = 1, 2, 3$ ) and a reference beam  $\mathcal{E}_R$ ; (ii) the beams  $\mathcal{E}_i$  are individually phase-modulated using acousto-optic modulators (AOMs) operating at distinct radio frequencies  $\Omega_i$  around 80 MHz; (iii) the pulses  $\mathcal{E}_{1,2,3}$  are delayed with respect to one another by  $\tau_{12}$  and  $\tau_{23}$ ; (iv) their intensities  $P_i \sim |\mathcal{E}_i|^2$ , which are varied to scan the pulse areas  $\theta_i$ , are controlled via fixed radio-frequency drivers. After steps (i)–(iv),  $\mathcal{E}_{1,2,3}$  propagate collinearly and are focused on the sample surface. The pulse shaper is also used to geometrically compensate the first-order chirp, attaining a focal spot close to the Fourier limit. FWM is retrieved in reflectance [note that, for the sake of readability, Fig. 1(a) shows a transmission configuration] by locking  $\mathcal{E}_R$  at the specific heterodyne frequency ( $\Omega_{\text{FWM}} = 2\Omega_2 - \Omega_1$  or  $\Omega_3 + \Omega_2 - \Omega_1$ ) and interfering it with the signal beam from the sample [red in Fig. 1(a)], which is phase-modulated with the same heterodyne frequency as  $\mathcal{E}_R$ . The FWM signal is retrieved by subtracting the intensities in channels A and B from each other, attaining the shot-noise limited detection.  $\mathcal{E}_R$  is also used to generate background free spectral interference on a CCD camera installed at the output of an imaging spectrometer.

In these experiments, the  $\mathcal{E}_{1,2,3}$  pulse durations, hereafter denoted as  $\tau$ , arriving at the QD position are in a few hundred femtoseconds up to ten ps range. Therefore, the theoretical treatment of the optical excitation has to go beyond the computationally handy ultrashort-pulse limit, where analytic expressions for the FWM signals can be derived [38,40]. To simulate the FWM signals of the optically driven QD, we model the QD exciton as a two-level system coupled to LA phonons via the deformation potential mechanism [41]. The equations of motion for the occupation and the polarization are then obtained with the well-established correlation-expansion approach [42]. The full set of equations of motion are given in Ref. [43]. Practically, to compute the FWM signal we drive the QD exciton with Gaussian laser pulses:



**Fig. 1.** Schematic picture of the experiment and the theory. (a) Experimental setup. AOMs label the excitation pulses with radio frequencies  $\Omega_i$ . The signal beam after the sample is mixed in an AOM with the reference beam into channels A and B to generate the stationary spectral interference at the CCD camera. FWM is detected background free by subtracting A and B. Note that the actual experiment is working in reflection from the sample. To keep the picture as simple as possible, the figure shows transmission geometry. (b), (c) Schematic picture of the theoretical simulation of the exciton state on the Bloch sphere. After each pulse with pulse area  $\theta_i$  the state is filtered with respect to phase factor  $\varphi_i$  corresponding to the final FWM phase  $\varphi_{\text{FWM}}$ . (b) For two-pulse FWM where  $\tau_{12}$  is scanned and (c) for three-pulse FWM where  $\tau_{23}$  changes.

$$\mathcal{E}_i(t) = \frac{\hbar}{2\sqrt{2}\pi\mu\tau} \exp \left[ -\frac{1}{2} \left( \frac{t-t_i}{\tau} \right)^2 - i\omega_L t + i\varphi_i \right],$$

in general with  $i = 1, 2, 3$ , the pulse areas  $\theta_i$ , the dipole moment  $\mu$ , and the pulse duration  $\tau$  at times  $t_i$ . The central energy is  $\hbar\omega_L$ , which is chosen to be in resonance with the exciton transition. We label each pulse with a phase  $\varphi_i$  with  $i = 1, 2$  for two-pulse and with  $i = 1, 2, 3$  for three-pulse FWM. These phases  $\varphi_i$  correspond to the radio-frequency shifts  $\Omega_i$  in our experimental heterodyne detection scheme. The FWM signal  $S_{\text{FWM}}$  is then isolated by filtering the resulting microscopic polarization  $p = \langle |g\rangle \langle x| \rangle$  with respect to the FWM phases  $\varphi_{\text{FWM}} = 2\varphi_2 - \varphi_1$  or  $\varphi_{\text{FWM}} = \varphi_3 + \varphi_2 - \varphi_1$ , resulting in the FWM polarization  $p_{\text{FWM}} \sim S_{\text{FWM}}$ . Here,  $|g\rangle$  denotes the ground state and  $|x\rangle$  the exciton state. Note that this filtering of the signal with respect to the phases directly mimics the heterodyne detection scheme. In particular, although both in experiment and in theory the lowest order contribution to the FWM signal is of the order  $\chi^{(3)}$ , the signals are not limited to this order. Instead, they include all nonlinear odd orders of the susceptibility  $\chi$ . This allows us to directly resolve Rabi oscillations both in experiment and in theory. By varying the delay  $\tau_{12} = t_2 - t_1$  in two-pulse FWM and  $\tau_{23} = t_3 - t_2$  in three-pulse FWM, we probe the coherence

and population dynamics, respectively. This can be seen from the Bloch sphere (spanned by the real and imaginary part of  $p$  and the exciton occupation  $f = \langle |x\rangle\langle x| \rangle$ ) schematics in Figs. 1(b) and 1(c). Due to the excitation with phase-modulated laser pulse train, for each repetition, the Bloch vector switches from its initial (gray) to its final state (black). The curved arrows represent the pulse areas of each driving pulse. The successive selection of the FWM phase after each pulse results in the final FWM polarization (orange arrow). In the experiment, optical heterodyning allows us to probe a FWM representation of the Bloch vector dynamics in its rotating frame. Changing the delay  $\tau_{12}(\tau_{23})$  probes the dynamics of the polarization (population) in the case of two-pulse (three-pulse) FWM, as can be seen from the selected Bloch vector (dotted black line and blue arrow) directly before  $\tau_{12}(\tau_{23})$ . The simulations reaching delay times over 100 ps are computationally demanding, which makes finding appropriate system parameters such as pulse areas, pulse duration, and QD geometry quite challenging. Note that we add a phenomenological long-time dephasing rate  $\beta$  for the polarization and a spontaneous decay rate  $\gamma$  for the exciton occupation to the model when simulating dynamics in the 100 ps range.

### 3. RESULTS

We have recently shown that the pulse areas  $\theta_i$  of the driving laser fields in FWM experiments have a strong influence on the system's dynamics between the pulses. For instance, the visibility of quantum beats, arising due to the splittings in the excitonic level structure of a QD system, depends on the pulse area [38]. Furthermore, in a two-pulse FWM experiment, the strength of the phonon-induced dephasing (PID) becomes more pronounced when increasing  $\theta_1$  towards  $\pi$  [29].

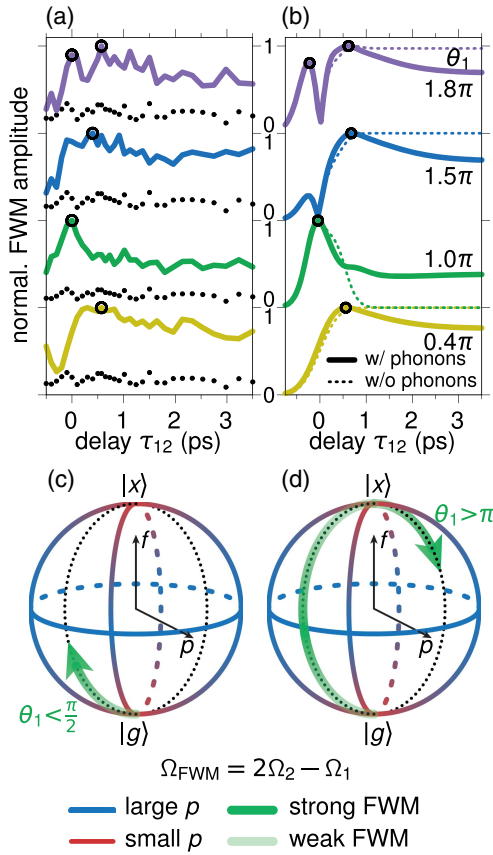
The pulse area dependence of the exciton quantum state is rather obvious, when considering the dynamics during the interaction with the laser field, i.e., Rabi oscillations take place. When keeping  $\tau$  fixed and increasing  $P_i$  and therefore  $\theta_i$ , the exciton is excited and de-excited during the pulse more often. In the picture of the Bloch vector, it performs more rotations, which means that the rotation speed, i.e., the Rabi frequency  $\Omega_R$ , increases. Additionally, the coupling between exciton dynamics and phonons depends on the instantaneous Rabi frequency and the phonon spectral density  $J(\omega_{ph})$ . For the deformation potential coupling between the QD exciton and LA phonons  $J(\omega_{ph})$  scales like  $\omega_{ph}^3$  for small phonon frequencies  $\omega_{ph}$  [44]. For typical self-assembled InGaAs/GaAs QDs with sizes in the range of a few nanometers, the spectral density forms a broad maximum at  $\omega_{ph,0}$  in the range of a few  $\text{ps}^{-1}$ , i.e., a few milli-electron volts [45]. The upper cutoff frequency is roughly given by  $\omega_{ph,max} = 2c/a$ , where  $c$  is the sound velocity and  $a$  the localization length of the exciton. Therefore, the strongest interaction between exciton and LA phonons lies in the range of  $\Omega_R \approx \omega_{ph,0}$ .

In the limit of ultrafast laser pulse excitation, in the range of  $\tau \approx 100$  fs, we have shown that simulations in the delta-pulse limit yield a satisfactory agreement with experiments [38,46]. In this limit, the properties of the excited phonons only depend on the final occupation of the exciton state [47]. Therefore, to sense the PID effects related with the variation of the pulse area, one needs to work with longer pulses. With this aim, we spectrally shape the initial laser beam, setting durations of  $\tau \approx 300$  fs. To enhance the influence of LA phonons, we set the temperature to

$T = 23$  K, increasing the phonon occupation of modes in the range of 7 meV by a factor of  $7 \times 10^6$  with respect to 4.2 K, while keeping sufficiently long dephasing of the zero-phonon line [46,48].  $T = 23$  K is also considered in the simulations. We first present the results obtained on a neutral exciton in an InAs QD embedded in a planar cavity, exhibiting a low quality factor  $Q_{\text{planar}} \approx 1.7 \times 10^2$ , as recently employed in Refs. [6,29,38]. The layer of annealed and capped InAs QDs (density  $2 \times 10^9 \text{ cm}^{-2}$ ) is placed in the center of a GaAs spacer. The spectrum of the driving laser pulses is tuned to cover the ground state to exciton transition as shown in Fig. S1(a) in Supplement 1. Also, the exciton to biexciton transition is slightly covered by the tail of the pulse spectrum, generating small signals for negative delays via the two-photon coherence [see Fig. S1(a) in Supplement 1]. A strong influence from the biexciton state would also lead to a beating for positive delays due to the biexciton binding energy [38]. Because the pulse spectrum has only little overlap with the exciton to biexciton transition, we do not resolve this beating for positive delays. Therefore, we conclude that the influence of the biexciton for the signals at positive delay times is negligible and neglect the biexciton state in our study, restricting it to a two-level system. In Figs. 2(a) and 2(b), we plot the two-pulse FWM amplitude as a function of  $\tau_{12}$ . The FWM phase is given by  $\varphi_{\text{FWM}} = 2\varphi_2 - \varphi_1$ , meaning that the first pulse creates a coherence and the second pulse converts this coherence into the FWM signal [see Fig. 1(b)]. We show results for four different pulse areas  $\theta_1$  at a fixed second pulse area of  $\theta_2 = \pi$ . Figure 2(a) presents the measurement and Fig. 2(b) the respective simulation results for small to large pulse areas from bottom to top. Note that all curves are normalized to unity. The pulse areas in the calculations are listed in the plot next to the respective graph. For the geometry of the QD, we choose slightly different parameters than in Ref. [29]. Here, the electron and hole localization lengths are  $a_e = 7$  nm and  $a_h = 1.5$  nm, respectively. These parameters give the best agreement between the measured data in Fig. 2(a) and the simulations in Fig. 2(b). Note that the dimensions of the exciton  $a_e$  and  $a_h$  are a spherical representation of the exciton, which leads to the same physical results as a lens-shaped model [49]. Therefore, these sizes must not be seen as the real size of the exciton wave function.

We start the discussion with the smallest measured pulse area at the bottom in Fig. 2(a) (yellow). The FWM amplitude builds up around  $\tau_{12} = 0$  and reaches a maximum at around 0.5 ps (marked by the circle). After that, the signal slightly decays within the next 3 ps. This behavior is well reproduced by the simulation in Fig. 2(b), including the coupling to LA phonons (solid) for  $\theta_1 = 0.4\pi$ . The drop of the signal within the first few ps results from the PID effect. Together, the phonons and the exciton in the QD form a new equilibrium state, the acoustic polaron. This polaron is accompanied by a static lattice displacement in the vicinity of the QD [50]. When the exciton, and therefore the polaron, is created faster than the typical time scale of the involved phonons, a phonon wave packet is emitted. This results in an irreversible loss of exciton coherence [46,51,52]. To emphasize that the signal drop results from the phonon coupling, we also provide simulations neglecting the exciton-phonon interaction, resulting in the dashed yellow curve in Fig. 2(b), visibly lacking the initial decay due to the PID effect.

Going over to the next larger pulse area in Fig. 2(a) (green), we clearly see that the drop of the measured FWM signal is significantly increased and reaches an almost stationary value within less



**Fig. 2.** Normalized two-pulse FWM generated with subps pulses on a QD exciton embedded in a low- $Q$  cavity. (a), (b) FWM amplitude as a function of the delay  $\tau_{12}$  for increasing pulse area of the first pulse from bottom to top. The FWM delay dependence probes the optically driven evolution of the exciton polarization, performing Rabi oscillations for excitations with high pulse areas. (a) Experiment, the  $P_1$  impinging the sample surface are (0.14, 1.1, 2.3, 2.7)  $\mu\text{W}$ ,  $P_2 = 1 \mu\text{W}$ , temperature  $T = 23 \text{ K}$ . The dots indicate the noise level. (b) Theory, solid lines with phonon coupling dotted lines without phonons.  $\theta_1$  as given in the plot. The circles mark maxima of the FWM signal. (c), (d) Schematic pictures of the FWM signal on the Bloch sphere; (c) for small pulse areas  $\theta_1$ ; (d) for pulse areas  $\theta_1$  exceeding  $\pi$ .

than 1 ps. Additionally, the circle shows that the maximum of the signal shifts to shorter delays. These findings are consistent with the simulation for  $\theta_1 = 1.0\pi$  in Fig. 2(b) (solid green). However, the calculated FWM signal without the exciton-phonon coupling (dashed green) goes exactly to zero after  $\tau_{12} = 1 \text{ ps}$ . To explain this, we take a look at the schematic Bloch sphere picture in Fig. 2(c), where the green arrow visualizes the movement of the Bloch vector during the first laser pulse excitation. When the first pulse area is small, i.e.,  $\theta_1 < \pi/2$ , the microscopic polarization  $p$  is maximal at the end of the pulse. Therefore, also the two-pulse FWM signal is maximal for small positive delays, i.e., after the first pulse. This is the case for the dashed yellow curve. However, for pulse areas  $\pi/2 < \theta_1 < \pi$ , the Bloch vector crosses the equator of the Bloch sphere, i.e., the state of maximum coherence, during the pulse. For the special case of a  $\pi$  pulse, the final polarization is zero, leading also to a vanishing FWM signal. Already here we see that the dynamics of the FWM signal resembles more involved movements of the Bloch vector during the action of the first laser pulse. The full model,

including the coupling to LA phonons, is shown as a solid green line in Fig. 2(b). We see that the stationary value of the curve strongly deviates from the dashed line. This mismatch is again a consequence of the PID effect. Going from a  $0.4\pi$  pulse to a  $\pi$ -pulse excitation, the dephasing influence of the phonon coupling should significantly increase because the final exciton occupation  $f$  is approximately twice as large [45,53]. Comparing the solid lines with the dashed lines of the same color (yellow and green), we see that this is clearly the case. A stronger excitation leads to a larger phonon wave packet and therefore to stronger dephasing.

When increasing the pulse area further, in the second curve from the top in Fig. 2(a) (blue), the signal drop decreases again, and the maximum of the signal also goes back to longer  $\tau_{12} \approx 0.5 \text{ ps}$ , as marked by the circle. We find the best agreement with the experiment for the simulation with  $\theta_1 = 1.5\pi$  in Fig. 2(b) (solid blue). The dynamics of the simulated signals, both with and without the exciton-phonon interaction, look very much like the  $\theta_1 = 0.4\pi$  case (yellow). The only significant difference of the blue curve is an additional minimum at  $\tau_{12} = 0 \text{ ps}$ . This is again a result of the Bloch vector dynamics. We are now dealing with the  $\theta_1 > \pi$  case, schematically shown in Fig. 2(d). The Bloch vector crosses the north pole of the sphere during the pulse, which leads to null polarization, and, in consequence, to a vanishing FWM signal. After 1 ps, the solid blue line in Fig. 2(b) is again governed by the PID drop of the signal. The additional minimum of the FWM signal around  $\tau_{12} = 0 \text{ ps}$  is obviously too unpronounced to be clearly resolved in the experiment in Fig. 2(a). When comparing the simulation with and without phonon coupling, we find that the drop of the signal, i.e., the PID effect, gets weaker with respect to the pulse area discussed above. This is in line with the previous explanation of the strength of the PID effect. For this pulse area, the final exciton occupation is smaller than for the  $\pi$  pulse, which leads to weaker dephasing.

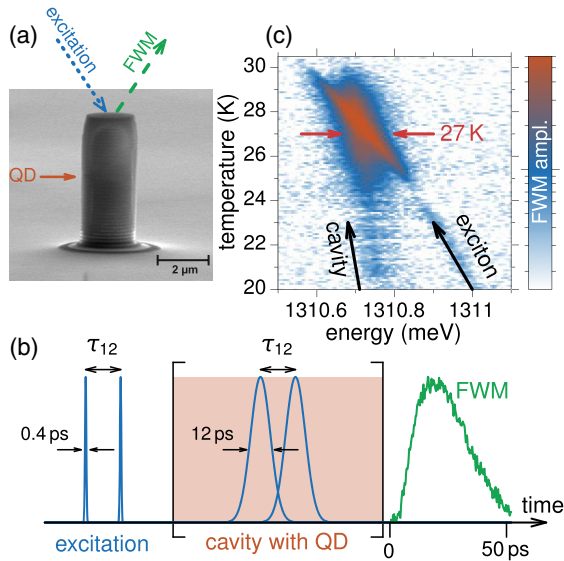
When we move to the largest considered pulse area at the top (violet), a clear minimum shows up in the measured signal in Fig. 2(a). This is in excellent agreement with the simulation for  $\theta_1 = 1.8\pi$  in Fig. 2(b) (solid violet). Here, the signal minimum is significantly more pronounced than in the case considered before. This is an instructive demonstration of the optically driven Rabi oscillations of the exciton state. The PID drop after  $\tau_{12} = 0.5 \text{ ps}$  is of a comparable strength as for the  $1.5\pi$  case in blue.

From the pulse area series in Figs. 2(a) and 2(b), we find that the effect of the PID and therefore the influence of the exciton-phonon interaction changes measurably with the pulse area  $\theta_1$  of the first driving laser pulse. For large positive delays, i.e.,  $\tau_{12} > \tau$ , pulse one arrives first, while for large negative delays, pulse two arrives first. Therefore, around  $\tau_{12} = 0$ , both pulses overlap. The additional dynamics evolving around  $\tau_{12} = 0$  happen during the presence of the first pulse. Because the  $\tau_{12}$  dependence of the two-pulse FWM signal represents the dynamics of the coherence, i.e., the microscopic polarization of the exciton  $p$ , the resolved oscillations for the largest considered pulse area stem from optically driven Rabi oscillations in the time domain.

It was shown that the exciton-phonon interaction leads to more involved movements of the Bloch vector during optical driving. The phonon lead to (i) dephasing, i.e., shrinking of the Bloch vector length and (ii) a mixture of real and imaginary part of the polarization [54], i.e., a movement out of the Rabi oscillation plane of the Bloch vector. For simplicity, the illustrating pictures

in Figs. 2(c) and 2(d) do not take these complexities into account. Instead, they capture well the origin of the Rabi oscillations observable in the FWM delay dynamics. Following this proof of principle demonstration, we now generalize this approach using a more suitable photonic nanostructure.

Shifting the focus from the PID phenomenon to the Rabi oscillations, we aim to drive the QD with laser pulses that are as long as possible, to stretch the dynamics in time. In parallel, we want to reach preferably large pulse areas, to generate as many Rabi periods as possible. Our FWM methodology relies on the detection of spectrally resolved interference between the QD emission and the reference  $\mathcal{E}_R$ , requiring a bandwidth well beyond the spectrometer resolution, in practice over 0.1 meV. As such, the experiment is limited to pulse durations  $\tau$  in the few ps range. To overcome this issue, we switch to a micropillar cavity [55,56] with a diameter of 1.8  $\mu\text{m}$  and a height around 10  $\mu\text{m}$ , as depicted in Fig. 3(a), which yields a 2 orders of magnitude higher quality factor of  $Q_{\text{pillar}} \simeq 2.4 \times 10^4$ . This structure, containing a layer of InAs QDs at the antinode of the cavity mode, allows us to access the photonic structure with subps pulses (namely,  $\tau \simeq 0.4$  ps). See Supplement 1 for spectra of spectral interference and laser pulses. We tightly focus them onto the top facet of the pillar and construct spectral interferences with  $\mathcal{E}_R$ , placed 12  $\mu\text{m}$  apart at the auxiliary pillar [56]. Crucially, due to the high quality factor, the optical field reaches the QD in the cavity in a retarded way. This stretches the pulses to durations of over 10 ps, as schematically shown in Fig. 3(b). Figure 3(c) shows a temperature scan of the FWM spectrum of the QD cavity system (a corresponding scan of the photoluminescence intensity is shown in Supplement 1). The resonances of exciton and cavity are marked in the picture. Around  $T = 27$  K both resonances cross, which shows that the coupled QD cavity system operates in the weak coupling



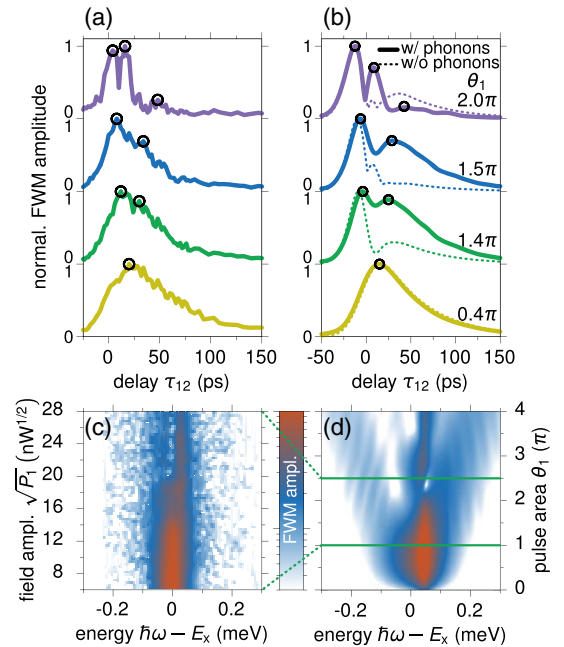
**Fig. 3.** Micropillar cavity system. (a) Scanning electron microscopy image of an exemplary micropillar cavity system with a diameter of 1.8  $\mu\text{m}$  and a height around 10  $\mu\text{m}$ ; (b) FWM spectra for varying temperatures, demonstrating operation in the weak coupling regime: exciton and cavity resonances shift in energy and cross at  $T \approx 27$  K; (c) schematic picture of the driving laser pulses and the measured FWM dynamics. The effective pulse duration  $\tau$  is increased by a factor of 30 inside the cavity. Green trace is the measured time-resolved FWM field (vertical logarithmic scale), illustrating its buildup owing to the high Q factor.

regime [55,57,58]. The possibility of adjusting the detuning very precisely makes this system prototypical to demonstrate the Purcell effect [59,60]. We do so by applying a three-pulse FWM measurement for different detunings between the cavity mode and the exciton transition. The results are shown in Supplement 1.

The enhancement of the intracavity field in the pillar structure allows for reaching larger  $\theta_i$  with respect to the low Q-factor planar cavity explored in Fig. 2. In the following, we choose the temperature to  $T = 27$  K, setting the exciton transition to be approximately in resonance with the cavity mode, as marked by the red arrows in Fig. 3(c). Working at these elevated temperatures results in a significant increase of the LA phonon influence than at 5 K [46]. In all simulations, we choose the temperature to be 25 K.

The results for the two-pulse FWM study are shown in Fig. 4 with the measurements in Fig. 4(a) and the simulations in Fig. 4(b). All curves are normalized to their respective maximum. We consider four different pulse areas, increasing from bottom to top. The second pulse area is fixed in the simulations to  $\theta_2 = \pi$ . The pulse duration  $\tau$  and the long time dephasing rate  $\beta$  are fitted to the smallest pulse area, and we found the best agreement for  $\tau = 12$  ps and  $\beta = 0.01/\text{ps}$ . For the simulations, we choose the same QD geometry as for the calculations shown in Fig. 2.

For the smallest considered pulse area, the measured FWM signal in Fig. 4(a) just forms one maximum around  $\tau_{12} = 25$  ps and decays for longer delays single exponentially with  $\beta$ . These dynamics are well reproduced by the simulation in Fig. 4(b). Additionally, there is hardly any difference between the calculation with and without exciton-phonon interaction



**Fig. 4.** Normalized two-pulse FWM with  $\tau \simeq 12$  ps pulses in a micropillar cavity. (a), (b) FWM amplitude as a function of the delay  $\tau_{12}$  for increasing pulse area of the first pulse from bottom to top. (a) Experiment,  $P_1 = (0.02, 0.24, 0.35, 0.55) \mu\text{W}$ ,  $P_2 = 0.08 \mu\text{W}$ ; (b) theory, with  $\theta_1$  as given in the plot and  $\theta_2 = \pi$ ; solid/dotted lines with/without coupling to phonons; (c), (d) FWM spectral amplitudes as a function of excitation power for  $\tau_{12} = 0$  illustrating emergence of the AT splitting with increasing  $\theta_1$ . Experiment in (c) against  $\sqrt{P_1}$  and theory in (d) against  $\theta_1$ .

(solid and dashed lines). This indicates that the optically driven dynamics are too slow to induce the PID drop of the signal. In other words, the polaron creation follows the exciton occupation adiabatically, and no phonon wave packet is emitted [45]. We can instructively describe the optical excitation process using the laser-exciton dressed state picture [61]. These states then have an energy splitting  $\Delta_{\text{AT}}$ , the so called Autler–Townes (AT) splitting [62], which is proportional to the instantaneous laser field amplitude. While in the exciton basis the LA phonons only couple to the exciton state, in the presence of a light field they lead to transitions between the dressed states. For small pulse areas, i.e.,  $\theta_1 \ll \pi$ ,  $\Delta_{\text{AT}}$  is in the  $\mu\text{eV}$  range, hence in the energy range, where the phonon spectral density is negligible [29]. Therefore, the phonons do not lead to efficient transitions between the dressed states, which allows them to evolve adiabatically.

The appearance of a splitting between the dressed states with increasing field strength can be seen in Figs. 4(c) and 4(d). There, we show the FWM spectrum centered around the exciton transition energy  $E_x$ . In the measurement in Fig. 4(c), it is plotted against  $\sqrt{P_1} \sim \theta_1$ , which is proportional to the field amplitude. The corresponding simulation is presented in Fig. 4(d) and shows a good agreement with the measurement. The spectra are taken for  $\tau_{12} = 0$ . While for small pulse areas the spectrum is dominated by a single line at the exciton energy, it splits according to the AT splitting of the dressed states, reaching values of approximately 0.1 meV in Fig. 4(c). Note that the AT splitting is time-dependent for pulsed excitations. Therefore, we are seeing a time-integrated version of the splitting between the dressed states in the FWM spectra. However,  $\Delta_{\text{AT}}$  is the direct spectral domain translation of the Rabi oscillations in the FWM signal. In agreement between the dynamics depicted in Figs. 4(a) and 4(b) and the spectral behavior in Figs. 4(c) and 4(d), we find clear signatures of the Rabi oscillations from pulse areas of  $\theta_1 \approx 1.5\pi$  onward. We additionally find that the line at positive detuning is stronger than the other one. This stems from a slight mismatch between the energies of the driving laser pulses and the exciton transition of  $\hbar\omega_L - E_x = -0.05$  meV, which was considered in the simulation in Fig. 4(d). This detuning agrees well with the separation between the exciton and cavity line in Fig. 3(b) at 27 K. Only for the case of exact resonance between the driving field and the transition energy are the dressed states equally occupied [61]. As we find here, already detunings in the few  $\mu\text{eV}$  range result in a significant mismatch of the two lines in the FWM signal. To check that the AT splitting is driven by the laser pulses, we performed the same measurement and simulation as in Figs. 4(c) and 4(d) but with a large delay of  $\tau_{12} = 30$  ps to reduce the overlap of the pulses. Here, no AT splitting was found (see Supplement 1).

Comparing the second pulse area in Fig. 4(a) (green) to the yellow curve, the maximum of the signal splits into two local maxima, with one moving to smaller and one moving to larger delays  $\tau_{12}$ . The maxima are marked by the circles. In the simulation in Fig. 4(b), we find the best agreement for  $\theta_1 = 1.4\pi$  (solid green line), where we already find clear signatures of the Rabi oscillations during the interaction with the first laser pulse. Here two distinct maxima of almost equal height are formed. Compared to the simulation without the exciton-phonon interaction (dashed green line), we find a significant deviation for the second maximum already for this pulse area. Without the coupling to phonons, the FWM signal drops very rapidly, and

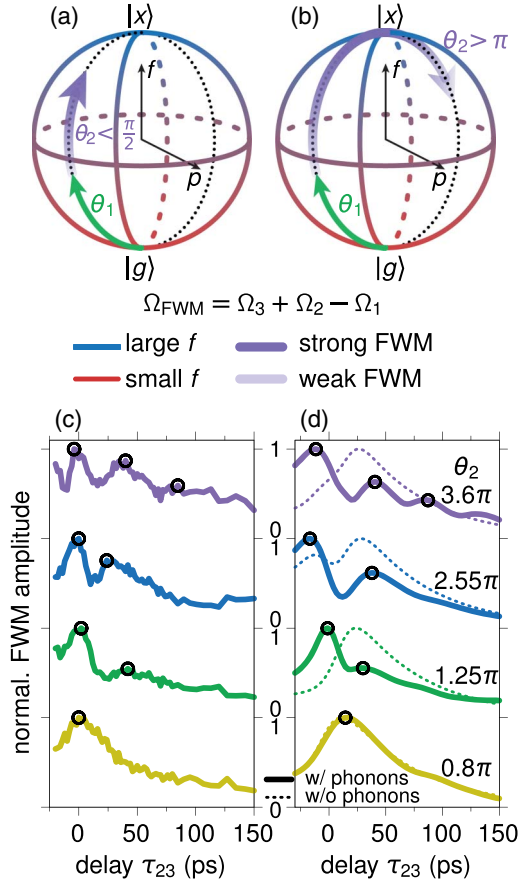
the second maximum is much smaller than the first one. There are two main effects enhancing the discrepancy between the two calculations: (i) For these pulse areas we have shown that  $\Delta_{\text{AT}}$  attains the 0.1 meV range. This allows for more efficient phonon-assisted transitions between the dressed states and therefore to strong dephasing during the first laser pulse. (ii) The exciton-phonon interaction leads to a renormalization of the pulse areas [26,63–65]. This makes a direct comparison of the pulse areas with and without phonon coupling difficult.

Stepping to the next larger pulse area in Fig. 4(a), i.e., from the green to the blue curve, does not alter the dynamics of the FWM signal significantly. We basically find a slightly larger difference in the height of the two maxima in the signal. This is reproduced by the modeled signal in Fig. 4(b). Here the minimum between the two maxima is more pronounced, as in the experiment, which we already found in a similar way for the shorter pulses in Fig. 2. The deviation between the simulation with and without phonon coupling remains remarkable because of the reasons pointed out earlier.

The most significant difference for the signal dynamics is found for the largest considered pulse area in the experiment, which is shown as a violet curve in Fig. 4(a). It forms a double peak structure within the first 25 ps followed by a minor maximum around  $\tau_{12} = 50$  ps. After that, the signal is basically null. The very same behavior is found in the simulation with  $\theta_1 = 2.0\pi$  in Fig. 4(b): Two strong and narrow peaks around  $\tau_{12} = 0$  ps are followed by a small maximum at  $\tau_{12} \approx 50$  ps. Together with the stunning agreement with the simulated curve in Fig. 4(b) (solid violet), this impressively shows that multiple Rabi oscillations are resolved in the coherence dynamics of the two-pulse FWM experiment. Without considering the coupling to phonons, the model gives the dashed line in Fig. 4(b). Here also three maxima build up, but the relative heights of the second and third maximum do not agree with the measured curve in Fig. 4(a) at all. This shows that coupling to phonons has a strong impact on the optically driven dynamics of the exciton quantum state.

The FWM technique allows us to use different pulse sequences that lead to different microscopic quantities determining the FWM signal. So far, we have studied the exciton polarization  $p$  by employing a two-pulse FWM experiment with  $\varphi_{\text{FWM}} = 2\varphi_2 - \varphi_1$ . We now turn to a three-pulse excitation with  $\varphi_{\text{FWM}} = \varphi_3 + \varphi_2 - \varphi_1$  where—in a two-level system—the FWM signal carries information about the occupation of the exciton state  $f$ . A schematic picture of the Bloch sphere for this experiment is shown in Figs. 5(a) and 5(b) [see also Fig. 1(c)]. The first pulse drives a microscopic coherence. The second pulse converts this coherence into an occupation of the exciton, which is then turned into the FWM signal by the third pulse. A larger occupation of the exciton state  $f$  will accordingly lead to a stronger FWM amplitude. The idea is to fix the first and third pulse areas to small values  $\theta_1, \theta_3 \lesssim \pi/2$  and change the second area  $\theta_2$ . This will then result in different occupations  $f$  and therefore different signal strengths. For large pulse areas  $\theta_2$ , the three-pulse FWM signal will resolve the optically driven Rabi oscillations projected on the exciton occupation  $f$ .

Figures 5(c) and 5(d) present the normalized experimental and theoretical results, respectively, for different pulse areas  $\theta_2$ , increasing from bottom to top. The FWM amplitude is plotted as a function of the delay  $\tau_{23}$  to investigate the exciton population



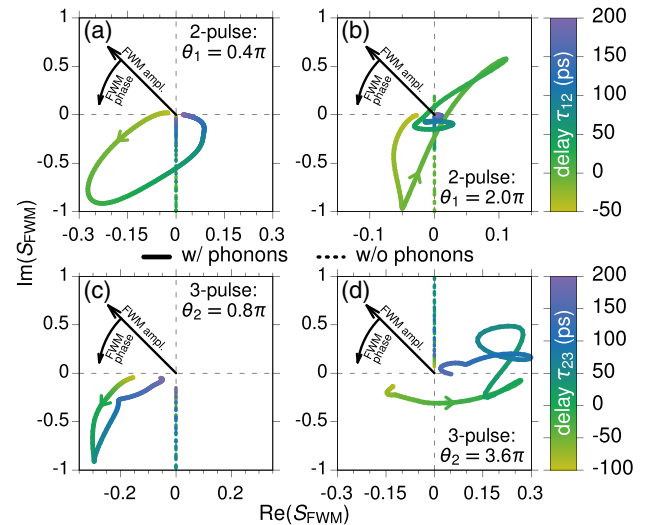
**Fig. 5.** Normalized three-pulse FWM with  $\tau \approx 12$  ps pulses in a micropillar cavity. (a), (b) Schematic pictures of the FWM signal on the Bloch sphere. (a) For small pulse areas  $\theta_2$ ; (b) for pulse areas  $\theta_2$  exceeding  $\pi$ ; solid/dotted lines with/without coupling to phonons; (c), (d) FWM amplitude as function of the delay  $\tau_{23}$  for increasing pulse area of the second pulse from bottom to top. The FWM delay dependence probes the optically driven evolution of the exciton occupation, performing Rabi oscillations for excitations with high pulse areas. (c) Experiment,  $T = 27$  K,  $\tau_{12} = 10$  ps,  $P_1 = 0.05 \mu\text{W}$ ,  $P_3 = 0.1 \mu\text{W}$ ,  $P_2 = (0.13, 0.3, 0.56, 1) \mu\text{W}$ ; (d) theory, with  $\theta_1 = 0.2\pi$ ,  $\theta_3 = 0.4\pi$  and  $\theta_2$  as given in the plot.

dynamics. The first delay is fixed to  $\tau_{12} = 10$  ps to enable the buildup of the intracavity field. We see for the smallest pulse area at the bottom (yellow) that the dynamics of the three-pulse FWM amplitude forms a single broad maximum. For long delays, the signal is dominated by the spontaneous decay. The decay rate was chosen to  $\gamma = 0.004/\text{ps}$  to give the best fit to the measurement. We found the best agreement between measurement and simulation for  $\theta_2 = 0.8\pi$ . The other pulse areas are chosen to be  $\theta_1 = 0.2\pi$  and  $\theta_3 = 0.4\pi$  in all simulations.

Comparing experiment and theory, we find an excellent agreement for each considered  $\theta_2$ . With increasing pulse strength, the first maximum shifts to shorter delays  $\tau_{23}$ , while at longer  $\tau_{23}$ , additional maxima emerge, as marked by the circles. These dynamics now happen during the interaction with the second laser pulse. Therefore, the signal is dominated by the Rabi oscillations of the exciton occupation  $f$ . For the two largest pulse areas (blue and violet), up to three distinct maxima can be found in the signal. When comparing the simulations including the exciton-phonon coupling (solid) with those omitting it (dashed), we find

clear qualitative differences. In the  $\theta_2 = 1.2\pi$  case (green), the maxima have a significant mismatch in their delay  $\tau_{23}$  of approximately 25 ps. Another striking example is the largest pulse area at the top, where the full model, including the phonon coupling, develops multiple modulations, while the uncoupled calculation yields a single broad peak. One obvious reason for this stronger discrepancy between the model with and without phonons in Fig. 5(b) compared to the results in Fig. 4(b) is the fact that the former one includes three pulses, while the latter one only two. Therefore, the impact of dephasing and pulse area renormalization come into play for one more pulse, further enhancing their influence. Additionally, we reach higher pulse areas, which results in general in a more efficient exciton-phonon interaction.

The FWM signal  $S_{\text{FWM}}$  is a complex quantity with real and imaginary part or amplitude and phase. This feature has, e.g., been used to distinguish between different coupling situations in few-level systems [35]. We here use it to further illustrate the dynamics of the Rabi oscillations and especially the influence of the exciton-phonon coupling. In Fig. 6 we plot real and imaginary parts of the simulated FWM signals and color-code the delay to visualize the dynamics of the signals. The arrow heads indicate the direction of the time evolution. In principle, Figs. 6(a), 6(b) and 6(c), 6(d) are more sophisticated representations of the data shown in Fig. 4(b) and 5(d), respectively. In Figs. 6(a) and 6(b), we show the smallest and largest pulse areas for the two-pulse FWM case and in Figs. 6(c) and 6(d) the smallest and largest pulse areas for the three-pulse FWM case. The remaining four considered pulse areas are given in Supplement 1. The simulations without exciton-phonon coupling are shown as dotted lines. These curves are restricted to the vertical axis, i.e., the FWM signal is purely imaginary. When the full model is considered (solid lines), real and imaginary parts of the FWM signals get mixed, and the curves in Fig. 6 show very involved dynamics. While for the small pulse area examples in Figs. 6(a) and 6(c) the signals are governed by a single loop, in the high area cases in Figs. 6(b)



**Fig. 6.** Entire complex FWM signal. Real and imaginary part of the FWM signal  $S_{\text{FWM}}$ . The delays are color-coded; the dotted lines show simulations without phonon coupling, and the solid lines with phonon coupling. (a), (b) For two-pulse FWM. The corresponding dynamics of the FWM amplitude are given in Fig. 4(b). (c), (d) For three-pulse FWM. The corresponding dynamics of the FWM amplitude is given in Fig. 5(d).

and 6(d) the signals perform multiple loops. These loops represent the Rabi oscillations, which we also found in the dynamics of the FWM amplitudes in Figs. 4 and 5. This way of analyzing the FWM signals exploits the full potential of the method in an appealing fashion.

#### 4. CONCLUSIONS

In summary, we have investigated the optically driven dynamics of the full quantum state of a single QD exciton in the presence of efficient coupling to acoustic phonons. By driving the system with sub- and superps laser pulses and applying two- and three-pulse FWM techniques, we could isolate the dynamics of the microscopic polarization and the exciton occupation, respectively. By this, we showed that for large pulse areas, involved dynamics of the Bloch vector in the form of Rabi oscillations take place. Especially, the coupling to LA phonons plays a decisive role for the optical control of the QD exciton: On the one hand, for short laser pulses ( $\tau < 1$  ps) phonons lead to the loss of coherence after the first laser pulse, due to the emission of phonon wave packets. On the other hand, for  $\tau \approx 10$  ps the dephasing happens already during the first pulse, and it therefore strongly depends on the applied pulse area. Only the accurate theoretical description of the exciton-phonon interaction allowed for a quantitative analysis and comprehension of the measured data. In the spectral domain, the observed Rabi oscillations represent the AT splitting, which we clearly observed in the FWM spectra.

We thus find that FWM is a versatile technique to study optically driven dynamics in many different aspects, not only in single QDs, but also in any other potential isolated quantum systems such as spins of QD trions [1–3] or single defect centers in insulators [4,5]. However, the time scales of spins are by a factor of 10–100 slower than our optically active exciton transition, which renders the investigation in the optical range far more challenging.

We have demonstrated that FWM spectroscopy of single emitters can be used to study the optically driven dynamics of the quantum state and at the same time works in a regime where the coupling to phonons is rather strong. This makes this technique also promising to investigate optical transitions in localized excitons in atomically thin structures such as transition metal dichalcogenides and hexagonal boron nitride or color centers in insulators. These systems stand out due to their functionality at elevated temperatures, making them an up-and-coming platform for quantum applications.

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See Supplement 1 for supporting content.

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