

Increase of intensity-dependent excitonic second- and third-harmonic generation induced by static electric fields demonstrated for the K exciton of a $2H$ homobilayer of MoS_2

Ruixin Zuo ¹, Matthias Reichelt ¹, Cong Ngo ¹, Xiaohong Song ², Weifeng Yang,^{2,3} and Torsten Meier ^{1,*}

¹Department of Physics, [Paderborn University](#), Warburger Strasse 100, D-33098 Paderborn, Germany

²School of Physics and Optoelectronic Engineering, [Hainan University](#), Haikou 570288, China

³Center for Theoretical Physics, [Hainan University](#), Haikou 570288, China



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We compute and analyze the dependence of excitonic second- and third-harmonic generation (SHG/THG) as a function of the optical excitation intensity in the presence of static electric fields by solving the semiconductor Bloch equations. Our simulations are performed for excitation of the strongly bound intralayer exciton of an inversion-symmetric homobilayer of MoS_2 with in-plane electric fields. We demonstrate that for resonant excitation at the $1s$ K -exciton the SHG and the THG show complex dependencies on both the strength of the static field and the peak amplitude of the optical pulse. For sufficiently intense optical excitation, the THG increases and the SHG increases superlinearly with the amplitude of the static field as long as exciton ionization is not yet dominating. Microscopic simulations demonstrate that these dependencies arise from an interplay between several effects including static and transient Stark shifts, exciton ionization, off-resonant Rabi oscillations, and a modified interference between optical nonlinearities induced by the intraband acceleration. Our findings offer several new possibilities for controlling the strong-field dynamics of systems with strongly bound excitons.

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I. INTRODUCTION

As is well known, homogeneous static electric fields are able to drastically change the electronic properties of crystals [1–7]. Modifications of the electron wave functions and the electronic dynamics by electrical biasing can be detected using suitable frequency- and time-domain optical techniques that excite and/or probe interband transitions. Indeed, since the late 1950s a number of intriguing effects induced by static electric fields have been observed in semiconductors and semiconductor nanostructures like quantum wells and superlattices; see, e.g., Refs. [8–18]. For example, field-induced tunneling of the electron wave function into the band gap and the accompanying oscillatory behavior above the band gap, known as the Franz-Keldysh (FK) effect [5–7], is visible in optical absorption spectra as finite absorption below the band-gap and above-band-gap oscillations [8–12]. Furthermore, electric fields localize the electronic wave functions, which are extended Bloch functions in the absence of a bias, in the direction in which they are applied. This Wannier-Stark (WS) localization [3,4] is visible in optical spectra when the field strength is sufficiently strong such that the separation between WS ladder states exceeds the optical linewidth and transitions to individual WS states can be resolved [13,14].

To be able to observe the time-domain counterpart of WS localization, i.e., Bloch oscillations (BO) [1,2], the period for one BO must be shorter than the dephasing times. Since the dielectric breakdown limits the amplitudes of static electric fields that can maximally be applied without destroying the crystal, WS localization [13,14] and BOs [15,16] were first detected in semiconductor superlattices which are artificial crystals with a unit cell that is much larger than lattice constants of natural crystals. As the splitting between WS states is proportional to the length of the unit cell and the BO period is inversely proportional to the length of the unit cell, the requirements to observe these phenomena could be reached in semiconductor superlattices for field strengths well below the dielectric breakdown fields.

When semiconductors are excited off-resonantly far below the band gap with ultrashort light pulses, the peak electric field amplitudes (typically several megavolts per centimeter up to some 10 MV/cm) can significantly exceed the breakdown fields without destroying the material. This regime is nowadays routinely reached in many strong-field experiments and is, e.g., utilized for the generation of high harmonics from semiconductors [19–31]. In such experiments the fields are often so strong that transient BOs, i.e., a time-dependent motion of the wave vector in field direction through the entire Brillouin zone during one cycle of the exciting optical field, take place during the excitation [20]. Thereby such highly intense pulses strongly modify the electronic properties on ultrashort timescales and can, e.g., realize transient WS localization which leads to distinct modifications of optical spectra [32].

Due to the Coulomb attraction between photoexcited electrons and holes, excitonic resonances, representing bound

*Contact author: torsten.meier@uni-paderborn.de

electron-hole states with typically hydrogen-like wave functions, appear as discrete absorption lines below the band gap [33,34]. The binding energy of excitons in semiconductors and semiconductor nanostructures depends strongly on the material and, in particular, on the dimensionality and varies from a few millielectronvolts up to about 100 meV. The wave function of the lowest $1s$ exciton is typically, at least to very good approximation, symmetric in space and therefore a static electric field will typically result in a quadratic (static or dc) Stark shift to lower energies [35–37]. Since an electric field corresponds to a potential that varies linearly in space and thus tilts the attractive Coulomb potential, it also leads to a finite probability that electrons and holes separate from each other, i.e., that the exciton ionizes. The reduced lifetime of the exciton due to ionization can be observed as a field-dependent broadening of the exciton peak in optical spectra [36,38] or as a more rapid decay in time-domain spectroscopy [18]. The situation changes for sufficiently strong static fields that lead to WS localization. In this regime the states in the field direction are localized and the Coulomb attraction couples the electron and hole WS states to form WS excitons [39,40].

For sufficiently intense resonant excitation, Rabi flopping of semiconductor excitons can be achieved [41,42]. Excitonic Rabi flopping differs distinctively from what one would expect for a simple two-level system and is significantly influenced by many-body correlations. When excitonic systems are excited off-resonantly below the resonance by intense fields off-resonant Rabi oscillations, i.e., an incomplete oscillation of the occupation and the optical polarization, can be induced [43,44]. They can be viewed as a kind of partial Rabi oscillations where no complete occupation of the upper state is realized, however, the polarization still changes sign during the dynamics. Furthermore, when semiconductors and semiconductor nanostructures are excited weakly detuned below the resonance, excitons shift towards higher energies due to the optical Stark effect [45–47]. The strength of this dynamical or ac Stark shift is proportional to the field intensity and inversely proportional to the detuning. In particular, when pulses with different polarizations are used, the excitonic Stark effect was shown to be strongly influenced by exciton-exciton interactions, i.e., many-body correlations [48]. Furthermore, also the excitonic dynamical FK effect has been reported in semiconductor quantum wells where a frequency- and intensity-dependent shift of the excitonic resonance under intense THz irradiation has been observed [49].

The family of two-dimensional (2D) transition-metal dichalcogenides (TMD) semiconductors shows numerous fascinating physical properties resulting from their reduced dimensionality and crystal symmetry, which are distinct from their bulk counterparts [50–71]. Due to the quantum confinement and decreased dielectric screening of the Coulomb interaction, excitons in 2D TMD are observable even at room temperature and dominate the optical properties. Two-dimensional TMD host exceptionally high exciton binding energies of several 100 meV and hence are ideally suited for studying exciton physics.

Due to the large exciton binding energies and the associated small excitonic Bohr radii, the amplitudes of static fields that induce significant changes in the excitonic properties

are one to two orders of magnitude higher than those for ordinary semiconductors and semiconductor nanostructures. Sufficiently strong field amplitudes lead to measurable excitonic Stark shifts which arise from the coupling to other exciton states and exciton ionization [72–75]. Recently, excitonic second-harmonic generation (SHG) has been studied in a WSe₂ monolayer beyond the perturbative regime and the results were interpreted in terms of intensity-dependent optical Stark and Bloch-Siegert shifts [76].

The in-plane breakdown fields of TMDs are typically on the order of megavolts per centimeter. As we show below, for such field amplitudes transitions to WS states appear in the linear absorption spectra. The very high absorption of TMD excitons leads also to sizable optical Stark shifts and off-resonant Rabi oscillations for excitation with far off-resonant optical fields with peak amplitudes in the range of a few megavolts per centimeter. Thus the strong-field physics of excitons in the presence of optical and static fields is influenced by an interesting interplay of several phenomena. As several times higher field amplitudes than the breakdown field can be used for pulsed excitation it is possible to investigate in TMDs ultrafast dynamical excitonic processes in highly nonperturbative regimes which are not reachable in most other systems.

In this paper, we present and discuss the results of microscopic simulations for resonant excitonic SHG and excitonic third-harmonic generation (THG). As a model system we investigate the strongly bound intralayer excitons of an inversion-symmetric homobilayer of MoS₂. We consider in-plane polarized optical pulses which are two- or three-photon resonant with the $1s$ K exciton and study the dependencies of SHG and THG on the excitation intensity and on the amplitude of a homogeneous static in-plane electric field. Our results are obtained by numerical solutions of the semiconductor Bloch equations (SBE) including the many-body Coulomb interaction in time-dependent Hartree-Fock approximation and inter- and intraband excitations in the length gauge [33,77,78]. The band structure and matrix elements are computed by density-functional theory [79].

Since the homobilayer of MoS₂ is inversion symmetric, SHG is only possible when the symmetry is broken which can be achieved in several different ways. For example one can use static electric fields [80,81] which is known as electric-field-induced second-harmonic generation, an effect that has been widely used in different settings and on various material systems, see, e.g., Ref. [82]. For static fields applied perpendicular to the layers remarkable switching and tunability of SHG from $2H$ stacked MoS₂ homobilayer was demonstrated [56] and the role of interlayer excitons mediating SHG has been investigated [66].

Here we consider optical and static fields which are collinearly polarized in the plane of the layers and show that the dependence of the excitonic SHG on the amplitude of the static field changes qualitatively with the intensity of the optical field. In particular, we find a regime in which the SHG increases superlinearly with the amplitude of the static field. Our microscopic simulations clarify that a combination of several effects including static and transient Stark shifts, exciton ionization, and off-resonant Rabi oscillations contribute to the obtained complex field dependencies. Also

the computed excitonic THG shows a very complex dependence on the amplitudes of the static and the optical fields. For moderate optical excitation intensity it exhibits an unusual increase as function of the amplitude of the static field when exciton ionization is not yet dominant. Besides the effects mentioned above, for the static-field-enhanced THG additionally higher-order optical excitations representing exciton-to-continuum-to-exciton transitions are relevant. Our analysis shows that the intraband acceleration modifies the interference between optical nonlinearities, leading to a more constructive interference in the presence of a static field.

The paper is structured as follows. Section II presents the modeling of the system and the theoretical description of the dynamics including the light-matter and the many-body Coulomb interactions. Some additional details are provided in the appendix. In Sec. III we present and discuss excitonic optical absorption spectra in bilayer MoS₂ with in-plane electric fields and the dependence of the excitonic SHG and THG on the strengths of the static and optical fields. To explain these findings we analyze how the intraband acceleration induced by static and oscillating fields changes the excitonic absorption. Furthermore, we investigate the influence of static and transient Stark shifts and of off-resonant Rabi oscillations on the dynamics. In Sec. IV we briefly summarize our main findings.

II. THEORETICAL MODEL

Our model system is a 2H stacked homobilayer of MoS₂ which consists of two van der Waals bonded monolayers which are rotated by 180° with respect to each other. In contrast to the nonequivalence of the two valleys *K* and *K'* including a finite splitting of bands which results from the lifted spin degeneracy present in a monolayer, the bands of the 2H homobilayer are spin-degenerate throughout the Brillouin zone (BZ) and have equivalent *K* valleys. It was shown that homobilayer TMDs can be used to tune the valley circular dichroism in a controllable way, e.g., by applying an interlayer bias [52,83–86]. Here we concentrate on intralayer excitonic effects in homobilayer MoS₂ excited and biased by in-plane polarized electric fields, see Fig. 1(a), and compute the dynamical optical response by numerically solving the SBE.

We start from a many-body Hamiltonian that contains three terms [33,77,78],

$$H = H_0 + H_{\text{LM}} + H_C. \quad (1)$$

The electronic band structure is described by

$$H_0 = \sum_{\lambda k} \epsilon_{\lambda k} a_{\lambda k}^\dagger a_{\lambda k}, \quad (2)$$

where λ combines the spin and band indices and $a_{\lambda k}^\dagger$ ($a_{\lambda k}$) is the creation (annihilation) operator of an electron with crystal momentum $\hbar k$ in band λ and $\epsilon_{\lambda k}$ is the band structure. The eigenfunctions of the crystal Hamiltonian are Bloch functions $\Psi_{\lambda k}(\mathbf{r}) = e^{i\mathbf{k}\cdot\mathbf{r}} u_{\lambda k}(\mathbf{r})$. Using the length gauge, the light-matter interaction is described in the dipole approximation by [78,87–89]

$$H_{\text{LM}} = -e\mathbf{E}(t) \cdot \sum_{\lambda\lambda'kk'} \mathbf{r}_{kk'}^{\lambda\lambda'} a_{\lambda k}^\dagger a_{\lambda'k'}. \quad (3)$$

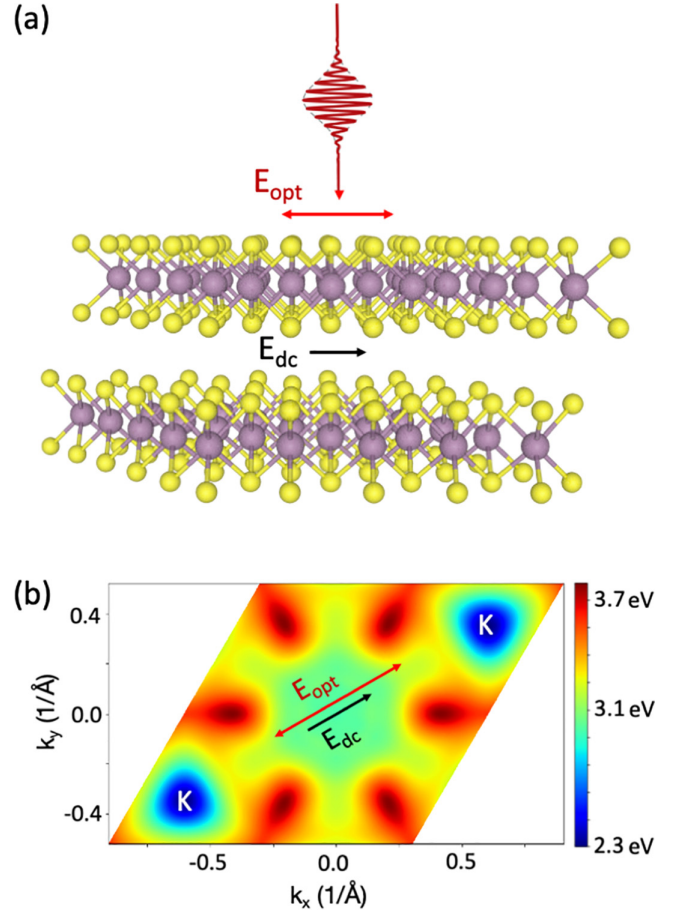


FIG. 1. (a) Illustration of the 2H homobilayer of MoS₂ and the considered electric fields. A linearly polarized optical pulse excites the material under normal incidence. The optical field and the static field are collinearly polarized in the plane of the bilayer system. (b) The interband transition energy in the 2D Brillouin zone. The arrows show the polarization directions of the optical and the static fields which coincide with the zigzag direction for all simulations described below.

Here $\mathbf{E}(t)$ is the electric field and

$$e\mathbf{r}_{kk'}^{\lambda\lambda'} = [\mathbf{d}_k^{\lambda\lambda'} + ie\delta_{\lambda\lambda'} \nabla_k] \delta(\mathbf{k} - \mathbf{k}') \quad (4)$$

are the matrix elements of the position operator multiplied by the electron charge e , where $\mathbf{d}_k^{\lambda\lambda'} = ie \int d^3r u_{\lambda k}^*(\mathbf{r}) \nabla_k u_{\lambda'k'}(\mathbf{r})$ denotes for $\lambda \neq \lambda'$ the interband dipole matrix elements and for $\lambda = \lambda'$ the Berry connection $\mathbf{d}_k^{\lambda\lambda}/e$ and ∇_k describes the intraband acceleration by the electric field. The many-body Coulomb interaction between photoexcited carriers reads

$$H_C = \frac{1}{2} \sum_{\substack{\lambda\lambda'kk'q \\ q \neq 0}} V_{q,k,k'}^{\lambda\lambda'} a_{\lambda k}^\dagger a_{\lambda'k'}^\dagger a_{\lambda'k'+q} a_{\lambda k-q} \quad (5)$$

with the quasi-2D Coulomb matrix element

$$V_{q,k,k'}^{\lambda\lambda'} = \int d^3r \int d^3r' u_{\lambda k}^*(\mathbf{r}) u_{\lambda'k'}^*(\mathbf{r}') \times W_q(z, z') u_{\lambda'k'+q}(\mathbf{r}') u_{\lambda k-q}(\mathbf{r}), \quad (6)$$

where $W_q(z, z')$ is the Coulomb potential including dielectric screening that is described by a dielectric function which is derived from Poisson's equation [90]; see Appendix A for more details. The Coulomb interaction leads to renormalizations of the single-particle band structure and the Rabi frequency. The latter results in the formation of bound electron-hole pairs, i.e., excitons, which strongly modify the optical properties.

The dynamics of the photoexcited carriers is analyzed using the Heisenberg equations of motion for the microscopic interband polarizations $p_k^{\lambda\lambda'} = \langle a_{\lambda'k}^\dagger a_{\lambda k} \rangle$ and the carrier occupations $n_k^\lambda = \langle a_{\lambda k}^\dagger a_{\lambda k} \rangle$. The resulting coupled equations of motion are known as the SBE which in time-dependent Hartree-Fock approximation read [33,77,78]

$$\frac{\partial}{\partial t} p_k^{\lambda\lambda'} = -\frac{i}{\hbar}(\varepsilon_k^\lambda - \varepsilon_k^{\lambda'}) p_k^{\lambda\lambda'} + \frac{e}{\hbar} \mathbf{E}(t) \cdot \nabla_k p_k^{\lambda\lambda'} + \frac{i}{\hbar} \Omega_k^{\lambda\lambda'} (n_k^{\lambda'} - n_k^\lambda) - \frac{p_k^{\lambda\lambda'}}{T_2}, \quad (7a)$$

$$\frac{\partial}{\partial t} n_k^\lambda = \frac{2}{\hbar} \text{Im} \left[\sum_{\lambda' \neq \lambda} \Omega_k^{\lambda\lambda'} p_k^{\lambda\lambda'} \right] + \frac{e}{\hbar} \mathbf{E}(t) \cdot \nabla_k n_k^\lambda, \quad (7b)$$

where

$$\Omega_k^{\lambda\lambda'} = \mathbf{d}_k^{\lambda\lambda'} \cdot \mathbf{E}(t) + \sum_{k'} V_{k-k',k,k'}^{\lambda\lambda'} p_{k'}^{\lambda\lambda'} \quad (8)$$

is the renormalized Rabi frequency,

$$\varepsilon_k^\lambda = \epsilon_k^\lambda - \sum_{k'} V_{k-k',k,k'}^{\lambda\lambda} n_{k'}^\lambda, \quad (9)$$

is the renormalized energy, and T_2 is the dephasing time of the microscopic polarizations.

The light field emitted by a coherently excited semiconductor contains two sources and is described by $E_{\text{out}}(t) \propto \frac{\partial^2}{\partial t^2} \mathbf{P}(t) + \frac{\partial}{\partial t} \mathbf{J}(t)$ [77,91] with the macroscopic polarization $\mathbf{P}(t)$ originating from interband transitions

$$\mathbf{P}(t) = \sum_{\substack{\lambda\lambda' \\ \lambda \neq \lambda'}} \int_{\text{BZ}} d\mathbf{k} \mathbf{d}_k^{\lambda\lambda'} p_k^{\lambda\lambda'} \quad (10)$$

and the macroscopic charge current $\mathbf{J}(t)$ due to intraband currents,

$$\mathbf{J}(t) = e \sum_{\lambda} \int_{\text{BZ}} d\mathbf{k} \mathbf{v}_k^\lambda n_k^\lambda, \quad (11)$$

where the integration runs over the first BZ and $\mathbf{v}_k^\lambda = \nabla_k \varepsilon_k^\lambda / \hbar$ is the group velocity in band λ .

We perform density functional calculations to obtain the ground state of the 2H homobilayer of MoS₂ using the ELK code [79] with the generalized gradient approximations. The band gap is adjusted using the scissors operator. The multiband twisted parallel transport gauge [78,87,92,93] is implemented on the eigenfunctions resulting in matrix elements that vary smoothly as function of wave vector. For the homobilayer MoS₂ system, the spin-degenerate bands are formed by orbitals from different layers. Due to interlayer hopping which hybridizes the valence band states of different layers, the splitting of bands occurs with the increase of the

number of layers. Particularly, the valence band splitting at the Γ point is much larger than at the K point for bilayer MoS₂ and therefore the band gap is indirect [94]. However, the lowest direct interband transition energy still occurs at the K point with a band gap of $E_g = 2.336$ eV. According to the optical selection rules [94], in each K valley, the top valence band couples to the lowest conduction band via an intralayer transition and couples to another energetically close conduction band via an interlayer transition, the latter being rather weak. In the present work, we consider optical intralayer transitions via a four-band model (two degenerate conduction and two degenerate valence bands around the band gap). The interband transition energy is shown in Fig. 1(b) and the peak strength of the interband dipole matrix elements for the two allowed transitions of opposite spins is $0.153e$ nm. In Appendix B, we rewrite the SBE for one spin, i.e., a two-band model (the transition for the other spin has the same magnitude under in-plane polarized exciting fields) and also present a perturbative expansion of the SBE which is used for analyzing the dynamics for not too strong optical excitation.

In our numerical evaluations, we consider optical and static electric fields which are collinearly polarized in the plane of the homobilayer, i.e., the total electric field is given by $\mathbf{E}(t) = E(t)\mathbf{e} = (E_{\text{opt}}(t) + E_{\text{dc}})\mathbf{e}$, where the unit vector $\mathbf{e}$ defines the polarization direction. Note that below we denote the peak amplitude of the optical field by E_{opt} . Throughout this paper the fields are polarized in the zigzag direction. We discuss the components of the interband polarization and the intraband current in this direction which allows us to ignore their vector characters.

Dephasing is modeled phenomenologically via a dephasing time of $T_2 = 50$ fs, which is a reasonable value considering the measured linewidth [95] and the dephasing time obtained for a MoSe₂ bilayer from four-wave-mixing experiments [67]. The optical pulses used in the numerical calculations have a Gaussian envelope with a full width at half maximum of 100 fs, which ensures that the computed signals are predominantly generated by the coherent dynamics but still provide a sufficient spectral selectivity.

III. RESULTS AND DISCUSSION

To determine the linear absorption, we solve Eq. (7a) for constant ground-state occupations using a weak ultrashort in-plane-polarized optical pulse. The spectral absorption is proportional to the imaginary part of the ratio between the Fourier-transformed macroscopic polarization and the optical electric field [33], i.e.,

$$\alpha(\omega) \propto \omega \text{Im}[P(\omega)/E_{\text{opt}}(\omega)], \quad (12)$$

where the pulse spectrum $E_{\text{opt}}(\omega)$ is broad and covers the excitation of exciton and continuum states. Figure 2(a) displays the linear absorption spectra of a homobilayer MoS₂ for different static electric fields E_{dc} which induce a time-dependent change of the wave vectors according to the acceleration theorem. Without a static field and when the Coulomb interaction is neglected, the absorption starts in a steplike fashion which reflects the 2D density of states at the band gap; see upper blue line in Fig. 2(a). Due to the electron-hole attraction the absorption spectra are dominated by strongly absorbing excitonic

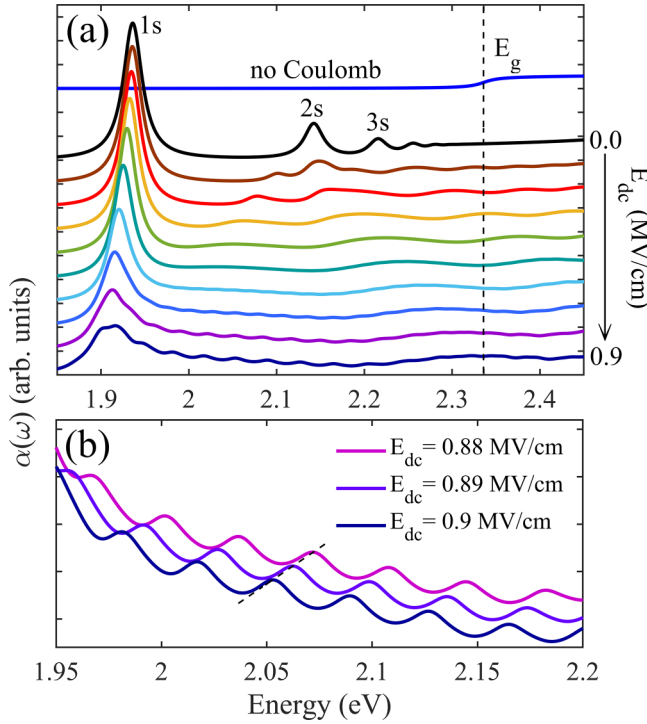


FIG. 2. Calculated linear absorption spectra for a freestanding $2H$ homobilayer MoS_2 . (a) Without electron-hole attraction and without a static field the comparatively weak absorption starts in a steplike fashion, which reflects the 2D density of states, at the band gap; see upper blue line. In the presence of the Coulomb attraction the absorption spectra are dominated by strongly absorbing excitonic resonances below the band gap. With increasing static field amplitude E_{dc} all excitons Stark-shift to lower energies. Simultaneously, the peak absorption is reduced and the absorption lines broaden due to exciton ionization. For $E_{dc} \gtrsim 0.7$ MV/cm weak periodic oscillations appear which originate from WS localization. The curves are shifted vertically for clarity. (b) When the static field is strong enough such that the splitting between adjacent WS states exceeds the homogeneous linewidth, optical transitions to the ladder of WS states lead to oscillatory features in the absorption spectra. As shown by the dashed line, the transition energy to a particular WS state shifts proportional to E_{dc} .

resonances below the band gap. Without an electrical bias, the linear absorption spectrum provides the energetic positions of the three lowest s excitons which resemble a modified hydrogenic series. The position $E_{1s} = 1.936$ eV and the binding energy of about 400 meV of the $1s$ exciton are in reasonable agreement with reported experimental and theoretical data for layered TMDs [96–98].

A static in-plane electric field breaks the spatial symmetry and induces a coupling among excitonic states of opposite symmetry, e.g., s and p excitons. The newly formed exciton states can be described as superpositions of the field-free exciton states. The energy of the lowest exciton shifts red with increasing field showing a quadratic dc Stark shift [75] which we denote by δ_{dc} . Apart from the hybridization, a static field tilts the attractive Coulomb potential in real space which results in a finite probability that the electron and hole separate, i.e., that the exciton ionizes, which reduces the exciton lifetime with increasing field amplitude [74,99]. The

dc Stark shift δ_{dc} and the broadening due to field-induced exciton ionization are clearly visible in Fig. 2(a). Due to their weaker binding energy which corresponds to a larger extension in real space, the higher excitons $3s$ and $2s$ are ionized already for significantly weaker fields than the $1s$ exciton.

For $E_{dc} \gtrsim 0.7$ MV/cm weak periodic oscillations appear near and above the $1s$ exciton in Fig. 2(a). The spacing between adjacent peaks scales linearly with E_{dc} and, as shown in Fig. 2(b), the position of an individual peak shifts proportional to E_{dc} . This behavior is exactly what is expected when WS localization is realized. Without Coulomb interaction the splitting between adjacent WS states in 2D lattices is given by $eE_{dc}a^*$, where a^* is the relevant effective lattice constant which depends on the direction in which the static field is applied [100,101]. For $E_{dc} = 0.7$ MV/cm, the splitting $eE_{dc}a^*$, with $a^* = 1.5a$ and the lattice constant $a = 3.18$ Å for a field applied in the zigzag direction [101], is about 33 meV and thus slightly exceeds the homogeneous linewidth which is already increased somewhat above its field-free values of $2\hbar/T_2 = 26$ meV due to exciton ionization. For semiconductor superlattices it has been shown that excitonic effects lead to a reduced WS splitting near the exciton resonance and correspondingly to an increased Bloch period, which is the time-domain counterpart of the WS splitting; see Refs. [39,40,102–104]. This reduced splitting is also present in Fig. 2. For example, the excitonic WS splitting seen for $E_{dc} = 0.9$ MV/cm is ≈ 36 meV, whereas without Coulomb interaction ≈ 43 meV would be expected. Unlike in semiconductor superlattices, due to the much larger exciton binding energy in the MoS_2 bilayer transitions to several WS states can be resolved above the $1s$ exciton. We compute SHG and THG by solving the SBE, Eqs. (7a) and (7b), for excitation pulses which are two- or three-photon resonant with the field-free $1s$ exciton, i.e., the photon energies are $\hbar\omega_0 = 1.936$ eV/2 or $\hbar\omega_0 = 1.936$ eV/3, respectively, for various static and optical field amplitudes. The resulting SHG and THG shown in Figs. 3 and 4 have been obtained by integrating $|P(\omega)|$ around $2\omega_0$ or $3\omega_0$ and dividing the result by E_{opt}^2 or E_{opt}^3 , respectively, i.e., the dependence on the peak amplitude of the optical field in the perturbative limit. For far off-resonant excitation the interplay between inter- and intraband transitions significantly influences the nonlinear optical response [105,106]. For the excitation conditions we consider here, the excited occupations are rather low and the resonantly excited excitonic polarization dominates. Therefore, the generated radiation is predominantly originating from the interband polarization, whereas the contributions from the intraband currents are negligible. We verified that the resulting signal is reduced by a few orders of magnitude when the term $\mathbf{E}_{opt}(t) \cdot \nabla_{\mathbf{k}} p(\mathbf{k})$ in the SBE is artificially switched off. The dominant pathways to SHG and THG are thus that in first order in the electric field a linear polarization $(p_k^{\lambda\lambda'})_{inter}^{(1)}$ is generated by an interband transition (note that as we start from the ground state, i.e., a completely filled valence band and an empty conduction band, no intraband excitation is possible in first order in the field). As outlined above, higher-order excitations occur predominantly via subsequent intraband transitions. Therefore, the second-order and third-order polarization $(p_k^{\lambda\lambda'})_{inter,intra}^{(2)}$ and $(p_k^{\lambda\lambda'})_{inter,intra,intra}^{(3)}$, which dominate the resonantly excited excitonic SHG and THG for not too strong optical fields,

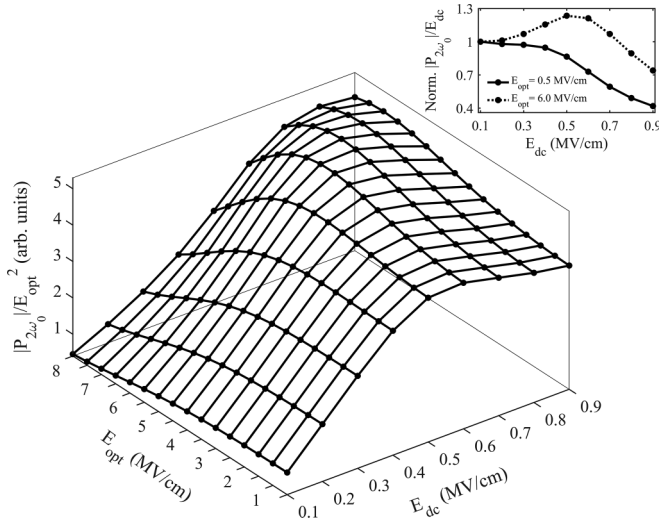


FIG. 3. Ratio between $|P_{2\omega_0}|$ and E_{opt}^2 with $|P_{2\omega_0}| = \int_{1.7\omega_0}^{2.3\omega_0} |P(\omega)| d\omega$ for various optical and static field amplitudes. The excitation frequency is $\hbar\omega_0 = 1.936$ eV/2. Inset: Normalized $|P_{2\omega_0}|/E_{dc}$ as a function of the static field amplitude for $E_{opt} = 0.5$ MV/cm (solid dotted line) and $E_{opt} = 6.0$ MV/cm (dashed dotted line). The symbols represent the calculated values and the lines are guides for the eye.

respectively, are generated from $(p_k^{\lambda\lambda'})_{inter}^{(1)}$ via subsequent intraband transitions.

For a centrosymmetric system excited by a single-color pulse the generation of even harmonics is forbidden. Applying a static electric field breaks the symmetry and enables SHG which in the perturbative limit scales linearly with the field amplitude E_{dc} . As expected, for weak optical driving, i.e.,

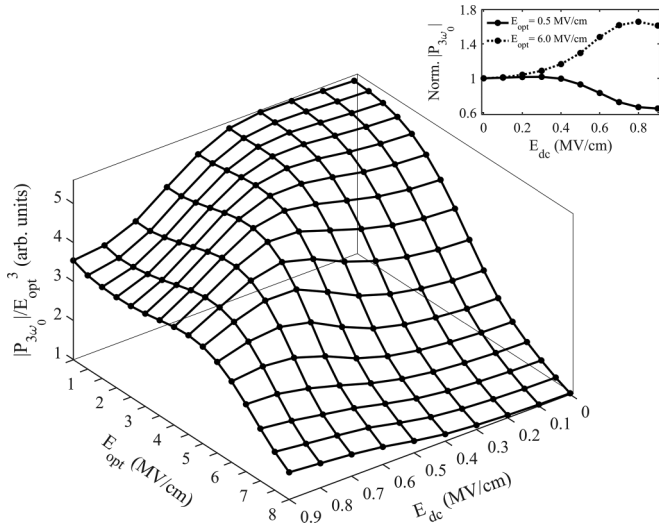


FIG. 4. Ratio between $|P_{3\omega_0}|$ and E_{opt}^3 with $|P_{3\omega_0}| = \int_{2.7\omega_0}^{3.3\omega_0} |P(\omega)| d\omega$ for various optical and static field amplitudes. The excitation frequency is $\hbar\omega_0 = 1.936$ eV/3. Inset: Normalized $|P_{3\omega_0}|/E_{dc}$ as a function of static field amplitude for $E_{opt} = 0.5$ MV/cm (solid dotted line) and $E_{opt} = 6.0$ MV/cm (dashed dotted line). The symbols represent the calculated values and the lines are guides for the eye.

small E_{opt} , the SHG shown in Fig. 3 starts to increase linearly with E_{dc} . Starting at about $E_{dc} \approx 0.5$ MV/cm the increase gets weaker and turns into a decrease for $E_{dc} \geq 0.7$ MV/cm. This dependence of the SHG on E_{dc} for weak E_{opt} originates from a combination of two effects which are visible in Fig. 2. With increasing E_{dc} the $1s$ exciton experiences a quadratic Stark shift δ_{dc} to lower energies such that the SHG is not excited fully resonantly. However, even if one would adjust for this Stark shift by tuning the excitation frequency accordingly, then the SHG would still decrease for larger E_{dc} since exciton ionization leads to a significant increase of the homogeneous linewidth, i.e., rapid dephasing.

For stronger optical excitation, i.e., larger E_{opt} , the SHG shows an unexpected dependence on E_{dc} . Above around $E_{opt} = 4$ –5 MV/cm the optical driving leaves the perturbative limit, as can be seen by the reduction of the line for $E_{dc} = 0.1$ MV/cm for larger E_{opt} in Fig. 3. At the same time, the SHG starts to show a clear superlinear, approximately quadratic increase as function of E_{dc} , see also the inset of Fig. 3, up to about $E_{dc} = 0.5$ –0.6 MV/cm. Furthermore, with increasing E_{opt} the maximal SHG is reached for larger E_{dc} .

We now turn to the dependence of the THG on E_{dc} and E_{opt} shown in Fig. 4. For weak optical driving the THG is unaffected by small bias fields, but it starts to decrease as function of E_{dc} when the Stark shift δ_{dc} and exciton ionization become relevant. For larger E_{opt} , when the optical excitation is beyond the perturbative limit, the THG shows an unexpected significant increase as function of E_{dc} (note that we plot the amplitude of the polarization, i.e., the intensity is enhanced up to about a factor of three). As clearly shown in the inset of Fig. 4 for $E_{opt} = 6.0$ MV/cm the maximal THG is generated for dc field amplitudes at which for weaker optical driving exciton ionization already significantly reduces the THG. As will be analyzed below, static (δ_{dc}) and dynamic Stark shifts [$\delta_{opt}(t)$], off-resonant Rabi oscillations, dc-field-induced exciton ionization and a partial stabilization in strong optical fields, as well as a modified interference between higher-order optical nonlinearities are relevant in the investigated regime and contribute to the dependencies of the SHG and THG shown in Figs. 3 and 4.

To gain further insight into the change of the excitonic properties under the action of an oscillating field, we calculate the linear optical absorption in the presence of a constantly oscillating field, i.e., a continuous wave (cw) field, which is considered only in the intraband acceleration of Eq. (7a). The linear absorption spectra for frequencies corresponding to the SHG and THG calculations and different field amplitudes are shown in Figs. 5(a) and 5(b). We clearly see that the peak of the $1s$ exciton shifts towards higher energies with increasing amplitude of the cw field, i.e., an optical Stark shift. This shift can be understood to arise from the intraband acceleration of electrons and holes induced by the optical cw field. The averaging over the periodic dynamic field-induced change of the interband transition energy shifts the effective gap of the interband absorption to higher energies. The effective gap increases by the ponderomotive energy $U_p \propto E_{cw}^2/\omega_{cw}^2$ [107].

The $1s$ exciton peaks seen in Figs. 5(a) and 5(b) basically follow the shift of the band gap to higher energies which is quadratic in E_{cw} and due to the smaller ω_{cw} larger for the THG than for the SHG excitation conditions. Unlike the

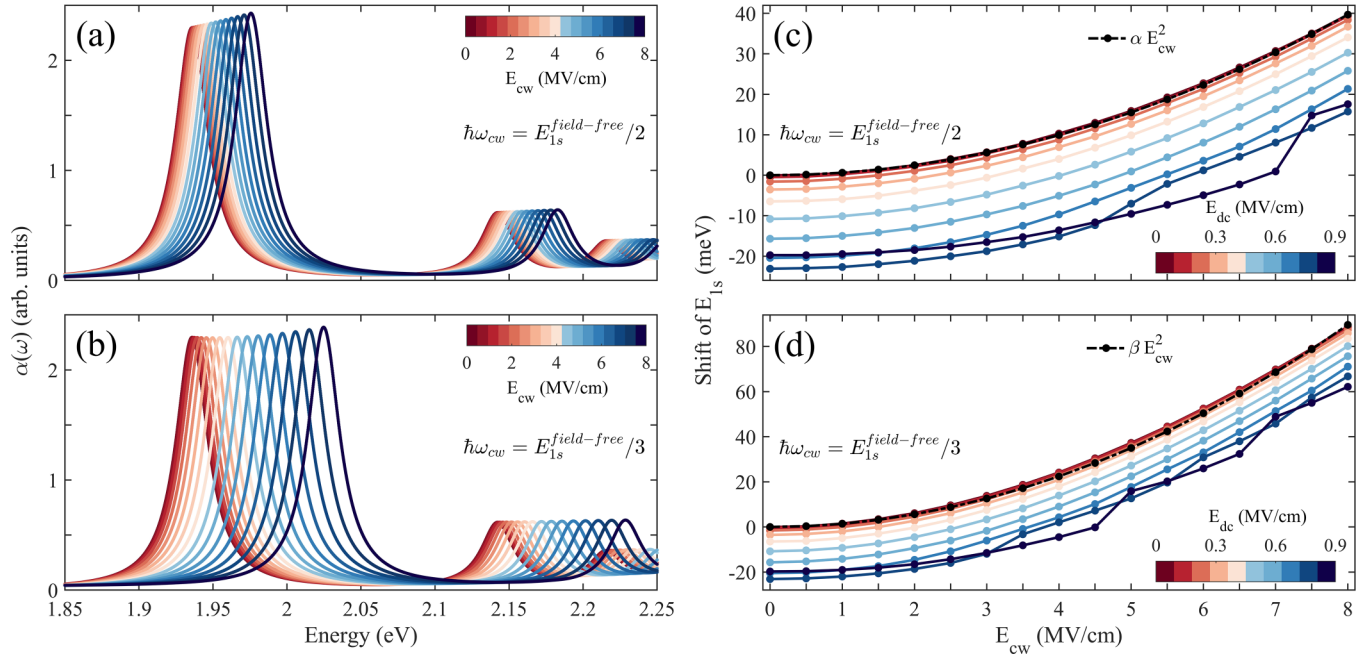


FIG. 5. Linear absorption spectra in the presence of a cw optical field with (a) $\hbar\omega_{cw} = 1.936$ eV/2 and (b) $\hbar\omega_{cw} = 1.936$ eV/3. [(c) and (d)] The energetic shift of the main exciton resonance compared to the field-free position of the $1s$ exciton due to both static and cw optical fields for the cw fields used in (a) and (b), respectively. The oscillatory results for $E_{dc} \geq 0.7$ MV/cm arise from WS localization. In this regime the largest excitonic WS peak shows an oscillatory dependence on E_{dc} .

suppression of the absorption peak and the spectral broadening in the presence of a static field, the shape of the absorption peak in Fig. 5(a) is nearly unchanged by the cw field. If we carefully determine the exciton binding energy by taking the difference of the exciton peak to the effective band gap in the presence of the cw field, then we find a small increase as function of E_{cw} . This can be understood to arise from the band width which is reduced in the polarization direction of the cw field due to the averaging of the transition energy over the oscillations of the wave vector induced by the optical field. As a result the effective mass near the band gap increases, i.e., a stronger anisotropy of the band structure and thus of the exciton evolves. This effect can also be viewed as the onset of dynamical localization [108,109] which was shown to lead to an increased anisotropy or even a change of the effective dimensionality of excitons [110].

The energy shifts of the main absorption peaks with respect to the field-free $1s$ exciton in the presence of both static and cw fields are shown in Figs. 5(c) and 5(d). As can be seen in Figs. 5(c) and 5(d) for a vanishing dc field, the optical Stark shifts of the $1s$ exciton can be fitted well by parabolic functions (black dash curves) of the field amplitude with $\alpha = 0.062e$ nm²/V for $\hbar\omega_{cw} = 1.936$ eV/2 and $\beta = 0.14e$ nm²/V for $\hbar\omega_{cw} = 1.936$ eV/3. In the presence of both cw and static fields, we obtain a competition between the dc and the optical Stark shifts which have opposite signs. Therefore the red shift induced by a static field can be compensated or overcompensated by a cw field. For the field amplitudes considered here, the blue shift induced by the cw field dominates for high E_{cw} , as exciton ionization limits the achievable dc Stark shifts and WS localization, which leads to oscillatory deviations from the quadratic dependence on E_{cw}

for $E_{dc} \geq 0.7$ MV/cm, modifies the nature of the excitons for strong E_{dc} .

Next we analyze the time-dependent frequencies of the coherent SHG and THG polarizations. For this analysis we filter out frequencies in the range of $1.5\omega_0 < \omega < 2.5\omega_0$ for SHG and $2.5\omega_0 < \omega < 3.5\omega_0$ for THG to obtain $\bar{P}(t)$. Then we transform $\bar{P}(t)$ into a rotating frame in order to concentrate on the slowly varying dynamics via $\tilde{P}(t) = \bar{P}(t)e^{-2i\omega_0 t}$ for SHG and $\tilde{P}(t) = \bar{P}(t)e^{-3i\omega_0 t}$ for THG, respectively. In these frames, subtle changes of the time-dependent phases of the SHG and THG polarizations can be determined via $\phi(t) = \text{Im}[\log(\tilde{P}(t))]$. If the dynamics is dominated by a single frequency which changes slowly as function of time, then the shift of the instantaneous frequency with respect to the field-free $1s$ exciton resonance is given by $\omega_{\text{shift}}(t) = \frac{\partial}{\partial t}\phi(t)$. As shown below, by this procedure we obtain important and detailed information on the static and transient optical Stark shifts of the $1s$ exciton resonance, i.e., δ_{dc} and $\delta_{\text{opt}}(t)$, and further phenomena.

Figures 6 and 7 show $\hbar\omega_{\text{shift}}(t)$ for the SHG and THG results presented in Figs. 3 and 4, respectively. Please note that in Appendix C the main features visible in Figs. 6 and 7 are transparently analyzed and explained on the basis of a simple linear p equation with static and time-dependent detunings. Figure 6(a) demonstrates that the SHG starts to oscillate with twice the excitation frequency which is identical to the frequency of the field-free $1s$ exciton, i.e., $\hbar\omega_{\text{shift}} = 0$. For weak optical fields, the optical Stark shift leads to a transient increase of $\hbar\omega_{\text{shift}}$ to positive values during the pulsed excitation. With increasing time the instantaneous frequency continuously reduces towards lower frequencies and reaches the position of the dc Stark

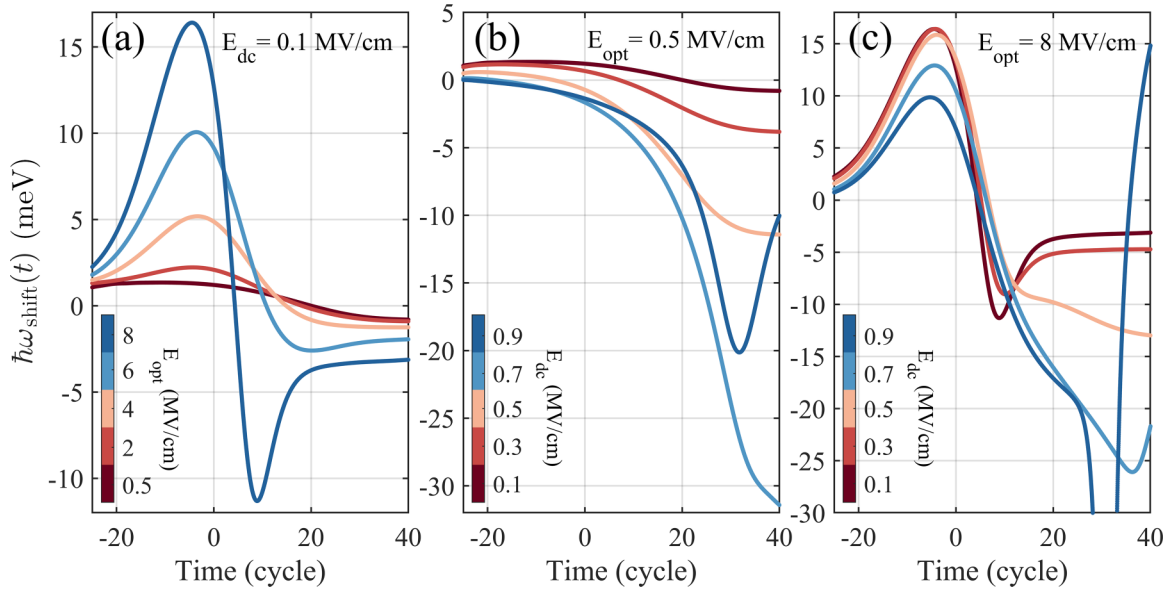


FIG. 6. $\hbar\omega_{\text{shift}}(t)$ of the SHG frequency for (a) varying the optical field amplitude with $E_{\text{dc}} = 0.1$ MV/cm, (b) varying the static field amplitude with $E_{\text{opt}} = 0.5$ MV/cm, and (c) the same as in (b) but for $E_{\text{opt}} = 8.0$ MV/cm. The excitation parameters are the same as those used in Fig. 3. The duration of an optical cycle is 4.27 fs.

red-shifted exciton after the excitation pulse. For stronger optical excitation the final value of $\hbar\omega_{\text{shift}}$ in Fig. 6(a) is reduced below the position of the red-shifted exciton due to band-gap renormalization. The temporal change from twice the driving frequency at early times to the dc-Stark shifted eigenfrequency of the exciton at later times is more clearly visible in Fig. 6(b) for E_{dc} up to about 0.5 MV/cm. Significant sharp oscillatory changes of $\hbar\omega_{\text{shift}}(t)$ appear in Fig. 6(a) for stronger optical driving starting at about $E_{\text{opt}} \geq 6$ MV/cm. This dynamics originates from off-resonantly driven Rabi oscillations. As can be seen in Fig. 3 for $E_{\text{dc}} = 0.1$ MV/cm,

with increasing excitation intensity the SHG starts to deviate from the perturbative quadratic scaling with E_{opt} which, as shown in Fig. 6(a), coincides with the start of an incomplete off-resonant Rabi cycle.

Considering the exciton as a two-level system with transition energy $E_{1s}(t)$ which depends on time due to the transient optical Stark shift and dipole matrix element d , the n -photon Rabi energy can be written as [111,112]

$$\hbar\Omega_R^{(n)}(t) = \sqrt{\left(\frac{(dE(t))^n}{E_{1s}(t)^{n-1}}\right)^2 + (\hbar\delta_n(t))^2} \quad (13)$$

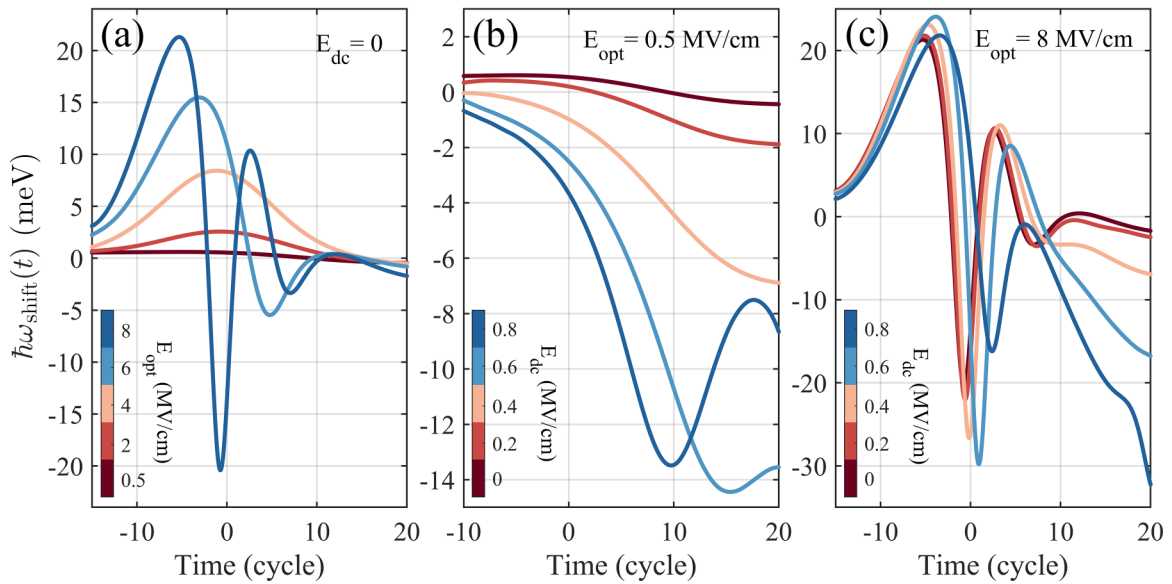


FIG. 7. $\hbar\omega_{\text{shift}}(t)$ of the THG frequency for (a) varying the optical field amplitude with $E_{\text{dc}} = 0.0$ MV/cm, (b) varying the static field amplitude with $E_{\text{opt}} = 0.5$ MV/cm, and (c) the same as in (b) but for $E_{\text{opt}} = 8.0$ MV/cm. The excitation parameters are the same as those used in Fig. 4. The duration of an optical cycle is 6.4 fs.

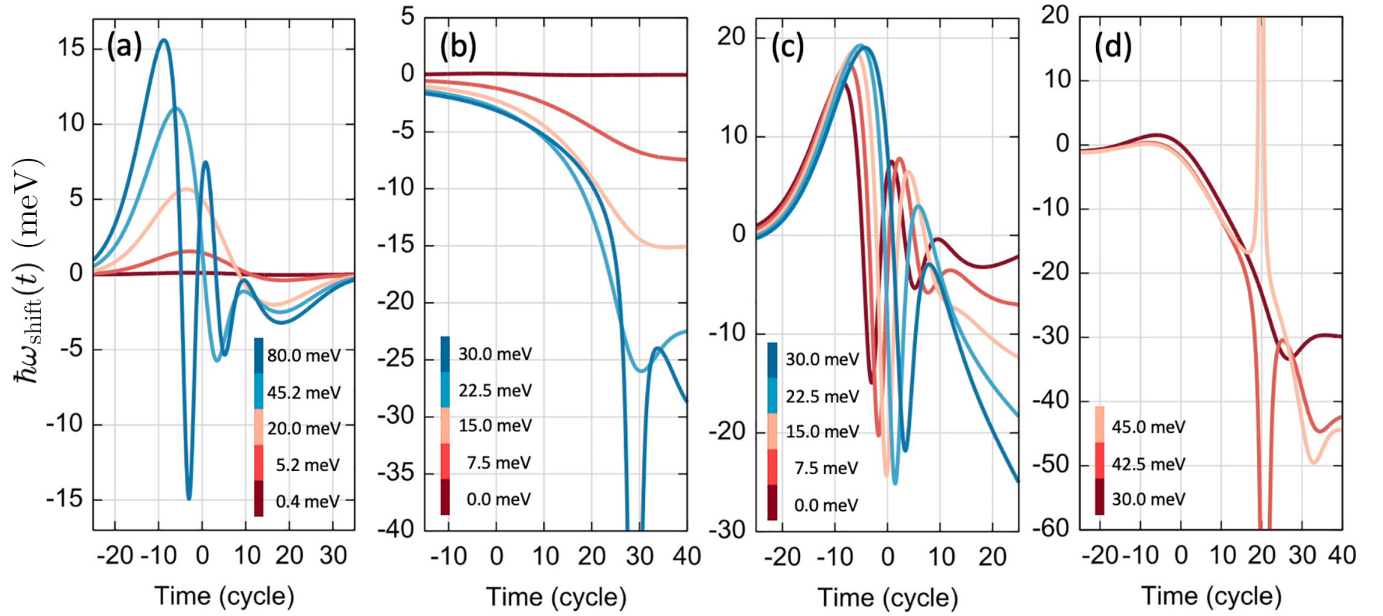


FIG. 8. Results for the instantaneous frequency shifts obtained by numerically solving Eq. (C1). The detunings δ_{dc} and $\delta_{\text{opt}}(t)$ mimic the effects of the static and optical fields that underlie the shifts shown in Fig. 7(a)–7(c). (a) No static field, i.e., $\delta_{\text{dc}} = 0$, and increasing optical fields amplitudes with maximal shifts $\delta_{\text{opt}}(t = 0) = 0.4$ –80.0 meV. (b) a weak optical field leading to $\delta_{\text{opt}}(t = 0) = 0.4$ meV and increasing static fields inducing shifts of $\delta_{\text{dc}} = 0$ –30.0 meV (c) a strong optical field leading to $\delta_{\text{opt}}(t = 0) = 80.0$ meV and the same increasing static fields inducing shifts of $\delta_{\text{dc}} = 0$ –30.0 meV. Panel (d) shows that divergences might occur for certain parameters as shown in Fig. 6(c). An explanation is provided in the text.

with the transient n -photon detuning $\hbar\delta_n(t) = E_{1s}(t) - n\hbar\omega_0$. For the conditions considered in this analysis, the light-matter coupling $d E(t)$ is much smaller than the transition energy and thus we can approximate

$$\Omega_R^{(n)}(t) \approx |\delta_n(t)|, \quad (14)$$

i.e., the effective Rabi frequency for the n -photon excitations is determined by the time-dependent detuning. The dynamics one obtains from a simple linear p equation with static and time-dependent detuning is analyzed in Appendix C using parameters which closely match the situation of the THG calculations. The results shown in Fig. 8 very well reproduce the features that appear in Fig. 7 and also almost all effects visible in Fig. 6. Only the shift to lower energies visible in Fig. 6(a) for strong optical excitation and long times which originates from band-gap renormalization is not included in the simple model that is evaluated in Appendix C.

Apart from the absence of a dc Stark shift, very similar dynamics is demonstrated in Fig. 7(a) also for the THG polarization. The deviation of the THG in Fig. 4 from the perturbative scaling with E_{opt} is exactly correlated with the behavior of $\hbar\omega_{\text{shift}}(t)$ in Fig. 7(a) where again off-resonant Rabi oscillations appear for sufficiently strong optical driving, see Appendix C. Due to the larger detuning of the optical pulses the number of Rabi cycles is larger for THG than for SHG.

When the optical field is relatively weak, e.g., $E_{\text{opt}} = 0.5$ MV/cm shown in Figs. 6(b) and 7(b), the detuning of the exciton with respect to $2\hbar\omega_0$ or $3\hbar\omega_0$, respectively, is given by the dc Stark shift δ_{dc} . Therefore, for not too strong dc fields $\hbar\omega_{\text{shift}}(t)$ starts at zero, i.e., initially follows the driving, and then continuously shifts red towards δ_{dc} when the optical pulse

is gone. The oscillations that appear in Figs. 6(b) and 7(b) for higher static fields are due to off-resonant Rabi oscillations that can become visible when the detuning is sufficiently large. For $E_{\text{dc}} = 0.9$ MV/cm WS localization leads to a splitting or weak oscillation of the exciton absorption, see Fig. 2, which also contributes to the phase dynamics.

For stronger optical fields, e.g., $E_{\text{opt}} = 8$ MV/cm shown in Figs. 6(c) and 7(c), the dynamics is dominated by off-resonant Rabi oscillations during the excitation, see Appendix C. The rapid changes seen in Figs. 6(c) for the largest dc field near 30 cycles originate from a destructive interference which leads to a vanishing polarization in the time domain and thus an ill-defined phase, as is explained in Appendix C and reproduced in Fig. 8(d).

As shown above, for the considered excitation conditions several effects contribute to the excitonic SHG and THG. Further insight into the nonlinear dynamics can be obtained by comparing the results obtained by full nonperturbative solutions of the SBE, i.e., Eqs. (7a) and (7b), with the perturbative contributions which have been computed using the expansion with respect to the optical field described in Appendix B. Figures 9(a) and 9(b) show the lowest-order perturbative contributions to SHG and THG, i.e., the response up to second- and third-order in the optical field, respectively. The SHG initially rises linearly and then decreases, whereas the THG is initially constant and then decreases as function of E_{dc} . For weak optical fields this corresponds to the expected dependencies and the decrease for stronger E_{dc} originates, as explained above, from the dc Stark shift which detunes the exciton and exciton ionization. The SHG and THG obtained by solving the full SBE are for small E_{dc} below the lowest-order perturbative results, but for larger E_{dc} above the lowest order

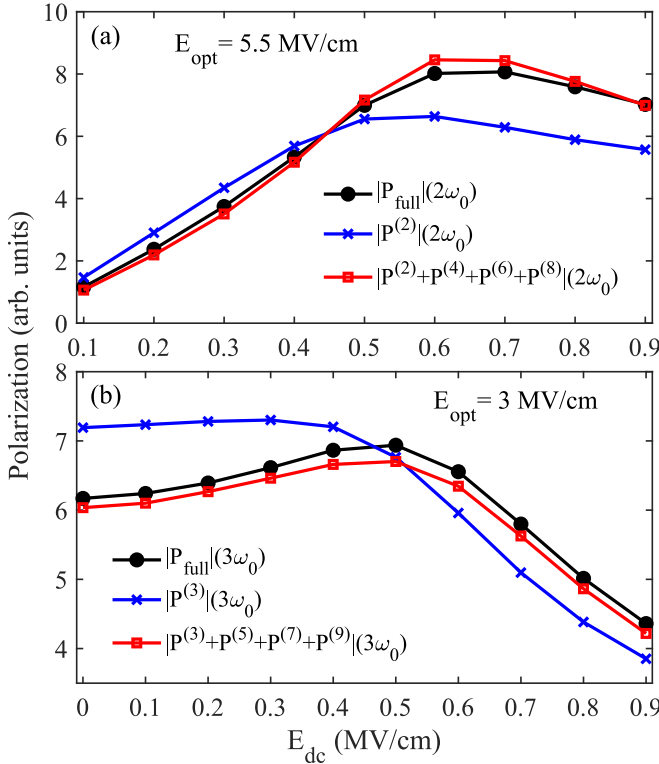


FIG. 9. (a) SHG computed as in Fig. 3 by integrating $P(\omega)$ around $2\omega_0$ as function of the amplitude of the static field E_{dc} for $E_{opt} = 5.5$ MV/cm. (b) THG computed as in Fig. 4 by integrating $P(\omega)$ around $3\omega_0$ as function of the amplitude of the static field E_{dc} for $E_{opt} = 3$ MV/cm. For both cases we show the nonperturbative results (black), the lowest-order perturbative contributions with respect to the optical field, i.e., up to second and third order, (blue), as well as the perturbative expansion with respect to the optical field including the four lowest contributing orders, i.e., up to eighth and ninth order, (red), respectively.

perturbative results. Figures 9(a) and 9(b) thus demonstrate that the quite unexpected results shown in Figs. 3 and 4, i.e., the superlinear increase of SHG with E_{dc} and the increase of the THG with E_{dc} , clearly originate from higher-order nonlinearities. Thus, whereas for a small bias the lowest and higher-order interfere destructively, for both SHG and THG the interference becomes constructive for $E_{dc} \geq 0.5$ MV/cm. The good agreement between the full results and the perturbative ones which include the four lowest contributing orders demonstrates furthermore that for the considered peak amplitudes of the optical field E_{opt} we are still in the perturbative limit. This is in fact not the case for more intense optical fields; however, also in this regime the full results remain above the lowest-order perturbative contributions. It should be noted that in Figs. 9(a) and 9(b) the decrease as function of the electrical bias starts at larger E_{dc} for the full result than for the lowest-order perturbative contributions. Besides a transient partial restoration of the resonant excitation of the exciton, when the blue shift $\delta_{opt}(t)$ partly cancels the dc red shift δ_{dc} , also a stabilization of the exciton against ionization in a strong optical field, i.e., a partial dynamic suppression of tunneling [113–115], contributes to this behavior.

IV. CONCLUSIONS

In summary, we have numerically solved the SBE including inter- and intraband excitations and the many-body Coulomb interaction to compute resonantly excited excitonic SHG and THG in the presence of in-plane electric fields. As a model system we investigated the strongly bound intralayer $1s$ K-exciton of an inversion-symmetric $2H$ stacked homobilayer of MoS_2 . It should be noted, that additional calculations for a generic two-band tight-binding model lead to qualitatively similar results which confirms that the obtained dependencies do not depend on details of the model system but are of general nature.

For weak optical excitation the dependence of the SHG and the THG on the amplitude of a static in-plane field shows an expected behavior. Although the SHG increases linearly with the field amplitude, the THG is constant for weak static fields and both decrease for stronger electrical biasing. The decrease originates from the quadratic dc Stark shift of the exciton to lower energies and from increased dephasing caused by exciton ionization. Very unexpected dependencies on the amplitude of the static field are demonstrated for stronger optical excitation. For elevated amplitudes of the optical field, we identified a regime in which the THG increases and the SHG increases superlinearly as function of the strength of the static field. Our detailed analysis demonstrates that several effects contribute to the computed signals. The static field and the optical pulses shift the exciton resonance in opposite directions. Thus the optical excitation can transiently reduce, transiently compensate or even overcompensate the red shift induced by the dc field. For stronger static fields, the exciton line broadens which reflects increased dephasing due to exciton ionization which reduces the nonlinear optical response. Additionally, for sufficiently strong static fields weak signatures of transitions to excitonic WS states appear in the linear absorption spectra. The static and transient shifts of the exciton line, as well as off-resonant Rabi oscillations which are visible in the dynamics of the instantaneous frequency of the SHG and THG polarizations have been explained in terms of a simple linear equation with static and time-dependent detunings of the transition frequency. Furthermore, we demonstrated that the static-field-induced intraband acceleration modifies the interference between optical nonlinearities.

The understanding gained by our detailed analysis can be used to design new avenues for controlling the strong-field dynamics of systems with strongly bound excitons in the time and frequency domains. By using pulsed Terahertz radiation instead of static fields additional temporal control and higher field strengths that may significantly exceed the breakdown fields and thus allow one to explore a broader range of strong-field phenomena, like dynamic WS localization [32] and others, might be achievable.

ACKNOWLEDGMENTS

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DATA AVAILABILITY

The data that support the findings of this article are openly available [116].

APPENDIX A: CALCULATION OF COULOMB POTENTIAL

To describe intralayer excitons in a freestanding bilayer MoS₂, we employ the dielectric model for a multilayer structure [90] and compute the quasi-2D intralayer Coulomb potential of a point charge located at the center of the layer. For a geometry of thickness $L = 2D$ (D is the layer-to-layer distance in the natural bulk configuration) with dielectric parameters:

$$\epsilon_{\parallel}(z) = \begin{cases} 1 & z < -\frac{L}{2} \\ \epsilon_{\parallel} & |z| < \frac{L}{2} \\ 1 & z > \frac{L}{2} \end{cases}, \quad \epsilon_{\perp}(z) = \begin{cases} 1 & z < -\frac{L}{2} \\ \epsilon_{\perp} & |z| < \frac{L}{2} \\ 1 & z > \frac{L}{2} \end{cases}. \quad (\text{A1})$$

The dielectric constant is 1 for the vacuum outside the system and $\epsilon_{\parallel}$ and $\epsilon_{\perp}$ are the background dielectric constants. The bulk dielectric constants $\epsilon_{\perp}$, $\epsilon_{\parallel}^B$ can be obtained from density functional calculations and $\epsilon_{\parallel}$ is determined via $\epsilon_{\parallel}^B = \epsilon_{\parallel} + \lim_{q \rightarrow 0} 4\pi e^2 \chi_L(\mathbf{q}, \omega)/D$, where $\chi_L(\mathbf{q}, \omega)$ is the longitudinal susceptibility.

The intralayer Coulomb potential is given by

$$W_q\left(z = \frac{D}{2}, z' = \frac{D}{2}\right) = \frac{2\pi}{\kappa q} \left(1 + c_1 e^{-\sqrt{\frac{\epsilon_{\parallel}}{\epsilon_{\perp}}} q_{\parallel} D} + c_1 e^{-\sqrt{\frac{\epsilon_{\parallel}}{\epsilon_{\perp}}} q_{\parallel} 3D} + 2c_2 e^{-\sqrt{\frac{\epsilon_{\parallel}}{\epsilon_{\perp}}} q_{\parallel} 2L}\right), \quad (\text{A2})$$

with

$$\begin{aligned} \kappa &= \sqrt{\epsilon_{\parallel}\epsilon_{\perp}}, \\ c_1 &= \frac{(\kappa + 1)(\kappa - 1)}{\mathcal{N}}, \\ c_2 &= \frac{(\kappa - 1)(\kappa - 1)}{\mathcal{N}}, \\ \mathcal{N} &= (\kappa + 1)^2 - (\kappa - 1)^2 e^{-2\sqrt{\frac{\epsilon_{\parallel}}{\epsilon_{\perp}}} q_{\parallel} L}. \end{aligned}$$

By analyzing the Coulomb matrix elements, we find that the $\mathbf{k}$ dependence of $V_{\mathbf{k}-\mathbf{k}', \mathbf{k}, \mathbf{k}'}^{\lambda, \lambda'}$ is nearly constant within a valley, i.e., in the regions around K and Γ in which the transition energy varies parabolically, see Fig. 1(b), but has different values in the two valleys. This allows us to reduce the $\mathbf{k}$ and $\mathbf{k}'$ dependencies of the Coulomb matrix elements, which would require huge computer memory, by approximating $V_{\mathbf{k}-\mathbf{k}', \mathbf{k}, \mathbf{k}'}^{\lambda, \lambda'} \approx V_{\mathbf{q}, \mathbf{k}=K, \mathbf{k}'=K-\mathbf{q}}^{\lambda, \lambda'}$ in most of the Brillouin zone and by $V_{\mathbf{k}-\mathbf{k}', \mathbf{k}, \mathbf{k}'}^{\lambda, \lambda'} \approx V_{\mathbf{q}, \mathbf{k}=\Gamma, \mathbf{k}'=\Gamma-\mathbf{q}}^{\lambda, \lambda'}$ for $|\mathbf{k} - \Gamma| \leq 0.3|\mathbf{b}_1|$, where $|\mathbf{b}_1| = 4\pi/\sqrt{3}a$ is the length of a reciprocal lattice vector and a is the lattice constant. Within this approximation the excitonic features rel-

evant for our analysis are described well and the numerical complexity when solving the SBE is kept within reasonable limits.

APPENDIX B: PERTURBATIVE EXPANSION OF THE SEMICONDUCTOR BLOCH EQUATIONS

For the 2H homobilayer MoS₂ considered here the valleys K and K' are equivalent. The interband transition energy is minimal at these valleys and degenerate bands of opposite spin are from upper and lower layers. Here we consider four bands around the band edge, i.e., the two lowest degenerate conduction and the two highest degenerate valence bands. In this four-band model, the interlayer coupling is rather weak and thus ignored. Throughout this manuscript we consider linearly in-plane polarized optical fields which can be described as a superposition of opposite circularly polarized fields that induce intralayer transitions of equal strength in both valleys. Using the electron-hole picture, the simplified two-band SBE for the dynamical variables $p_{\mathbf{k}} = \langle a_{v\mathbf{k}}^{\dagger} a_{c\mathbf{k}} \rangle$ and $n_{\mathbf{k}} = \langle a_{c\mathbf{k}}^{\dagger} a_{c\mathbf{k}} \rangle = 1 - \langle a_{v\mathbf{k}}^{\dagger} a_{v\mathbf{k}} \rangle$ read [33,77]

$$\begin{aligned} \frac{\partial}{\partial t} p_{\mathbf{k}} &= -\frac{i}{\hbar} \left[\epsilon_{\mathbf{k}}^c - \epsilon_{\mathbf{k}}^v + 2 \sum_{\mathbf{k}'} V_{\mathbf{k}-\mathbf{k}'}^{cc} n_{\mathbf{k}'} - \frac{i\hbar}{T_2} \right] p_{\mathbf{k}} \\ &+ \frac{i}{\hbar} \left(\mathbf{d}_{\mathbf{k}}^{cv} \cdot \mathbf{E}(t) + \sum_{\mathbf{k}'} V_{\mathbf{k}-\mathbf{k}'}^{cv} p_{\mathbf{k}'} \right) (1 - 2n_{\mathbf{k}}) \\ &+ \frac{e}{\hbar} \mathbf{E}(t) \cdot \nabla_{\mathbf{k}} p_{\mathbf{k}}, \end{aligned} \quad (\text{B1a})$$

$$\begin{aligned} \frac{\partial}{\partial t} n_{\mathbf{k}} &= \frac{2}{\hbar} \text{Im} \left[\left(\mathbf{d}_{\mathbf{k}}^{cv} \cdot \mathbf{E}(t) + \sum_{\mathbf{k}'} V_{\mathbf{k}-\mathbf{k}'}^{cv} p_{\mathbf{k}'} \right)^* p_{\mathbf{k}} \right] \\ &+ \frac{e}{\hbar} \mathbf{E}(t) \cdot \nabla_{\mathbf{k}} n_{\mathbf{k}}. \end{aligned} \quad (\text{B1b})$$

The perturbative expansion of the SBE is an effective approach to quantitatively distinguish between inter- and intraband excitation pathways when the excitation intensity is sufficiently weak [88,117]. In the following, we expand the microscopic quantities $X(\mathbf{k}, t) = p(\mathbf{k}, t)$, $n_{e/h}(\mathbf{k}, t)$ in powers of the optical field as $X(\mathbf{k}, t) = \sum_{m=0}^{\infty} X^{(m)}(\mathbf{k}, t)$ with $X^{(m)} \propto E_{\text{opt}}^m$. The initial conditions are $p^{(0)}(\mathbf{k}, t = -\infty) = 0$ and $n^{(0)}(\mathbf{k}, t = -\infty) = 0$. The resulting perturbative set of the two-band SBE reads

$$\begin{aligned} \frac{\partial}{\partial t} p_{\mathbf{k}}^{(1)} &= \left[-\frac{i}{\hbar} (\epsilon_{\mathbf{k}}^c - \epsilon_{\mathbf{k}}^v) - \frac{1}{T_2} + \frac{e}{\hbar} \mathbf{E}_{\text{dc}} \cdot \nabla_{\mathbf{k}} \right] p_{\mathbf{k}}^{(1)} \\ &- 2\frac{i}{\hbar} \mathbf{d}(\mathbf{k}) \cdot \mathbf{E}_{\text{dc}} n_{\mathbf{k}}^{(1)} + \frac{i}{\hbar} \mathbf{d}(\mathbf{k}) \cdot \mathbf{E}_{\text{opt}}(t) \\ &+ \frac{i}{\hbar} \sum_{\mathbf{k}'} V_{\mathbf{k}-\mathbf{k}'}^{cv} p_{\mathbf{k}'}^{(1)}, \end{aligned} \quad (\text{B2a})$$

$$\begin{aligned} \frac{\partial}{\partial t} n_{\mathbf{k}}^{(1)} &= \frac{2}{\hbar} \text{Im} [\mathbf{d}^*(\mathbf{k}) \cdot \mathbf{E}_{\text{dc}} p_{\mathbf{k}}^{(1)}] + \frac{e}{\hbar} \mathbf{E}_{\text{dc}} \cdot \nabla_{\mathbf{k}} n_{\mathbf{k}}^{(1)}, \quad (\text{B2b}) \\ \frac{\partial}{\partial t} p_{\mathbf{k}}^{(m)} &= \left[-\frac{i}{\hbar} (\epsilon_{\mathbf{k}}^c - \epsilon_{\mathbf{k}}^v) - \frac{1}{T_2} + \frac{e}{\hbar} \mathbf{E}_{\text{dc}} \cdot \nabla_{\mathbf{k}} \right] p_{\mathbf{k}}^{(m)} \\ &- 2\frac{i}{\hbar} \mathbf{d}(\mathbf{k}) \cdot \mathbf{E}_{\text{dc}} n_{\mathbf{k}}^{(m)} + \frac{e}{\hbar} \mathbf{E}_{\text{opt}}(t) \cdot \nabla_{\mathbf{k}} p_{\mathbf{k}}^{(m-1)} \end{aligned}$$

$$\begin{aligned}
& -2\frac{i}{\hbar}\mathbf{d}(\mathbf{k}) \cdot \mathbf{E}_{\text{opt}}(t)n_k^{(m-1)} + \frac{i}{\hbar} \sum_{k'} V_{k-k'}^{cv} p_{k'}^{(m)} \\
& -2\frac{i}{\hbar} \sum_{j \leq m} p_k^{(j)} \sum_{k'} V_{k-k'}^{cc} n_{k'}^{(m-j)}, \quad (\text{B2c})
\end{aligned}$$

$$\begin{aligned}
\frac{\partial}{\partial t} n_k^{(m)} &= \frac{2}{\hbar} \text{Im}[\mathbf{d}^*(\mathbf{k}) \cdot \mathbf{E}_{\text{dc}} p_k^{(m)}] + \frac{e}{\hbar} \mathbf{E}_{\text{dc}} \cdot \nabla_{\mathbf{k}} n_k^{(m)} \\
&+ \frac{2}{\hbar} \text{Im}[\mathbf{d}^*(\mathbf{k}) \cdot \mathbf{E}_{\text{opt}}(t) p_k^{(m-1)}] + \frac{e}{\hbar} \mathbf{E}_{\text{opt}}(t) \cdot \nabla_{\mathbf{k}} n_k^{(m-1)} \\
&+ \frac{2}{\hbar} \sum_{j \leq m} \text{Im} \left[p_k^{(j)} \sum_{k'} V_{k-k'}^{cv} p_{k'}^{*(m-j)} \right]. \quad (\text{B2d})
\end{aligned}$$

The static electric field is considered in the homogeneous part of the respective equations and thus does not couple different orders since we expand with respect to the optical field. Please note that the static field is switched on smoothly prior to the optical excitation. Furthermore, interband excitations induced by the static field, i.e., the terms $\propto \mathbf{d}(\mathbf{k}) \cdot \mathbf{E}_{\text{dc}}$ in Eqs. (B2a) and (B2d) have a negligible influence on the numerical results that we present.

E_{dc} leads to temporal change of the wave vector $\hbar \dot{\mathbf{k}}(t) = -e\mathbf{E}_{\text{dc}}$ and couples $p_k^{(m)}$ and $n_k^{(m)}$ ($m \geq 1$). $n_k^{(1)}$ is nonvanishing only in the presence of the static electric field due to interband transitions that could be induced by the static field. Since prior to the excitation the polarization and occupations vanish, in first order the interband excitation induced by the optical field initiates the entire dynamics. Higher orders of $p_k^{(m)}$ ($m > 1$) are generated by intraband excitations via the source term proportional to $p_k^{(m-1)}$ and interband excitations which are proportional to $n_k^{(m-1)}$.

APPENDIX C: LINEAR p EQUATION WITH STATIC AND TIME-DEPENDENT DETUNINGS

The main mechanisms underlying of the dynamics of the instantaneous frequency shift displayed in Figs. 6 and 7 can be understood by analyzing a single equation for the linear microscopic polarization,

$$\frac{\partial}{\partial t} p(t) = -i(\omega_{1s} + \delta_{\text{dc}} + \delta_{\text{opt}}(t)) p(t) + \frac{i}{\hbar} d E(t) - \frac{p(t)}{T_2}. \quad (\text{C1})$$

Here $\hbar\omega_{1s}$ is the energy of the 1s exciton without applied fields, $\hbar\delta_{\text{dc}}$ describes the Stark shift to lower energies induced by the static field, and $\hbar\delta_{\text{opt}}(t)$ represents the time-dependent optical Stark shift to higher energies. We solve Eq. (C1) for resonant excitation using effective parameters, which, however, turn out to be quite similar to the shifts that are obtained in the full calculations. For instance, in the linear spectra shown in Fig. 2, the shifts for static fields in the range 0–0.8 MV/cm are roughly 0–20 meV. To obtain similar dynamical features of the linear polarization model as in the THG calculations, the maximal $\hbar\delta_{\text{dc}}$ has to be increased to about 30 meV. The three-photon resonant optical cw fields with amplitudes from 0–8 MV/cm produces blue shifts of up to about 90 meV in Fig. 5(d), whereas we use here a

maximal shift of $\hbar\delta_{\text{opt}}$ of 80 meV. The static shift is mimicked by a constant value for δ_{dc} with negative values (red shift), whereas $\delta_{\text{opt}}(t)$ is time dependent, positive (blue shift), and follows the Gaussian envelope of the excitation pulse $E(t)$, i.e., the maximal shift occurs at $t = 0$. The frequency of the excitation pulse is resonant with ω_{1s} . The envelope of $E(t)$ and the dephasing time T_2 are chosen as in the full calculations.

Although the simple linear model cannot fully incorporate all contributions and dynamics features that determine the SHG and the THG, with the simplified linear Eq. (C1) it is more straightforward to understand the main effects that determine the dynamics of the instantaneous frequency shift. In Fig. 8(a), without a dc field, i.e., for $\delta_{\text{dc}} = 0$, a change of the frequency can only occur when $\delta_{\text{opt}}(t)$ is finite, i.e., during the presence of the optical pulse. Therefore, the shift vanishes before and after the excitation by the optical pulse. For weak $\delta_{\text{opt}}(t)$ we see in Fig. 8(a) that the instantaneous frequency shift follows the envelope of the pulse, i.e., is proportional to $\delta_{\text{opt}}(t)$. When the transient detuning $\delta_{\text{opt}}(t)$ is large enough a single or even multiple off-resonant Rabi-type oscillation of the polarization occurs during the pulsed excitation. This explains the increasingly oscillatory response shown in Fig. 8(a) with increasing $\delta_{\text{opt}}(t)$.

In a second scenario, we consider different dc fields, i.e., $\hbar\delta_{\text{dc}} < 0$ and weak optical field, i.e., a small $\delta_{\text{opt}}(t)$. The instantaneous frequency shift starts at zero, i.e., the polarization oscillates initially with the driving frequency, see Fig. 8(b), and $\hbar\omega_{\text{shift}}$ approaches $\hbar\delta_{\text{dc}}$ when the optical driving is over. The linear polarization can be thought of being composed of two parts $p(t) = p_{\text{driven}}(t) + p_{\text{free}}(t)$ which oscillate with the driving frequency and with the eigenfrequency, respectively. At early times we have that $|p_{\text{driven}}(t)| > |p_{\text{free}}(t)|$. This relation will reverse at some t_0 during the pulsed optical excitation and after the excitation $p_{\text{driven}}(t)$ vanishes and only $p_{\text{free}}(t)$ is finite. In addition to this dynamic change from a driven towards a free oscillation, for large-enough δ_{dc} additional oscillations appear in Fig. 8(b). Similarly as in Fig. 8(a), these arise off-resonant Rabi-type oscillations which appear when $\hbar\delta_{\text{dc}}$ exceeds the spectral width of the exciting pulse.

In Fig. 8(c) we consider the same δ_{dc} Fig. 8(b); however, $\hbar\delta_{\text{opt}}(t)$ is increased to a peak blue shift of 80 meV at $t = 0$. Therefore the results are at early times similar to the curve shown for $\hbar\delta_{\text{opt}}(t = 0) = 80$ meV in Fig. 8(a), however, with increasing time when the optical driving decays $\hbar\omega_{\text{shift}}$ approaches the values considered for $\hbar\delta_{\text{dc}}$.

In Fig. 8(d), we show that the linear p equation can also reproduce the kind of divergences, as obtained in Fig. 6(c) for SHG. Here again, the polarization can be understood as the superposition of a contribution at the excitation frequency and a contribution that follows the time-dependent detuned resonance. For all considered cases these two contributions will have the same magnitude at a particular time t_0 , i.e., $|p_{\text{driven}}(t_0)| = |p_{\text{free}}(t_0)|$. Under certain conditions at this time t_0 the phases of these two contributions will be exactly opposite, i.e., vary by π , such that they interfere destructively leading to a vanishing polarization, i.e., $p(t_0) = 0$. At such a zero crossing, the phase of p becomes ill defined which appears as jumps in plots of the instantaneous frequency.

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